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ASSESSMENT OF THE PRESENT STATE OF POLLUTION BY CADMIUM, COPPER, ZINC AND LEAD IN THE MEDITERRANEAN SEA

In co-operation with

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1. INTRODUCTION

During the MED POL II pilot project (Baseline studies and monitoring of metals, particularly mercury and cadmium, in marine organisms) which was jointly co-ordinated by FAO and UNEP, a considerable amount of data on mercury, cadmium as well as on other heavy metals in marine organisms was compiled.

This assessment of pollution by Cd, Cu, Zn and Pb in the Mediterranean is mainly based on data compiled by the Pilot Project as well as on data presented during the ICSEM/UNEP Workshops or published in other scientific publications.

For these heavy metals there are enough data for an overall but still incomplete picture of levels in Mediterranean waters sediments and marine organisms.

2. SOURCES AND INPUTS OF CADMIUM, COPPER, ZINC AND LEAD IN THE MEDITERRANEAN

Rocks and soils contain different amounts of heavy metals depending on factors such as rock type and mineral composition.

Cadmium concentrations in igneous and metamorphic rocks vary from 0.001 to 1.8 µg/g. In shales and phophorites, cadmium concentrations can reach as high as 90 to 340 µg/g. Typical concentrations of lead in the earth's crust range from 10-70 µg/g in igneous and metamorphic rocks and from 10 to about 100 µg/g in carbonaceous shales and phosphate rocks (GESAMP, 1983).

Copper and zinc are found concentrated in minerals of basaltic rocks.

Sulfide deposits are the principal ores for Cu, Zn and Pb. Cadmium, a relatively rare element, occurs in Zn bearing sulfide ores (Fleischer, 1972).

Geologic weathering and erosion of the earth's crust release and transport heavy metals into the marine environment mainly through rivers and surface runoff. Other natural sources include deep sea volcanism and the atmosphere.

Anthropogenic activities such as agriculture, mining, industrial processing of ores and metals as well as the use of metals and metal components have resulted in increased inputs of heavy metals into the oceans. Other sources including combustion of fossil fuels, smelting and the use of lead gasoline release heavy metals into the atmosphere which are subsequently transported to the oceans (Förstner and Wittmann, 1983).

Cadmium is produced as a by-product of Cu refining and Pb processing. It can be carried from contaminated agricultural soils, mining wastes and mining waters by surface runoff into the sea. Appreciable amounts of Cd are released into the environment by Cu, Pb, Ni and Zn smelters, through the combustion of fossil fuels, the disposal of sewage sludges and by industrial processes. Cadmium is used in the electroplating industry, for the manufacture of pigments, alloys and solders, in alkaline batteries and as a polymerizing catalyst in PCB production (GESAMP, 1983).

Copper is mined in the form of chalcopyrite and is used in various industrial processes such as the electrical industry, in alloys, as a chemical catalyst, in antifouling paints as an algicide and wood preservative (UNEP/WHO, 1982).

Sources of Pb include mining and smelting of ores such as galena, cerusite and anglesite and industrial processes involving the manufacture of batteries, pigments, alloys, and tetraetylleads. Lead is released into the atmosphere by steel production, the combustion of fossil fuels and by the use of leaded gasoline (UNEP/WHO, 1982; GESAMP,1983).

Zinc is produced by smelting of sphalerite ores and is used for the manufacture of alloys, paints and batteries. Other industrial sources include pulp and paper, organic chemicals, fertilizers, petroleum refining, basic steel works and non ferrous metals works (UNEP/WHO, 1982; Förstner and Wittmann, 1983).

Heavy metal sources into the marine environment include surface runoff, domestic and industrial effluents disposed through outfalls, dumping of sewage and industrial sludges and the atmosphere. Metals are appreciably enriched in the suspended load of wastewaters and in sewage sludges as a result of industrial wastes, corrosion within the urban water supply network and urban runoff transporting metals such as Pb and Zn from the surface of streets and highways (Zafiropoulos, 1976; Förstner and Wittmann,1983).

Rivers transport metals that are dumped by industries or urban centres or are the result of increased erosion due to mining or agriculture. The atmosphere over urban and industrial centres can be enriched in Cd, Cu, Pb and Zn, when compared to average crustal material by as much as four orders of magnitude (Förstner and Wittmann, 1983). Rainfall or dry deposition transport these metals in the marine environment.

Inputs of Pb and Zn into the Mediterranean from land-based sources have been assessed by the joint ECE/UNIDO/FAO/UNESCO/WHO/IAEA/UNEP Project (MED POL X) (UNEP/ECE/UNIDO/FAO/UNESCO/WHO/IAEA,1984).

This project involved the assessment of heavy metal inputs from domestic sewage, industrial wastewater and river waters:

a. Domestic sewage. The assessment was based on data such as resident population, tourism and industry. Direct data on heavy metal concentrations and domestic sewage flows were available in only some cases.

b. Industrial waste water. For the assessment of heavy metal inputs from industrial waste water a flexible approach was used. Where direct information on analytical results of heavy metal concentrations in industrial effluents was available it was used with wastewater flows to calculate inputs. When no direct data were available indirect information such as production figures, water consumption and number of employees was used. In collecting information on heavy metal, but also other pollutant loads in industrial effluents the following problems were encountered:

i) Information on the exact location of industrial plants relative to the coastline was often lacking;

ii) Industries were not consistently classified;

iii) Production and employee figures were insufficiently differentiated;

iv) Experience on trace contaminants in industrial wastewaters was scarce;

v) Reporting systems from country to country were not comparable.

c. River discharges. Rivers were very rarely monitored for heavy metal concentrations. Sample pretreatment and analytical techniques used, varied widely from country to country making comparisons difficult.

Average pollutant concentrations and mean water discharges were available for 30 out of a total of 68 Mediterranean rivers. For the remaining 38 heavy metal input estimation was based on extrapolation.

Atmospheric inputs, which for the case of metals such as Pb are very important were not considered at all by this project. For Cd the lack of data made the estimation of total inputs impossible. The results of this project are summarized in Tables 1 and 2. Rivers are the most important sources of Pb and Zn in the Mediterranean carrying 3200 and 18000 tons per year respectively. About 79% of the total Pb inputs are due to pollution. For Zn the respective percentage is 84%.

Table 1. Estimated loads of lead and zinc in the Mediterranean from land-based sources in tons/year

	Pb	Zn
Pollution loads originating in the coastal zone	1600	6900
- Domestic	200	1900
- Industrial	1400	5000
Loads carried by rivers into the Mediterranean	3200 (2700-3800)	18000 (14000-22000)
- Pollution	2200	14000
- Background	1000	4000
Total Mediterranean loads	4800 (4300-5400)	25000 (21000-29000)
- Pollution	3800	21000

Table 2. Estimated loads of lead and zinc in the Mediterranean regions (% of total in brackets)

Region	tons/year (%) Pb	Zn
I	90 (2)	300 (1)
II	1360 (28)	5200 (21)
III	120 (2)	700 (3)
IV	630 (13)	3000 (12)
V	1440 (30)	8600 (35)
VI	230 (5)	1600 (6)
VII	100 (2)	500 (2)
VIII	440 (9)	2500 (10)
IX	180 (4)	1100 (4)
X	230 (5)	1200 (5)
Total	4820	24700

From UNEP/ECE/UNIDO/FAO/UNESCO/WHO/IAEA, 1984

High inputs enter the Mediterranean into the Adriatic (region V) and the northwestern basin (region II) (Fig. 1), which are bordered by industrialized countries and receive major river discharges. Regions IV and VIII (Tyrrhenian and Aegean Seas) receive moderate amounts of these metals (14% of total inputs). The other Mediterranean regions (I, III, VI, VII, IX and X) receive less than 9% of total loads.

The results of similar studies for the estimation of heavy metal inputs in other marine areas, even if not necessarily relevant for the Mediterranean, can provide useful information.

In southern California, an area receiving only limited surface runoff, it was found that the most important sources of Cu and Zn were the atmosphere and municipal wastewater. For Pb the most important source was found to be the atmopshere (Young et al., 1973).

In marine areas receiving large amounts of surface runoff like Puget Sound in N. Western United States, the most important sources for Zn and Cu were rivers. In contrast atmospheric inputs of Pb were 50% higher than river inputs (Zafiropoulos, 1976).

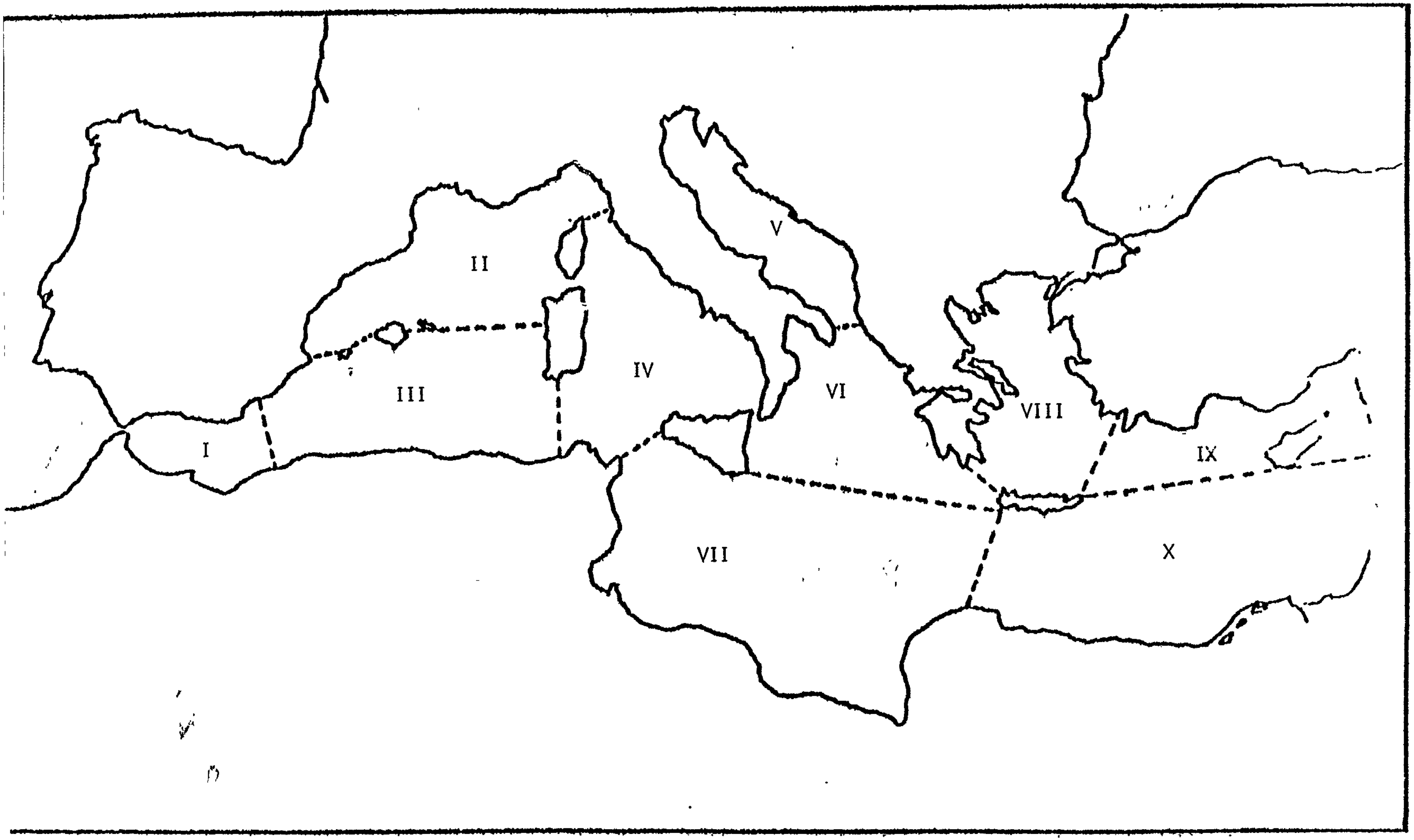

Figure 1. Mediterranean Regions

Work done by Chesselet et al. (1979) and Buat-Menard et al. (1980) has indicated that heavy metal concentrations in suspended particulates in the open Mediterranean waters could not be the result only from crustal weathering and particles of planktonic composition. Atmospheric inputs were indicated as a significant source.

Palumbo and Iannibelli (in press) have recently reported that Cd, Cu, Pb and Zn atmospheric inputs into the Bay of Naples are considerable.

Arnold et al. (1983) have estimated atmospheric inputs of heavy metals in the western Mediterranean. In Table 3 inputs in the western Mediterranean are extrapolated for the whole Mediterranean and compared with total land based inputs as estimated by Project MED POL X.

Atmospheric inputs of Pb (40000 tons/year) are 8 times higher than the combined industrial, domestic and riverine inputs. Zinc atmospheric inputs (27500 tons/year) are comparable with land based inputs estimated by MED POL X. It should be emphasized, though, that this extrapolation could be of dubious value. Chester et al. (1981) have reported a gradient of decreasing Pb atmospheric concentrations from the Western to the Eastern Mediterranean. Particulate Pb fluxes to the Mediterranean have been estimated by Chester et al. (1983) to be between 1600 and 6500 tons/year and are considerably lower than the flux of 40,000 tons/year estimated by extrapolation from Arnold et al. (1983) data.

Table 3. Comparison of atmospheric inputs and land based inputs of heavy metals into the Western Mediterranean.

	tons/year		
	Western Med.^{x}	Total Med.*	Total land-based source inputs$^+$
Pb	16000	40000	4820
Zn	11000	27500	25000
Cu	1000	2500	-
Cd	140	350	-

^{x}Arnold et al. (1983) Area of West Med. 10^6 km^2
*Total Med. area = 2.5×10^6 km^2
$^+$ UNEP/ECE/UNIDO/FAO/UNESCO/WHO/IAEA, (1984)

The exchange of waters through the Straits of Gibraltar can also be a heavy metal source for the Mediterranean. This has been recently reported by Boyle et al. (in press) for Cu and Cd. According to these authors a significant portion of heavy metal enrichment of the Mediterranean Sea may originate in coastal waters outside the basin.

3. FATE OF HEAVY METALS

Heavy metals entering the marine environment through different sources can undergo physico-chemical changes such as dilution/dispersion, precipitation, adsorption or desorption, absorption and release by marine organisms, removal to the sediments and resolubilization.

Where rivers meet the sea the increase of salinity and the sedimentation of river particulates can result in removal to the sediments or mobilization from particulates to the dissolved phase of heavy metals.

In the Rhine river estuary Cu, Pb and Zn are mobilized from the suspended particulates to the water column (Bryan, 1976).

However this is not always true for other estuaries. Oregioni et al. (1979) report that more than 90% of Zn and Pb are carried as suspended matter in the Var and Rhone rivers and can be considered to be deposited rapidly in their estuaries. Cauwet and Faguet (1981) and Buscail and Cauwet (in press) report that river mouths serve as traps for heavy metals that are deposited to the sediments. Kitano and Sakata (1978) report that the sedimentation in the estuarine areas removes most of Cu and Zn discharged by surface runoff.

A possible mechanism is that heavy metals deposit along with Fe rich flocs in the stream-sea boundary (Turekian, 1977). Zutic et al. (in press) report that a considerable fraction of dissolved trace constituents, including heavy metals, will bind to organic aggregates which are produced by flocculation of riverine organic matter and in situ primary production where river and sea-water mix.

Although sedimentation seems to be a major process responsible in removing heavy metals to the sediments in the estuarine area, some mobilization should also be expected to take place. Apart from Fe and Mn information is insufficient to determine a systematic behaviour of heavy metals in estuarine areas (Bewers and Yeats, 1981).

Heltz et al. (1975) have studied the behaviour of heavy metals including Cd, Cu, Pb and Zn discharged from a wastewater treatment plant into Chesapeake Bay in USA. They suggested that these metals are removed to the sediments. For Cd they report a possible remobilization from the sediments to the water column. Morel et al. (1975) have studied the fates of heavy metals in the Los Angeles wastewater discharge. Their data indicate that heavy metals can be mobilized from particulates but these will deposit before such solubilization can take place.

In the open waters adsorption of heavy metals on the hydrous oxide surface of iron and manganese oxides and clay minerals seems to be the most important ultimate removal mechanism (Yuan-Hui Li, 1981a, 1981b). The same author reports that the settling of faecal pellets is the predominant transport mechanism of heavy metals from surface to deep waters.

Fowler (1977) in studying the concentrations of heavy metals in zooplankton particulate products in the Mediterranean has concluded that the sinking of moults and faecal pellets of zooplankton contain relatively high concentrations of heavy metals and may play a very important role in marine biogeochemical cycles.

Lead particulates entering the sea through the atmosphere, due to the poor solubility of Pb compounds, are removed to the sediments. Lead pollutants in the coastal sediments of southern California are derived mainly from the combustion of lead additives in gasoline as Chow et al. (1973) have shown.

Heavy metals, in solution or in particulate form can be accumulated by marine organisms. Macroalgae absorb the soluble heavy metal forms, bivalves which are filter feeders can obtain heavy metals not only from food and from solution but also from the ingestion of inorganic particulate material. Fish can accumulate heavy metals both from food and solution. Metal uptake from solution can occur through the gill or the gastro-intestinal wall (Phillips, 1977).

Cadmium uptake by marine organisms is believed to occur both through the gills and through food. However there is little evidence of food chain magnification. The food chain is considered to be the main source of Zn in tissues of marine organisms but as for Cd there is no evidence of magnification (IRPTC, 1978).

4. CONCENTRATIONS OF CADMIUM, COPPER, ZINC AND LEAD IN THE MEDITERRANEAN

4.1 Sea-water

Heavy metal analysis in sea-water can be subject to serious errors arising from sampling, preconcentration and determination steps. The improvement of techniques and the control of contamination of samples with the use of clean laboratories and ultra-clean reagents has led to a decrease in the concentrations of heavy metals reported in the literature (Bruland et al., 1978a, 1978b) (see also Table 4).

Prior to 1975 relatively few conclusions could be drawn concerning the biogeochemistry and spatial distributions of heavy metals. This is believed to be a result of contamination during sampling and analysis. Research after 1975 has lead to the conclusion that heavy metals such as Cd, Cu and Zn are found in oceanic waters in concentrations that are significantly lower than previously thought. It was also found that these metals appear in well defined distributions in the world oceans (Bruland and Franks, 1979).

Heavy metal concentrations, especially in coastal waters, can depend on factors such as input variability, mixing of different water masses, transport and dilution processes and biological activity. Thus, the interpretation and comparison of heavy metal concentrations in sea-water is rather difficult. Furthermore, analytical methods usually determine different parts of the total heavy metal concentration. In comparing heavy metal data, the values for total, dissolved and particulate forms have to be considered.

Data prior to 1979 have been reviewed by UNEP (1978). In Table 4 more recent data on concentrations of Cd, Cu, Pb and Zn in Mediterranean open waters are summarized. The analytical methods used by authors are also included since they can influence results.

Table 4. Cadmium, Copper, Lead and Zinc concentrations in open waters of the Mediterranean (µg/l)

REGION	METHOD	Cd	Cu	Pb	Zn	REFERENCE
II	ASV	0.15	0.4	-	2.7	Huynh-Ngoc and Fukai, 1979
	ASV	0.11	0.10	-	1.2	Huynh-Ngoc and Fukai, 1979
IV	ASV	0.11	0.18	-	0.9	"
	ASV	0.05-0.09	0.13-0.19	0.018-0.09	-	Nürnberg, 1977
VI-VII	ASV	0.15	0.7	-	1.8	Huynh-Ngoc and Fukai, 1979
VIII	ASV	0.07	0.3	-	3	"
X	ASV	0.04	0.04	-	0.9	"
Recent data						
I-II		0.004	0.11	-	-	Boyle *et al.*, in press
IV-VI-VII		0.010	0.15	-	-	"
II	Dowex/ Extraction/ AAS	0.06	0.48	-	-	Frache *et al.*, 1980, Baffi *et al.*, 1983 a,b,c
II		0.008	-	0.05-0.14	-	Copin-Montegut *et al.*, (in press)
III	ASV	0.005-0.010	0.06-0.13	0.025-0.075	-	Laumond *et al.*, 1983
Mediterranean	Freon extraction AAS or ASVO	0.017±0.007	0.21±0.07	-	0.40±0.16	Kremling and Petersen, 1981
Oceans	Extraction or Chelex/ 100 AAS	0.07-0.7	0.3-2.8	-	1-13	Brewer, 1975
Background Oceanic values (Recent data)		0.01-0.07 0.01-0.1	0.1-0.3	0.005-0.015	0.01-0.6	Förstner and Wittmann, 1983 ICES, 1980

Cadmium

Older data on Cd concentrations in the open Mediterranean waters suggest a range from less than 0.05 up to 0.60 µg/l (UNEP, 1978).

Huynh-Ngoc and Fukai (1979) have reported mean concentrations of dissolved Cd in different Mediterranean regions that range from 0.04 to 0.15 ug/l and a Mediterranean open water average of 0.13±0.02 ug/l. Laumond *et al.* (1983) however, have reported much lower values for the Western Mediterranean (0.005 -0.10 µg/l). In the Tyrrhenian Sea, according to Nürnberg (1977), Cd concentrations range from 0.05-0.09 µg/l.

Kremling and Petersen (1981) report a Mediterranean open water average of 0.017±0.007 µg/l for Cd. According to the authors these values are close to oceanic results gathered under similar conditions.

In a more recent study (Boyle *et al.*, 1984), Cd concentrations in the sea of Alboran were reported to be 0.004 µg/l in surface waters with a maximum of 0.012 µg/l at 500 m depth. In the Central Mediterranean, mean Cd concentration in surface waters was 0.010 µg/l. The authors report that Cd concentrations in the Mediterranean are slightly elevated compared with the open Atlantic. Copin-Montegut *et al.* (in press) report an average concentration of 0.008 µg/l in surface waters of the Western Mediterranean and of 0.001 µg/l in the Atlantic.

ICES (1980) has reported a range of 0.001 to 0.10 µg/l in oceanic waters. These values are very close to Mediterranean concentrations.

Cadmium concentrations in Mediterranean coastal waters are reported to be as high as 1.4 µg/l, considerably increased compared with recent open Mediterranean values of 0.004-0.017 µg/l (Table 5).

The question that arises for Cd, and for the other heavy metals, is whether this considerable increase is a real one or a result of contamination during sampling and analysis. This question can be partially answered by taking into account data from papers that are reporting minimum concentrations close to typical open water values of 0.005+0.01 µg/l. Thus certain coastal areas of Spain and Italy undoubtely have increased Cd concentrations (see Table 5).

In the coastal Lagoon of Mar Menor in Spain which is influenced by mining activities De Leon *et al.* (1983) report Cd concentrations of 0.04 to 0.09 µg/l. Breder *et al.* (1981) have found small increases in Cd concentrations moving towards the mouth of several Italian estuaries. Background concentrations of 0.004 to 0.008 µg/l increased to 0.016-0.029 µg/l inside the estuaries.

Fukai and Huyhn-Ngoc (1976) studying the concentrations of Cd, Cu and Zn in coastal and offshore waters in area II did not observe any significant differences except in areas with high anthropogenic inputs.

In coastal and offshore waters of the Ligurian sea no systematic differences in Cd concentrations have been found although some stations close to input sources showed considerably high concentrations (Frache *et al.*, 1980; Baffi *et al.*, 1983 a, b, c, and 1984).

Table 5. Cadmium, Copper, [illegible]d and Zinc concentrations in Mediterra[illegible] coastal waters (μg/l)

REGION	METHOD	Cd	Cu	Pb	Zn	REFERENCE
II						
-Coastal waters	ASV	0.01-0.8	0.1-22.4	-	1-9	Fukai and Huynh-Ngoc, 1976
-Ligurian Sea	Dowex A-1/AAS	0.03	1.4	-	-	Frache et al., 1980
-Var lagoon, France	APDC extraction/ AAS	0.9	4.1	21	-	Chabert and Vicente, 1981
-Italian estuaries	Filtration/ASV	0.004-0.029	0.11-0.95	0.025-0.95		Breder et al., 1981
-Lagoons, Spain	Freon Tf extraction/AAS	0.040-0.09	-	0.11-0.58	3.3-4.3	De Leon et al., 1983
-Ligurian coasts, Italy	Filtration/Dowex A-1/AAS					
	Dissolved	< 0.002-1.4	<0.010-7.4	-	-	Baffi et al., 1983, 1984
	Particulate	< 0.06	<0.05-0.58	-	-	"
-Monaco	APDC/Ext./AAS		0.4-0.8	-	1-2	Veglia and Vaissiere, in press
IV						
-River Tiber mouth	AAS	0.1-0.6	0.2-0.6	0.1-0.6	1.4-3.3	Pettine et al.. 1982
V						
-Adriatic Sea	NAA	-	3.4	4.7	-	Marijanovic et al., 1983
	NAA	-	0.6-50	-	1-36	Grancini et al., 1976
VI						
-Sicilian coasts	Dissolved	0.01-0.47	0.25-7	0.02-3.7	0.2-16	Alpha et al., 1982
	Particulate	0.02-0.13	0.12-4	0.13-3.5	1-17	
VIII						
-Evoikos, Gera Gulf, Greece	Filtration/Chelex 100/AAS					
	Dissolved	-	1.7-1.9	2.3-2.9	4-13	Scoullos and Dasenakis, 1983;
	Particulate	-	1-1.2	1.2	1.6-2.7	Scoullos et al., 1983
-Elefsis Bay, Greece	Dissolved	-	-	-	18	Scoullos, 1981
	Particulate	-	-		2.4	"
-Saronikos Gulf,	NAA/Total	-	0.5-1.4	-	2.4-32	Zafiropoulos, 1983
Greece	ASV	0.15-0.70	1.0-3.5	-	3.7-18	Huyuh-Ngoc and Zafiropoulos, 1981
-Northern Greece	APDC-MIBK	0.16-0.52	0.7-2.1	3.5-20.5	13-23	Fytianos and Vasilikiotis, 1983
-Izmir Bay, Turkey	Extraction/AAS	0.01-0.03	-	0.05-0.20	-	Gücer and Yaramaz, 1980
X						
-Alexandria, Egypt	APDC- MIBK Extraction/AAS		70		210	El Sayed and El Sayed, 1981
-Mediterranean coastal waters		-	0.02-0.49	0.016-4.4	0.016-11	Reviewed by Bernard, 1983

Copper

Ranges of Cu concentrations in the open waters of the Mediterranean as reviewed by UNEP (1978) were between less than 0.03 and 3 µg/l.

Average dissolved Cu concentration in Mediterranean open waters has been reported by Huynh-Ngoc and Fukai (1979) to be 0.33±0.09 µg/l. Nürnberg (1977) has reported lower values (0.13-0.19 µg/l) for the Tyrrhenian Sea, and Laumond et al. (1983) a range of 0.06-0.13 µg/l for the western Mediterranean.

Kremling and Petersen (1981) report an average of 0.21±0.07 µg/l for the Mediterranean open waters which is very close to oceanic values obtained under similar conditions.

Recent data (Boyle et al., in press) suggest that Cu concentrations in the Mediterranean are actually lower (mean values of 0.11 and 0.15 µg/l for the Alboran Sea and the Central Mediterranean respectively). According to these authors these values are slightly higher than those for the Atlantic.

Maximum values for Cu concentrations reported in different coastal areas range from 0.6-50 µg/l and are considerably increased compared to open water concentrations. In the coastal area of Alexandria concentrations as high as 70 µg/l have been reported (El Sayed and El Sayed, 1981).

In the coastal waters near Cadiz, Spain, Cu concentrations of up to 8.6 µg/l have been reported. These are believed to be the result of copper mining activities in the area (UNEP, 1978).

Lead

Few data on concentrations of Pb in the open Mediterranean waters have been reported. These vary between 0.018 and 0.14 ug/l (Laumond et al., 1983; Nürnberg, 1977; Copin-Montegut et al., (in press). Oceanic values range from 0.005 to 0.015 ug/l (Förstner and Wittmann, 1983) (Table 5). It seems that Pb occurs in relatively increased concentrations in Mediterranean waters. However, this is a rather tentative conclusion in view of the scarcity of Mediterranean data for Pb and the notorious difficulties in accurate Pb analysis.

In coastal areas Pb concentrations of up to 21 µg/l have been reported (see Table 5). Breder et al. (1981) report Pb concentrations that increase from 0.025 µg/l, a value close to open Mediterranean values, to 0.95 µg/l in Italian estuaries. Alpha et al. (1982) report a range of 0.02 to 3.7 µg/l along the Sicilian coasts. Very high concentrations of Pb have been reported (3.5-21 µg/l) in Thermaikos Gulf, Greece, where a tetraethyl lead industry is operating (Vasilikiotis et al., 1982; Fytianos and Vasilikiotis, 1983).

Zinc

Ranges of Zn concentrations in Mediterranean open waters as reviewed by UNEP (1978) were between 0.1 and 86 µg/l.

According to Huynh-Ngoc and Fukai (1979) the average dissolved Zn concentration in the Mediterranean is 2±0.2 µg/l.

Kremling and Petersen (1981) report a Mediterranean average of 0.40±0.16 µg/l and that these concentrations are close to oceanic values.

Zinc concentrations in coastal waters of up to 210 µg/l have been reported and are considerably increased compared to open Mediterranean concentrations (Table 5).

4.2 Sediments

Concentrations of heavy metals in Mediterranean coastal sediments have been studied extensively over the past 6 years. In contrast, very few data exist for the open Mediterranean sediments.

Sedimentation processes will deposit heavy metals, along with terrigenous and biogenic material on the sea bottom. These sedimentation processes are very important in river deltas but also in areas receiving domestic sewage as well as industrial effluents and solid wastes. The concentrations of heavy metals in sediments will thus depend not only on pollution inputs but also on factors such as organic carbon content, mineralogical characteristics, grain size, and sedimentation rates.

The analytical determination of heavy metals in sediments involves as a first step the solubilization of the sample. A wide variety of reagents, usually acids, is used by different investigators ranging from total solubilization by $HF-HClO_4-HNO_3$ to simple extraction by dilute HCl. Some investigators analyze the whole sediment sample, others the fraction of less than 200, 63 or 5.5 µm. It is obvious that due to these factors data on heavy metals in sediments are not easily comparable.

Distribution of heavy metal concentrations determined on the whole sediment may be misleading in identifying areas contaminated by industrial or urban activities. Donazzolo et al. (1984a and b) studying heavy metal concentrations in North Adriatic sediments found that these depend on sediment fine fraction composition, specific surface area and accumulation level in the less than 63 um fraction (pelite). They report that 74-86% of the total concentration of Cd, Cu, Pb and Zn is bound in the pelite fraction.

The relation of heavy metal concentrations in sediments and minerological characteristics or grain size was recognized also in earlier work in the Mediterranean (Grancini et al., 1976, Fascardi et al., 1984).

Cosma et al. (1979, 1982, 1983) have compared different extraction methods for sediment analysis. Reagents such as EDTA, NH_2OH $HCl-CH_3COOH$ and 0.5N HCl extract amounts of Cd, Cu and Pb ranging from 10 to 48% of the total.

Another difficulty in comparing heavy metal concentration in sediments is defining what the natural background values are for a certain area. These natural background values will depend on factors such as grain size, organic carbon content and mineralogical characteristics.

Cadmium

Early publications have reported Cd concentrations in Mediterranean marine sediments to range between 0.1 and 2.3 µg/g (UNEP, 1978). Data reported since 1978 are summarized in Table 6. Minimum concentrations reported range from 0.1 to 10 µg/g. Donazzolo et al. (1984a) report a probable background Cd concentration, as calculated from core samples, of 1.2 µg/g. Frigniani and Giordani (1983) report concentrations of 0.5-2.5 µg/g in offshore sediments whereas Voutsinou-Taliadouri (1983) a value of 0.4 µg/g for Aegean sea sediments. A probable background Cd concentration should be in the range of 0.1 to 2.5 µg/g

Close to source inputs, industrial or urban, Cd concentrations have been reported to range, from 0.3 to 10 µg/g. Very high concentrations (32-64 µg/g) have been reported in sediments of Spanish lagoons (De Leon et al., 1983) in Izmir Bay, Turkey (Uysal and Tuncer, in press) and in the harbour of Alexandria (Saad et al., 1981).

Copper

Relatively few data have been reported on Cu concentrations in offshore sediments. Frignani and Giordani (1983) reported a range of 10-44 µg/g and Voutsinou-Taliadouri (1983) an average of 20±7 µg/g. Core sample analysis has resulted in values ranging from 15 to 30 µg/g (Donazzolo, 1984a, Cauwet and Monaco, 1983). Shaw and Bush (1978) report average values of Cu in deep sea sediments of the Cilician basin of 42 µg/g.Background Cu concentrations in Mediterranean sediments should thus be in the range of 10-44 µg/g with a probable average of 15 µg/g.

Copper concentrations in coastal sediments (table 6) are considerably increased over these background values. High concentrations have been reported in spanish coastal lagoons, in the river Rhône Delta, close to Cannes in the Ligurian sea, in the Gulf of Trieste and Izmir Bay. In the western harbour of Alexandria (Saad et al., 1981) a value of 1890 µg/g has been reported.

Lead

Concentrations of Pb in sediment core samples range from 12 to 32 µg/g (Oregioni, 1980). Donazzolo et al. (1984a) have reported background values for Pb, derived from core samples to be around 23 µg/g. Offshore sediments have Pb concentrations ranging from 15 to 94 µg/g (Frigniani and Giordani, 1983, Voutsinou-Taliadouri, 1983).

In coastal sediments relatively high Pb concentrations have been reported (100-3300 µg/g). These high concentrations occur near source inputs in Spain, Marseille, Cagliari, Gulf of Trieste, and Thermaikos Gulf (table 6).

Zinc

Offshore sediments have Zn concentrations that range from 20 to 78 µg/g (Frigniani and Giordani, 1983; Voutsinou-Taliadouri, 1983). In sediment core samples concentrations range from 50 to 85 µg/g (Donazzolo et al., 1984a; Cauwet and Monaco, 1983). Average Zn concentrations in deep sea sediments from the Cilician basin have been reported by Shaw and Bush (1978) to be around 117 µg/g. Zinc concentrations in coastal sediments are considerably increased over these background values. In the coast of Spain concentrations of up to 6200 ug/g have been reported (De Leon et al., 1983) in the area of Marseille 2550 ug/g (Arnoux et al., 1983); in Kastela Bay, Yugoslavia

1300 µg/g (Stegnar et al., 1979) in Saronicos Gulf, Greece 1360 µg/g (Angelidis et al., 1983) and in Izmir Bay, Turkey 860 µg/g (Uysal and Tuncer, in press). In other coastal areas maximum concentrations range from 100-650 µg/g (Table 6).

It is evident that Cd, Cu, Pb and Zn concentrations in coastal sediments in areas receiving industrial effluents, solid wastes and domestic sewage as well as in river deltas and estuaries are considerably increased over Mediterranean background values. The concentrations reported by researchers depend not only on the actual degree of heavy metal pollution in the area but also on the extraction technique used, as well as on the proximity of stations to source inputs. Evidently in some cases the very high values reported are not representative of the whole areas studied.

4.3. Marine biota

The knowledge of the exact concentrations of heavy metals in seawater and sediments does not necessarily lead to the prediction of the amounts of metals that marine organisms will uptake. Thus we cannot predict whether a marine ecosystem will be adversely affected or whether the consumers' health will be at risk by the consumption of marine organisms on the basis of data on concentrations in water and sediments only.

Marine organisms can uptake to a different degree different physico-chemical forms of heavy metals. Algae are probably uptaking the soluble heavy metal content of seawater. Bivalve molluscs on the other hand are filter-feeders and thus are uptaking heavy metals from food and inorganic particulate ingestion or directly from seawater. This makes bivalves advantageous indicator organisms for the assessment of heavy metal pollution. Many variables can affect heavy metal concentrations in bivalves including age, weight, size, sex, season of sampling, physical factors such as temperature and salinity but it is still not very clear how these variables exactly affect heavy metal concentrations (Phillips, 1977).

Fowler and Oregioni (1976) studied the variation of heavy metal and concentrations in Mytilus galloprovincialis reported maximum concentrations in spring samples. They suggest that this is probably a result of the reproductive state of the mussels but also because of high particulate metal loads caused by increased winter runoff.

Fish assimilate metals from both food and seawater. The uptake can occur across the body surface and particularly across the gills or across the gastro-intestinal wall. Heavy metal uptake from food is probably the most important assimilation route (Phillips, 1977).

Concentrations of heavy metals in fish muscle appear to vary with age, size or weight of the fish with its feeding habits, season and other factors. With the exception of Hg, fish do not always reflect ambient heavy metal concentrations and are thus considered poor indicators. However, it is important to monitor heavy metal concentrations in fish muscle in order to assess possible adverse effects in human consumers.

Variations of heavy metal concentrations with size, season or tissue analysed have been reported for Mediterranean marine organisms. Majori et al. (1979) report significant variations of Cd, Cu, Pb and Zn concentrations in Mytilus galloprovincialis that sometimes correlate with size. Hornung and Oren (1981) report an inverse relationship between Cd, Cu, Pb and Zn concentrations in Donax trunculus and size.

Table 6. Cadmium, Copper, Lead and Zinc concentrations in Mediterranean sediments (µg/g dry weight)

REGION	METHOD	Cd	Cu	Pb	Zn	REFERENCE
II						
-Var lagoon, France	$HF-HClO_4-HNO_3$	3.7	15.4	26.4	-	Chabert and Vicente, 1981
-Coastal lagoon,	$<63\mu m$	10-32	10-94	200-2000	500-6200	Leon et al., 1983
Spain	Conc.HNO_3	0.1-0.3	-	10-50	-	Peiro et al., 1983
-River Ebro Delta	HNO_3	0.12-0.37	7.9-21.5	22-48	33-104	Obiols et Peiro, 1981
-River Rhône Delta	HNO_3-HClO_4	0.25-5	20-55	9	90-140	Added et al., 1981; Cauwet and Monaco, 1983; Badie et al., 1983
-Marseille	$<200\mu m$ $HCl-HNO_3$	1.8-3	29-34	28-1250	120-2550	Arnoux et al., 1981
-Cannes	$<63\mu m$ $HNO_3-H_3PO_4-HCl$	1.8-7	15-80	30-100	50-300	Ringot, 1983
-Gulf of Nice	HNO_3-HCl	$<0.7-2.4$	$<2.1-32$	3-112	-	Flatau et al., 1983
-Italian Estuaries	HNO_3-HCl	0.21-0.55	33-53	30-43	-	Breder et al., 1981
-Ligurian Sea	$HF+HClO_4$	-	14-145	30-250	60-970	Cosma et al. 1979, 1982, 1983
-Offshore sediments	HNO_3-HCl	-	30-49	10-28	130-260	Arnoux et al., 1983
	HNO_3	0.7-1.7	-	-	-	Frignani and Giordani, 1983
III						
-Coast of Spain	-	0.02-10	4-230	23-3300	27-1050	De Leon et al., in press
IV						
-Cagliari lagoon	0.4NHCl	-	10-70	64-670	-	Contu et al., in press
-Offshore sediments	HNO_3	0.5-2.5	10-44	19-94	20-56	Frignani and Giordani, 1983
V						
-Gulf of Trieste		0.3-5.3	9-139	18-470	27-650	Majori et al., 1979
-Gulf of Venice	HNO_3	0.1-3.1	34-37	5-54	48-450	Angela et al., 1981
-Kastela Bay Yugoslavia	NAA$<100\mu m$		14-42	-	53-1300	Stegnar et al.,1979
-River Po delta	HNO_3	0.16-1.7	1.3-50	9.-73	24-244	Fascardi et al., 1984
-Mali Ston, Yug.	NAA	0.08-0.22	13-22	-	40-100	Vukadin et al., in press
-Northern Adriatic	-	$<0.05-5.6$	2.3-52	5.3-96	1.7-870	Donazzolo et al, 1984a,1984b
-Offshore sediments	HNO_3	0.80-1.2	15-30	21-43	54-78	Frignani and Giordani, 1983

Table 6. (Cont'd)

REGION	METHOD	Cd	Cu	Pb	Zn	REFERENCE
VI						
-Patraikos Gulf, Greece	HF-HNO$_3$ HClO$_4$	-	23-100	10-40	280-430	Varnavas and Ferentinos, 1983
-Kalamata Bay, Greece	HF-HNO$_3$-HClO$_4$	-	11-56	8-40	-	Varnavas et al., in press
-Gulf of Catania	HNO$_3$	2.2-4.6	3.8-2.5	4.5-19	25-236	Castagna et al., 1982
-Offshore sediments	HNO$_3$	0.6-1.1	24-29	22-27	55-78	Frignani and Giordani, 1983
VIII						
-Thermaikos-Kavala Gulf, Greece	< 63µm HNO$_3$-HClO$_4$	0.6-1.1	0.6-2.3	6-28	10-28	Fytianos and Vasilikiotis, 1983
-Evoikos Gulf,	< 61µm 0.5N HCl	-	9	37	20	Scoullos and Dasenakis, 1983
Greece	< 55µm 0.5N HCl	-	-	-	7-95	Angelidis et al., 1981
-Gera Gulf, Greece	< 61µm 0.5N HCl	-	8-160	9-122	12-390	Scoullos et al., 1983
Saronikos Gulf, Greece	< 55µm 0.5N HCl	-	-	-	5-1360	Angelidis et al., 1983
-Thermaikos Gulf, Greece	< 45µm HNO$_3$	0.40-2.5	10-50	25-130	8-240	Voutsinou-Taliadouri, 1983
-Pagassitikos Gulf, Greece	< 45µm HNO$_3$	-	30	30	130	Voutsinou-Taliadouri, 1983
-East Aegean offshore	< 45µm HNO$_3$	0.4	20	15	40	Voutsinou-Taliadouri, 1983
-Izmir Bay	HCl-HNO$_3$	0.2-49	14-870	20-280	53-860	Uysal and Tuncer, in press
IX						
-Erdemli, Turkey	HNO$_3$-HClO$_4$-HF	-	31	57	65	Balkas et al., 1979
-Alexandria		2.8	48	190	180	El Sokkary, 1979
-Alexandria Harbour	HNO$_3$-HCI	-	27	-	53	El Sayed et al., 1981
-Abu Kir Bay, Egypt	HNO$_3$	2	12	-	100	Saad et al., 1981
-River Nile delta	HF-HNO$_3$	-	5-77	-	2-120	Moussa, 1983
	HCl-1N		6-74		20-100	Tomma et al., 1981
-Cilician basin	HF-HNO$_3$-HClO$_4$	-	33-50	-	54-81	Ozkan et al., 1980
X						
-Damietta estuary Egypt	HNO$_3$	0.16-2	29-280	-	20-425	Saad and Fahmy, in press
-Western Harbor Alexandria	HNO$_3$-HClO$_4$	7-64	30-1890	-	23-470	Saad et al., 1981
XIII						
-Black Sea, Nearshore	HNO$_3$	1.3-4.8	10-100	22-88	37-250	Pecheanu, 1983
Offshore		2.8	52	37	75	Pecheanu, 1983
Mediterranean		0.1-2.3	-	31 9-95	31-2500	UNEP, 1978

Significant seasonal variations of Cd, Cu and Zn in the liver and gonads of Mullus barbatus appear to be related to the sexual physiology of this fish (Lafaurie et al., 1981).

Uysal and Tuncer (1983) found differences of Cd, Cu, Pb and Zn in Mullus barbatus, Mullus surmuletus and Sardina pilchardus according to length and season.

Voutsinou-Taliadouri and Satsmadjis (1982) observed higher Cd, Cu, Pb and Zn concentrations in marine organisms from Saronikos Gulf during the summer period.

Correlation between several heavy metal concentrations have been reported for Mullus barbatus and can provide a useful insight in the biochemistry of these metals (Zafiropoulos, 1981).

Heavy metals can accumulate in different degrees in different parts of marine organisms. Copper concentrations are higher in the liver and digestive track of Mullus barbatus and Engraulis encrasicolus (Capelli et al., 1981). Significant differences in Zn content of dark and white muscle as well as liver of Auris rochei has been reported by Andreotis and Papadopoulou (1981). The same findings have been reported for Sarda sarda (Capelli et al., 1983a).

Another question of considerable importance is whether food-chain magnifications occur for heavy metals in the marine environment. Campesan et al., (1981) found no clear evidence of heavy metal accumulation in a partial food web in the lagoon of Venice. Papadopoulou et al. (1979) found that Zn concentrations decrease with food level in part of a pelagic foodchain of the Aegean Sea. Bernhard and Andreae (1984) report that the concentrations of Cd, Cu and Zn at first increase along the foodchain, reaching a maximum with crustaceans, and then decrease in fish.

During the MED POL II Project (PHASE I) concentrations of mercury, cadmium and other heavy metals such as copper, zinc, manganese, lead, nickel and chromium were determined in Mediterranean marine organisms. Data on mercury concentrations compiled by this Project have already been assessed (UNEP/FAO/WHO,1983)

A considerable amount of data on levels of Cd, Cu, Pb and Zn in Mediterranean marine organisms have been reported in recent years in the literature and mainly during Workshops on Pollution of the Mediterranean. Publications considered but not cited are included in an additional reference list.

The present assessment is mainly based on data on Cd, Cu, Pb and Zn metal concentrations in marine organisms reported by participating institutes. The results of the intercalibration exercise were considered in the statistical treatment of results in order to exclude analytical errors.

It should be pointed out that average concentrations reported should not be interpreted as representative mean values for a given region, or the whole Mediterranean, due to differences between conditions prevailing at the individual sampling stations. Most samples were collected from coastal areas receiving industrial effluents or domestic sewage and are therefore representative of these "polluted" areas. There is a considerable variability of concentrations reported. Standard deviations are sometimes higher than the respective arithmetic means.

Although the computer processed data do not show any significant regional differences of heavy metal concentrations in marine organisms, researchers have reported increased values from areas with high heavy metal inputs.

In the lagoon of Mar Menor, Spain, dumping of industrial wastes has resulted in the accumulation of Pb and Zn in bivalves (De Leon *et al.*, 1983). High concentrations of Pb and Cd as compared to clean areas were found in samples of *Mullus barbatus*, *Mytilus galloprovincialis* and *Merluccius merluccius* from gulfs in Northern Greece receiving industrial and domestic effluents (Vasilikiotis *et al.*, 1983). In the Bay of Izmir, Cu and Pb concentrations are increased in *Mytilus galloprovincialis* collected from polluted areas (Tuncer and Uysal, 1983).

In contrast, other researchers report no significant increase in heavy metal contents of marine organisms in areas receiving industrial effluents or domestic sewage (Roth and Hornung, 1977; Grimanis *et al.*, 1981, 1983; Balkas *et al.*, 1982, Orlando and Mauri, 1983; Capelli *et al.*, 1983b).

In the open waters of the Mediterranean the majority of pelagic organisms analysed had heavy metal concentrations within the range of values that have been reported for the same groups from other geographical areas (Fowler *et al.*, 1979).

Stegnar *et al.*, (1979) report that Zn and Cu concentrations in mesopelagic fish are the same as concentrations found in coastal fish. Stoeppler and Nürnberg (1979) studying heavy metal levels in marine biota from the Mediterranean and other European seas report concentrations of Pb less than 10 µg/kg and of Cd around 3 µg/g.

Cadmium

Average Cd concentrations in *Mullus barbatus* and *Mytilus galloprovincialis* from different Mediterranean regions are summarized in Tables 7 and 8.

In *Mullus barbatus* Cd average regional values range from 17 to 50 µg/kg (fresh weight). In view of the considerable variability of data (standard deviations of more than 35%) there seem to be no significant differences between regional means.

The overall Mediterranean average (335 samples) is 46 µg/kg with a standard deviation of 67. Most data however are below 60 µg/kg.

Table 7. Cadmium concentrations in Mullus barbatus µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	136	1.0	590	50	90
VI	50	5.0	52	26	14
VII	11	5.5	49	17	15
VIII	46	15	162	47	39
X	21	14	65	39	14

Table 8. Cadmium concentrations in Mytilus galloprovincialis µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	105	40	1060	190	120
V	72	25	475	160	100
VI	25	24	52	38	6
VIII	76	5	780	100	124

Excluding 5% of the values which have been considered unreasonably high the overall Mediterranean average is 34±28µg/kg (Table 9).

Cadmium concentrations in Mytilus galloprovincialis range from 5 to 1060 ug/kg (f.w.). Region VI has the lower average (38±6 µg/kg). Most values are below 250 µg/kg and the Mediterranean average (excluding 5% of higher values) is 120±80 µg/kg.

A few samples of Mytilus edulis have been analysed and their mean concentration is 85±34 µg/kg (Table 9).

In the North Sea, mean values reported for Mytilus edulis ranged from 30 - 500 µg/kg (ICES, 1974, 1977a and b). In the ICES North Atlantic baseline study values reported were between 90 and 330 µg/kg (ICES, 1980). In the Oslo

Commission area (1983) Cd concentrations in Mytilus edulis have been reported to range between 43 and 12600 µg/kg with a mean of 1040. Compared with these values Mediterranean values are not increased.

Cadmium concentrations in Thunnus thynnus thynnus were reported in samples from area II. Mean concentration was 38±43 µg/kg. In Thunnus alalunga the mean of reported concentrations were 23±6.5 µg/kg.

Average Cd concentrations in other Mediterranean marine organisms are summarized in Table 9. The highest mean was reported in Mullus surmuletus (140 µg/kg).

Table 9. Average cadmium concentrations in Mediterranean marine organisms µg/kg (fresh weight)

Species	No. of samples	Mean	Standard Deviation
Donax trunculus	16	80	26
Engraulis encrasicolus	81	34	25
Merluccius merluccius	27	63	34
Mugil auratus	10	47	85
Mullus barbatus	318	34	28
Mullus surmuletus	218	140	83
Mytilus galloprovincialis	265	120	83
Mytilus edulis	10	85	34
Nephrops norvegicus	61	50	39
Parapenaeus longirostris	27	46	55
Thunnus alalunga	38	23	6.5
Thunnus thynnus thynnus	111	38	43

Copper

Regional mean concentrations of Cu in Mullus barbatus range from 380-930 µg/kg (Table 10). No significant variations between regions can be observed due to the considerably high standard deviations of the means.

Copper concentrations in Mytilus galloprovincialis are considerably higher with means ranging from 1000-1900 µg/kg (Table 11). These values are within the ranges reported for Mytilus edulis in the North Sea (600-9400 µg/kg) (ICES, 1977a, and b).

Copper concentrations in other Mediterranean marine organisms are summarised in Table 12. Fish have the lowest concentrations (400-2100 µg/kg) whereas bivalves and crustaceans have considerably higher concentrations (1300-8500 µg/kg).

Table 10. Copper concentrations in _Mullus barbatus_ µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	153	200	1300	405	172
IV	208	2.5	1000	380	127
VII	10	360	2700	930	684
VIII	60	220	1470	600	280
X	23	69	2550	800	560

Table 11. Copper concentrations in _Mytilus galloprovincialis_ µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	55	504	4800	1500	900
IV	85	70	6000	1900	1100
V	58	163	4400	1000	900
VIII	13	750	2800	1600	600

Table 12. Average copper concentrations in Mediterranean marine organisms µg/kg (fresh weight)

Species	No. of samples	Mean	Standard Deviation
Donax trunculus	19	3500	1800
Engraulis engrasicolus	97	990	560
Mugil auratus	31	700	960
Mullus barbatus	444	400	140
Mullus surmuletus	20	600	540
Mytilus galloprovincialis	204	1300	700
Nephrops norvegicus	303	5700	1900
Parapenaeus longirostris	22	8500	8000
Penaeus kerathurus	12	5200	2700
Sarda sarda	27	2100	1700

Zinc

Mean concentrations of Zn in *Mullus barbatus* from different Mediterranean regions range from 3500-5100 µg/kg (Table 13). The Mediterranean average (excluding the 5% highest values) is 3900±900 µg/kg (Table 14). There are no significant variations between Mediterranean regions.

In *Mytilus galloprovincialis* mean concentrations range from 17000-45000 µg/kg. Overall ranges are between 3150 and 97700 µg/kg. North Sea data have been reported to be of similar levels (ICES, 1974, 1977a, 1977b).

In Table 14, Zn concentrations in other Mediterranean marine organisms are summarized. Mean concentrations in fish range from 2900 µg/kg in *Upeneus moluccensis* to 18000 µg/kg in *Engraulis encrasicolus*. Bivalves and crustaceans have higher concentrations (11000-41000 µg/kg).

Table 13. Zinc concentrations in *Mullus barbatus* µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	132	100	7100	4000	970
IV	221	400	7000	4000	1000
VII	11	2700	5800	4300	860
VIII	40	2570	6890	3500	800
IX	12	3660	7400	5100	1040
X	23	3060	5870	4400	650

Table 14. Average zinc concentrations in Mediterranean marine organisms µg/kg (fresh weight)

Species	No. of samples	Mean	Standard Deviation
Donax trunculus	17	21000	17000
Engraulis encrasicolus	75	18000	6700
Mugil auratus	66	10600	15000
Mullus barbatus	435	3900	900
Mullus surmuletus	24	10000	14000
Upeneus moluccensis	13	2900	1100
Mytilus galloprovincialis	179	27000	13000
Nephrops norvegicus	279	15000	2800
Parapenaeus longirostris	19	11000	3400
Penaeus kerathurus	22	22000	16000
Carcinus mediterraneus	13	41000	29000

Table 15. Zinc concentrations in Mytilus galloprovincialis µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	26	13000	60200	28000	10700
IV	84	3150	63000	34000	11200
V	58	2500	64250	17000	12000
VIII	21	9200	97700	45000	24600

Lead

Mean concentrations of Pb in Mullus barbatus in three Mediterranean regions range from 60 to 370 µg/kg. Overall ranges were 23 to 610 µg/kg (Table 16).

Lead concentrations in Mytilus galloprovincialis were considerably higher with means of 600-1800 µg/kg. (Table 17).

There is a considerable variation in reported concentrations. Standard deviations are more than 100% of the means.

The overall Mediterranean average (excluding the 5% highest values) is 800±800 µg/kg (Table 18). Data reported from the Canadian coast in ICES North Atlantic baseline study show a similar variation but a lower overall average of 330 µg/kg (ICES, 1980). GESAMP (1983) reports Pb concentrations in Mytilus sp. of less than 1000 µg/kg.

Mean Pb concentratins in other Mediterranean organisms range from 70-1200 µg/kg (Table 18).

Table 16. Lead concentrations in Mullus barbatus µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	173	23	243	60	31
X	22	145	610	370	121

Table 17. Lead concentrations in *Mytilus galloprovincialis* µg/kg (fresh weight)

Region	No. of samples	Minimum	Maximum	Mean	Standard Deviation
II	101	50	6800	600	790
IV	85	50	16100	1800	2400
V	92	50	7825	840	1300
VIII	80	55	8260	1100	1500

Table 18. Average lead concentrations in Mediterranean marine organisms µg/kg (fresh weight)

Species	No. of samples	Mean	Standard Deviation
Donax trunculus	19	1200	650
Mullus barbatus	435	70	45
Mytilus galloprovincialis	344	800	800
Thunnus thynnus thynnus	53	117	170

5. HEALTH EFFECTS

Cadmium

The main route of exposure to cadmium for general human populations is through food, and the contribution by other routes (the working environment, ambient air, drinking water and tobacco) to the total intake is usually small. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) have proposed a Provisional Tolerable Weekly Intake (PTWI) of 0.4 - 0.5 mg per individual of 70 kg bodyweight. This estimate, however, is only provisional, and the Committee has pointed out that it should be revised when more precise data and better evidence become available. Such revision is still ongoing, and to date no environmental health criteria for cadmium have yet been issued.

Absorption of cadmium into the blood averages 5% from the gastrointestinal tract. The remaining 95% are removed with the faeces. It is rapidly cleared from the blood and accumulates principally in the kidneys and the liver. In time, concentrations occurring in the latter organ are re-distributed to the former, where it accumulates in the tissues and/or is excreted in the urine. Repeated exposure results in distribution to, and buildup in, most other organs in the body.

The kidney is the critical organ for effects of cadmium poisoning in man. In high-level inhalation exposure, the lungs may also be affected. This, however, would not occur in intake through seafood. Renal tubular dysfunction may occur when the concentration of cadmium is around 200 µg/g for dry weight in the renal cortex (a factor of about 10 higher than current levels in background-exposed individuals).

Compared to other food classes, the health risk associated with dietary intake of cadmium from seafood would appear to be of minor consequence, with the exception of crustaceans and molluscs. These organisms however, are not a substantial portion of the common diet. Where relatively high concentrations are found, this source of cadmium could be of significance to the total cadmium intake for particular regional population groups.

Copper

According to available evidence, copper in seafood or in seawater does not present any hazard to man.

Zinc

According to available evidence, zinc has not been implicated in any human diseases derived from the consumption of seafood.

The main route of exposure to zinc in man is through inhalation of fumes. There have also been reports of human cases of zinc poisoning associated with the prolonged consumption of water from galvanized pipes. Adverse effects of zinc include metal fume fever from inhalation of fumes, throat irritation, coughing, dyspnoea, muscle and joint pain, gastric irritation, peptic ulcers and various liver effects. There is so far no evidence that zinc in excess is carcinogenic, mutagenic or teratogenic.

Lead

For general human populations, the major contribution of lead to the total daily intake is from food, though water and air may provide significant contributions under certain conditions.

Anaemia is a characteristic early toxic effect of lead in man. In this regard, children appear to be more sensitive than adults. The nervous system is also affected, the effects themselves varying with the duration and intensity of exposure. Lead effects on the brain are much more commonly associated with childhood lead poisoning than in poisoning as it is seen in adults. With chronic lead poisoning, striking effects, referred to as lead encephalopathy, may occur. The major features are dullness, restlessness, irritability, headaches, muscular tremors, hallucinations, and loss of memory and ability to concentrate. These signs and symptoms may progress to delirium, mania, convulsions, paralysis and coma. Signs and symptoms of encephalopathy in infants and young children are quite similar to those reported to occur in adults.

Lead also affects the kidneys and the gastrointestinal tract. In the latter, as a symptom of lead poisoning, colic is a fairly consistent early warning of potentially more serious effects likely to occur with prolonged periods of exposure. Lead colic is also associated with effects on the blood and high degrees of anaemia. Impairment of thyroid function and adrenal function has also been reported in cases of lead poisoning.

A provisional tolerable weekly intake (PTWI) of 3.0 mg of lead for adults was recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1972. (FAO/WHO, 1972). This is equivalent to about 430 µg per day.

In view of the fact that man may have several sources of exposure to lead, it would not appear that seafood consumption on its own presents a health hazard. However, this would have to be considered both in absolute terms (i.e. contamination levels and consumption) and in association with exposure to lead from other sources.

6. CONCLUSIONS

1. Sources of Pb and Zn in the Mediterranean should be considered at present as a first approximation. No estimation of Cd and Cu input has been made due to the lack of data. The new survey of land-based sources will provide more recent information.

2. Very few data exist on heavy metal concentrations in open Mediterranean waters. Coastal waters show increased concentrations of heavy metals where sources are situated. In view of recent progress in analytical techniques, it is questionable whether all these data are reliable. The very few latest data that have been reported and can be considered reliable show that Cd, Cu, and possibly Pb are increased in Mediterranean waters as compared to the Atlantic.

3. Concentrations of Cd, Cu, Pb and Zn are generally considerably increased in sediments of coastal areas receiving sewage, effluents, solid wastes or river discharges. It is impossible, though, to assess the degree of heavy metal pollution without reliable background values. The wide variety of extraction and solubilization methods used in sediment analysis and the general lack of information on grain size, organic carbon and mineralogical content makes comparison of heavy metal concentrations in sediments difficult.

4. Data on concentrations of heavy metals in marine organisms show a considerable variability. It cannot be ascertained whether this is due to natural variability reflecting different environments, or to poor analytical results, or to the spatially uneven sampling. With the exception of Pb, the other heavy metals studied in mussels do not show any evidence of increased concentrations when compared with North Sea or Atlantic mussels.

5. It is impossible at present to evaluate whether considerably increased concentrations in marine organisms from polluted areas of the Mediterranean occur since very few samples have been collected from relatively clean areas.

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