

Hazardous substances in the European marine environment: Trends in metals and persistent organic pollutants

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Summary

The main objective of this report is to evaluate the causes, concentrations, trends and impacts of hazardous substances ⁽¹⁾ in European coastal waters, and to identify how the assessments can be improved. Based on data provided by member countries and marine conventions to the European Environment Agency's working database, Marinebase, riverine and atmospheric inputs and concentrations in mussels and fish were assessed for six substances. The data availability of other hazardous substances is insufficient for European assessments. Four of these substances, cadmium, mercury, lead and lindane, are included in the water framework directive's list of hazardous substances; whereas the remaining two, DDT ⁽²⁾ and PCB ⁽³⁾, are addressed by the UN convention on persistent organic pollutants (Stockholm POPs Convention).

Input and concentration data and further information for cadmium, mercury, lead, DDT, lindane and PCB from the marine conventions — Helsinki Commission (HELCOM), Oslo and Paris Commission (OSPAR), United Nations Environment Programme/Mediterranean Action Plan (UNEP/MAP) — and EEA national reference centres were examined. The data available for the project were scarce and, with the exception of some regions, inadequate for fully assessing the state and trends of hazardous substances at a European level. The input data did not adequately represent all member countries. In particular, information on concentrations in mussels and fish were lacking from the Black Sea, most of the Mediterranean, the Iberian Coast, Bay of Biscay, as well as the coasts bordering the western and eastern North Sea.

Driving forces and pressures

Emission sources and emission patterns differ with regard to the various hazardous substances. The main sources for cadmium, mercury and lead include waste incineration

and industrial activities. The main source for DDT and lindane is most likely leaching from soil (actually not a source but a pathway to the environment — the original source was the application of pesticides). For PCB, the main source is disposal of products containing PCB.

No data on emissions to water of hazardous substances were available for this assessment. Future assessments of emissions will be based on data from the European Pollution Emission Register (EPER) under the integrated pollution prevention and control (IPPC) directive, for which a database is being developed during 2003. Pressure assessment in this report is therefore based on literature review and data on riverine inputs and direct discharges to seas as well as atmospheric deposition.

Direct discharges and riverine inputs of cadmium, mercury, lead, lindane and PCB into the North-East Atlantic have decreased (1990–1999). These trends are based on an aggregation of all available annual data sets. However, no assessment of direct and riverine inputs to the Baltic Sea, Black Sea or Mediterranean Sea was possible due to the lack of data.

The atmospheric inputs of cadmium, mercury and lead to the North Sea, measured via atmospheric deposition, have also decreased in the period 1987–1995.

State and impact

The assessment of levels and trends of concentrations of hazardous substances in coastal waters, and the identification and presentation of hot spots in a series of maps, is the focus of this report. Concentrations in marine organisms decreased for all six hazardous substances in mussels in the North-East Atlantic and the Mediterranean Sea. However, trends for concentrations in fish in the North-East Atlantic and Baltic Sea are less obvious.

(1) The European Union has defined 'hazardous substances' as substances or groups of substances that are toxic, persistent and liable to bio-accumulate; and other substances or groups of substances which give rise to an equivalent level of concern (EU Water Framework Directive 2000/60/EC, Art 2).

(2) The synthetic organochlorine insecticide, dichlorodiphenyltrichloroethane.

(3) Polychlorinated biphenyl (PCB) results assessed in this report are defined as PCB_n or the sum of congeners: CB28, CB52, CB101, CB118, CB138, CB153 and CB180.

Assessments of local levels and trends as presented in a series of maps show:

Cadmium:

Concentrations of cadmium in mussels (both *Mytilus edulis* and *M. galloprovincialis*) during the period 1995–1999 (2001 for Black Sea) indicate that levels were elevated in the estuaries of large rivers, in areas with point source discharges and in some harbours. No ‘high’ concentrations (above EU food stuff limit values) were found for any fish.

At seven locations, different from the observed hot spots, increasing trends in cadmium concentrations of mussels and fish were observed. A total of 283 temporal trends have been analysed statistically on a station-by-station basis, of which only 27 were statistically significant: 20 downwards and seven upwards.

Mercury:

Concentrations of mercury in blue mussels in Europe in the period 1995–1999 were slightly higher than background levels in most areas. The general picture indicates a diffuse, widespread exposure of coastal mussel populations to mercury, presumably mainly atmospheric deposition, but with no real hot-spots. Concentrations were at background levels in fish in the North-East Atlantic and Baltic Sea.

Only four coastal locations showed increasing trends in mercury concentrations in mussels and fish. A total of 278 temporal trends have been analysed on a station-by-station basis, of which only 20 were significant, 16 down and four up.

Lead:

Concentrations of lead in the period 1995–1999 were below the estimated upper limit for OSPAR background in blue mussels in areas far from local or regional sources. However, concentrations of the metal were above background level along most of the European coasts.

At several locations, concentrations of lead in blue mussels were above the limits for human consumption, indicating several hot spot areas. Lead concentrations were generally low in livers of Atlantic cod and in herring muscle in the Baltic Sea.

At nine coastal locations, increasing trends in lead concentrations in mussels were observed, and in most cases these were

different from the observed hot spots. A total of 266 temporal trends were analysed on a station-by-station basis, of which only 39 were significant, 30 down and nine up.

DDT:

Concentrations of DDT in blue mussels in the period 1995–1999 were low in the North-East Atlantic and Baltic Sea.

No increasing trends were observed except for fish at one location. A total of 178 temporal trends were analysed, of which only 15 were significant, and 13 were down.

Lindane:

Data availability was limited to the Mediterranean and Black Sea. Along the French Mediterranean coast in the period 1995–1999 lindane concentrations in mussels were low.

No increasing trends were observed. A total of 25 temporal trends (all mussel monitoring stations from the French Mediterranean coast) had been analysed, of which seven were significant, all down.

Polychlorinated biphenyls (PCB₇):

Levels of PCB₇ in blue mussel in the period 1995–1999 reflect the general exposure pattern for this group of substances and also indicate that long-distance atmospheric transport is less important in temperate parts of Europe than regional or local sources. Concentrations of PCB₇ in mussels in the North-East Atlantic at several locations are at ‘high’ levels (more than 10 times above OSPAR background reference levels). Concentrations of PCB₇ in fish in the Baltic Sea and northern North-East Atlantic Ocean indicate only one hot spot.

Time trend analysis showed no increasing trends. A total of 208 temporal trends were analysed, of which only 18 were significant, all down.

Responses

Response measures on the six substances cover marketing, use, emission, foodstuff limit values and environmental concentrations, and are quite comprehensive. The substances have been regulated for several years. This is reflected in the overall decrease in concentrations of these substances in inputs and in marine organisms.

EU has placed cadmium, mercury, lead and lindane on its list of priority substances for the water framework directive with the target of cessation or phasing out of discharges, emission and losses by 2020. Although DDT and PCB are not on this list, they are given priority in the OSPAR Marine Convention, which has the same general target as the EU. Within the framework of HELCOM, DDT and PCB are banned substances according to Annex I, Part 2 of the 1992 Helsinki Convention (HELCOM, 1992b).

With regard to cadmium, mercury, lead, PCB and other hazardous substances, numerous OSPAR decisions and recommendations as well as HELCOM recommendations have been adopted. The Stockholm POPs Convention prohibits the production and use of PCB, and restricts the production and use of DDT. Parties to the Convention for the protection of the Mediterranean Sea against pollution have identified substances or groups of substances whose discharge, dumping or land-based discharge is prohibited or limited (Barcelona Convention and protocols). Parties to the Convention on the protection of the Black Sea against pollution have identified similar groups of substances and have protocols to reduce pollution of these harmful substances.

Several hazardous substances other than the six assessed in this report are listed by EU water directives and marine conventions but

many are not regulated to the same degree regarding marketing, use, emissions, food stuff limit values, etc. The data availability of these other substances is too poor for present assessments.

Recommendations

Recommendations to improve the availability of data for future assessments of hazardous substances in the coastal and marine environment include the following:

- development of a European database for atmospheric deposition as well as riverine and direct discharges, based on existing regional initiatives;
- harmonised European methods for calculating input loads as well as harmonising monitoring;
- development of improved method for time trend analyses;
- development of European spatial and temporal monitoring for hazardous substances in sediment and biota;
- gathering of spatial information on emission sources;
- development of limit values or environmental standards as a basis for classification and assessments;
- development of an environmental quality classification system covering coastal and marine waters;
- investigation of the usefulness of biological effects methods for European monitoring and assessment.

1. Introduction

For several years, the EEA has been developing maps of selected hazardous substances in coastal waters. An EIONET (European Environment Information and Observation Network) review by EEA member countries on proposed maps was undertaken in November 2000 and several comments for changes were received from EIONET. The EIONET workshop in November/December 2000 gave further comments as did the Inter-Regional Forum workshop in June 2001. DG Environment comments were received in September 2001. Based on these comments and updated data sets from annual data collections, the European Topic Centre on Water undertook a new assessment in 2002 and a report has been produced around the new maps. This report covers the assessment of the information presented in the maps as well as aggregated information on overall trends and their relations to impacts, sources of pollution and reduction measures.

1.1. Definition of hazardous substances

Within existing legislation, no consistent use is made of the terms 'hazardous substances', 'dangerous substances', 'harmful substances' and 'priority substances'. This report uses the term 'hazardous substances', defined as substances or groups of substances that are toxic, persistent and liable to bio-accumulate; and other substances or groups of substances which give rise to an equivalent level of concern (EU Water Framework Directive 2000/60/EC Article 2). HELCOM and OSPAR use the same definition for hazardous substances and their lists of priority hazardous substances include all six substances reviewed in this report.

A first list of chemicals has been established and 33 substances have been identified as 'priority substances' (2455/2001/EC). Of these, 11 are identified as priority substances (1) and 14 are subject to a review for identification as possible priority

hazardous substances (2). This will replace the List I of 129 'dangerous substances' of the Dangerous Substances Directive 76/464/EEC.

Parties to the Convention for the protection of the Mediterranean Sea against pollution (Barcelona Convention) have identified substances or groups of substances whose discharge is prohibited or limited (UNEP/MAP, 1995a (Barcelona Convention), UNEP/MAP, 1995b (Mediterranean dumping protocol), UNEP/MAP, 1996 (Mediterranean land-based sources protocol)). Parties to the Convention on the protection of the Black Sea against pollution (Bucharest Convention) have identified similar groups of substances (BSC, 1992). The Bucharest Convention has protocols to reduce pollution of these harmful substances.

Exposure to excessive loads of hazardous substances can result in a series of undesirable effects to the ecology of the seas and health of human populations. Hazardous substances may affect organisms by inhibition of vital physiological processes (acute toxicity), or they may cause effects that may ultimately threaten populations (chronic toxicity). For example, they may be carcinogenic or have the ability to induce impairment of reproduction. If a substance is persistent, i.e. its degradation process exceeds a certain time span, it remains in the environment and leads to a continuous and/or long-term exposure.

Substances with a high lipophilicity tend to bio-accumulate in organisms and subsequently bio-magnify in the food web. In aquatic organisms, bio-accumulation is the process that causes an elevated chemical concentration in the organism relative to the ambient water. This process involves all exposure routes. Many synthetic contaminants, such as pesticides and PCB, concentrate in the fatty tissues of organisms. This presents a particular problem for

(1) Brominated diphenylether (only pentabromodiphenylether), cadmium, chloroalkanes (C10-13), hexachlorobenzene, hexachlorobutadiene, HCH (lindane), mercury, nonylphenols, polyaromatic hydrocarbons (PAHs), pentachlorobenzene, tributyltin compounds.

(2) Anthracene, atrazine, chlorpyrifos, Di (ethylhexyl) phthalate (DEHP), diuron, endosulfan, isoproturon, lead, naphthalene, octylphenol, pentachlorophenol, simazine, trichlorobenzenes, trifluralin.

marine organisms with high fat content such as mammals. Humans may be affected by the bio-accumulation of hazardous substances through consumption of contaminated marine organisms.

1.2. Chemical policies for European waters and seas

The future European chemical policy is outlined in the White Paper (COM(2001) 88). The policy aims to achieve by 2020 that chemicals are only produced and used in ways that do not lead to a significant negative impact on the human health and the environment. The White Paper refers to all chemicals that exist and aims to improve the present slow risk assessment of existing and new chemicals. Substances that are produced or imported in high quantities will undergo more intensive toxicity testing than those of low quantities. All substances exceeding production volumes of more than one tonne will be registered together with basic information (estimated at 30 000 chemicals). Substances exceeding production volumes of 100 tons will undergo evaluation of toxicity testing programmes (estimated at 5 000 chemicals). Substances with certain hazardous properties that give rise to very high concern (carcinogenic, mutagenic and reprotoxic substances and POPs) will require authorisation (estimated at 1 400 chemicals). This future chemical policy should give an overall protection from hazardous substances to the environment and human health.

Specific measures to reduce riverine inputs, direct discharges and atmospheric deposition of a selection of hazardous substances and to protect the marine environment are being taken as a result of various initiatives at different levels. These include the:

- UN global programme of action for the protection of the marine environment against land-based activities;
- Dangerous Substances Directive (76/464/EEC);
- Water Quality Framework Directive (2000/60/EU);
- OSPAR convention 1998 for the protection of the marine environment of the North-East Atlantic;
- Helsinki convention 1992 on the protection of the marine environment of the Baltic Sea area (HELCOM);
- Mediterranean action plan (MAP);
- Convention on the protection of the Black Sea against pollution;

- Arctic monitoring and assessment programme (AMAP);
- conferences on the protection of the North Sea (e.g. NSC, 2002).

The target of these initiatives is a substantial reduction of the input of hazardous substances to coastal waters, thereby improving the biological state.

The Water Framework Directive (2000/60/EC) aims at the cessation or phasing out of discharges, emissions and losses of priority hazardous substances and the progressive reduction of discharges, emissions and losses of hazardous substances, respectively, within 20 years after the adoption of these proposals by the European Parliament and the Council. The ultimate goal is to achieve concentrations in the marine environment near background values for naturally occurring substances and close to zero for man-made synthetic substances. This has largely replaced (and will totally replace) the Dangerous Substances Directive (76/464/EEC). Goals similar to the water framework directive have also been outlined by OSPAR, HELCOM and North Sea conferences. No similar target has been formulated for the Mediterranean Sea or the Black Sea.

To summarise, the main EU policy objectives are:

- to achieve levels of water quality that do not give rise to unacceptable impacts on, and risks to, human health and the environment (1600/2002/EC), i.e. to achieve levels of contamination that do not exceed environmental quality standards (EQS);
- enhanced protection and improvement of the aquatic environment, inter alia, through specific measures for the progressive reduction of discharges, emissions and losses of priority substances; to ensure the progressive reduction of pollution of groundwater and prevent its further pollution; and the cessation or phasing out of discharges, emissions and losses of priority hazardous substances (2000/60/EC, Article 4);
- by 2020, to ensure that chemicals are only produced and used in ways that do not pose significant threats to human health and the environment (COM(2001)264, p.11);
- to eliminate pollution of the waters by the dangerous substances in the families and groups of substances in List I of the Annex, and to reduce pollution by the dangerous

- substances in the families and groups of substances in List II (76/464/EEC);
- to achieve a situation where the use and levels of pesticides in the environment do not give rise to significant risks to, or impacts on, human health and nature. This will include an overall reduction in the risk associated with the use of pesticides. (1600/2002/EC, chap. 5.5);
 - to substantially reduce operational discharges from oil installations and ships, and to eliminate illegal discharges from these sources by 2010 (EU Marine Strategy, COM (2002) 539).

1.3. Objectives of report and data used

This report cannot deal with all hazardous substances nor with all substances of the different policy lists for cessation of discharges, emissions and losses of hazardous substances, simply because the data and information are not available. The assessment presented here will therefore focus on six substances, three heavy metals (cadmium, mercury and lead) and three organic substances (DDT, lindane and PCB₇) for which the data availability is best.

The main objectives of this report are:

- to evaluate the inputs and concentrations of six selected hazardous substances in Europe's marine and coastal waters on the basis of existing data and information, and to present the information on maps and time trend graphs;
- to evaluate the causes for trends and elevated levels of these six hazardous substances;
- to identify gaps in information.

Data for the following six hazardous substances and media have been collected and analysed:

- **Cadmium, mercury and lead** inputs and concentrations in the North-East Atlantic (including the North Sea) and in the Baltic, and in mussels in the Mediterranean in the period 1985–1999.
- **DDT** concentrations in the North-East Atlantic (including the North Sea) and in the Baltic 1985–1999.
- **Lindane** input in the North-East Atlantic (including the North Sea) and in the Baltic, and concentrations in mussels in the Mediterranean in the period 1985–1999.
- **PCB₇** input and concentrations in the North-East Atlantic (including the North

Sea) and in the Baltic in the period 1985–1999.

Cadmium, mercury, lead, and lindane are on the EU's list of priority substances for the Water Framework Directive (2455/2001/EC). Even though DDT and PCB are not, these substances are on the OSPAR list of 15 chemicals for priority action (OSPAR 1998a). They are characterised as 'severely hazardous' on UNEP's list of 31 chemicals in need of strict control (<http://www.pic.int>).

Within the framework of HELCOM, DDT and PCB are banned substances according to Annex I, Part 2 of the 1992 Helsinki Convention (HELCOM 1992a). With regard to cadmium, mercury, lead, PCB and other hazardous substances, numerous HELCOM recommendations have been adopted, such as HELCOM 1998b, 2002a-e. Protocols have been agreed to prohibit the discharge of organohalogenes, which includes PCB, in the Mediterranean (CPM 1995b, 1996) and Black Sea (CPB 1992b, 1992c). DDT and PCB are banned or restricted by the Stockholm convention on persistent organic pollutants (POPs Convention).

Input data

As part of the work programme for the EEA, the Norwegian Institute for Water Research (NIVA) compiled a European marine database on water quality parameters called Marinebase (EEA, 2001a). This database includes input data for cadmium, mercury, lead, lindane and PCB (1990–1999) from the 1998 OSPAR summary report on riverine inputs and direct discharges (RID), supplemented with data provided by OSPAR for the years 1997–1999. The data concern the North-East Atlantic. No data on inputs of DDT were available.

For the Baltic Sea, HELCOM (1998a) could only provide data on 1995 inputs.

The OSPAR's comprehensive atmospheric monitoring programme (CAMP) provides input data of cadmium, mercury and lead (1987–1995) but only covers the North Sea.

No input data or time series data were available for the Mediterranean.

For the Black Sea, a short series of input loads from priority point sources in Romania were taken from a report of the Black Sea Commission (BSC, 2002).

Concentration data

The report comprises an evaluation of hazardous substances data, which were made available through collection from marine conventions and national reference centres. Due to quality assurance issues and to achieve adequate geographical and temporal representation, mussels (blue mussel — *Mytilus edulis*, Mediterranean mussel — *M. galloprovincialis*) and fish (Atlantic cod — *Gadus morhua*, herring — *Clupea harengus* and flounder — *Platichthys flesus*) were selected as state indicators.

Most of the data collected are from the Commission of the convention for the protection of the marine (OSPAR) and the Baltic marine environment protection commission (HELCOM). They are provided through the International council for the exploration of the seas (ICES) acting as a data centre for OSPAR and HELCOM. France and Greece provided the data that were selected for the Mediterranean. Romania provided the data that were selected for the Black Sea. The data are stored in the EEA’s working database Marinebase (EEA, 2001a).

Concentrations of hazardous substances in blue mussels and fish constitute state indicators for coastal water quality over time.

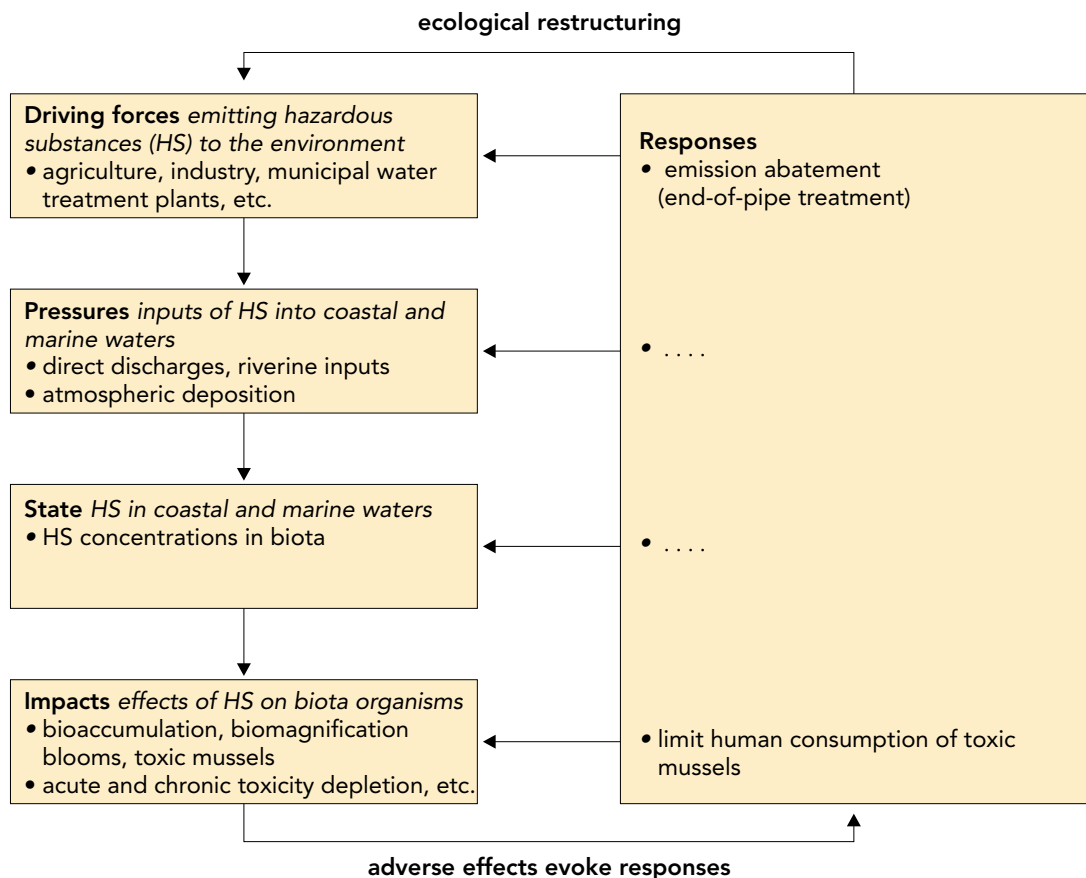
An advantage to using biota concentrations as indicators, as opposed to using water or sediment, is that they are of direct ecological importance as well as being important to human health and fisheries. Mussels are attached to shallow-water surfaces, thus reflecting exposure at a fixed point (local pollution). They are also abundant, robust and widely monitored in a comparable way. Mussels are, however, restricted to the coastal zone. Atlantic cod is a widely distributed and commercially important fish species in the North Atlantic. It is a predator and as such will also to some extent reflect contamination levels in its prey. Herring is a commercially and ecologically important species both in the Baltic and the North-East Atlantic. Flounder is found in some of the most highly contaminated estuaries and is a common species along most European coasts.

Structure of this report

The structure of the report generally follows the DPSIR assessment framework, where D = Driving forces, P = Pressures, S = State, I = Impacts and R = Responses. The DPSIR framework for hazardous substances in the marine environment is illustrated in Figure 1.1. The data analysis of this report covers pressure and state indicators, while driving forces, impact and responses have been assessed by literature review.

Figure 1.1

DPSIR assessment framework for hazardous substances (HS) in coastal waters. (Revised from EEA, 2001b).



In chapter two, a brief characterisation of the different European seas is given concerning drainage areas, run off, major surface currents and water exchange components.

In chapter three, the impacts of hazardous substances in the different European seas are described.

Chapter four deals with sources (driving forces) emissions, riverine inputs and atmospheric deposition. The current levels and recent developments of inputs are analysed.

In chapter five, an overview of the data material used for the present report is presented, with a focus on six hazardous substances and their spatial and temporal extent. The aim is to identify areas where reporting of monitoring data needs to be improved in order to better assess the state of

hazardous substances and to determine the presence of trends.

In chapter six, the present levels and trends (state) of six hazardous substances along the European coasts are assessed. The main emphasis is put on analyses of concentrations in mussels and fish from the coastal areas of countries bordering the North East Atlantic, the Baltic Sea, Mediterranean and Black Seas. Levels and time trends of hazardous substances in these regions are shown on maps in Figures 5.2–5.24.

Chapter seven is a brief review of the important legislation to combat the described hazardous substances.

In chapter eight, some recommendations on monitoring and assessment are given with regard to improvement of assessments of hazardous substances in the marine environment.

2. The seas of Europe

Hazardous substances are brought with freshwater from land via rivers, pipes and groundwater seepage to the coastal margin of the marine environment. Figure 2.1 shows the catchment areas of the different European regional seas. Offshore sources include dumping or incineration at sea, maritime transport and oil/gas production activities. Atmospheric deposition to the surface water of the seas is also an important source of contamination.

Exposure to hazardous substances is to a large extent determined by the hydrography. The sensitivity increases with the residence time and the strength of the stratification of the water column. Over time and in space, contaminated discharges are mixed with marine water bodies by water exchange, tidal currents, local coastal currents and large-scale circulation currents.

This report focuses on levels and trends of hazardous substances in mussels and fish in

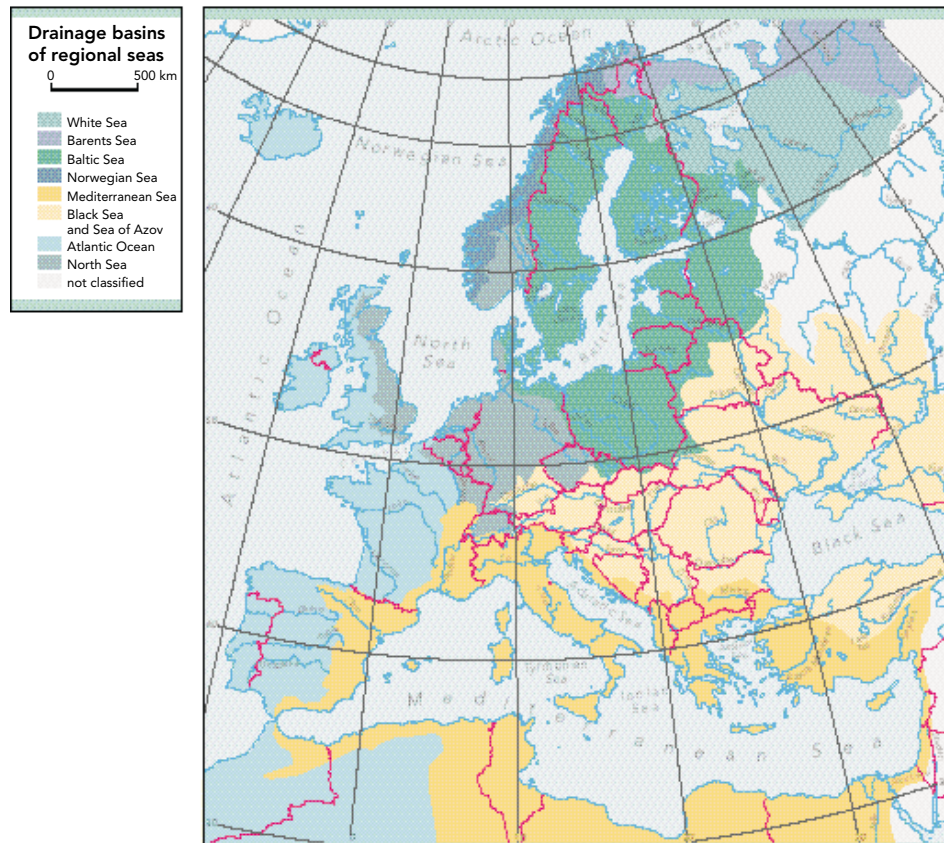
coastal areas. For that reason, this chapter does not include a description of deep waters and deep-water current systems. A comprehensive description of water masses and current circulation patterns in the different seas can be found in OSPAR Quality Status Report (QSR) 2000 for Regions I-V (2000), HELCOM (1996a) and EEA/UNEP (1999).

2.1. Baltic Sea

The Baltic Sea is the largest brackish water area in the world and consists of several sub-basins separated by sills. The transition area (the Belt Sea and Kattegat) to the North Sea is narrow and shallow with a sill depth of 18 m. At irregular intervals, major inflows supply large volumes of highly saline and oxygen-rich water to the Baltic Sea. Only these inflows are able to renew the stagnant bottom water of the deep basins of the Baltic Proper. The Baltic Sea is shallow (mean depth 52 m, maximum depth 459 m) with a water volume

Figure 2.1 Catchment areas and drainage basins of regional seas

Source: EEA, 1999.



of 21 700 km³ and a surface area of 415 000 km², including the transition area. The overall residence time is 25–35 years. In general, ice covers the northern part for about 4–6 months, but sea ice occurs in some winters also in the rest of the Baltic Sea area.

The drainage area is 1 745 136 km² (Figure 2.1). The mean net outflow from the Baltic Sea is 15 000 m³ s⁻¹.

2.2. Norwegian Sea

The only mainland that borders the Norwegian Sea is central and northern Norway. The Norwegian catchment area covers 168 000 km² of mainly mountain areas with little anthropogenic input (Figure 2.1). Two surface current systems run more or less parallel north-east up the coastline. The Norwegian coastal current is running near shore. When it enters the Norwegian Sea from the south-west, it transports approximately one million m³ s⁻¹ and has a salinity that generally exceeds 30. (OSPAR, 2000, QSR for Region I). The total input of the North Atlantic current in the Norwegian Sea is eight million m³ s⁻¹ (OSPAR, 2000, QSR for Region I; AMAP, 1998). In addition to the water masses transported by the two current systems near and offshore, a number of different water masses are found in fjords and other estuaries along the coastline. A wide range of water characteristics exist in coastal waters, depending on estuary type, residence time, freshwater input, mixing conditions, etc.

2.3. Greater North Sea

The Greater North Sea covers approximately 750 000 km² and has a volume of about 94 000 km³. Without strict boundaries it is often divided into seven sub-areas: the Southern North Sea, the Central North Sea, the deeper Northern North Sea, the Norwegian Trench, Skagerrak, the shallow Kattegat as a transition zone to the Baltic Sea and the Channel as a transition zone to the North East Atlantic. The shallow Southern North Sea includes the large Wadden Sea tidal area, the German Bight and the Southern Bight.

The catchment area for rivers discharging to the North Sea is 850 000 km² (Figure 2.1). The total run-off of freshwater is on average 300 km³ y⁻¹, but there are large year to year differences in run-off (OSPAR, 2000, QSR for Region II). The most important rivers are the

Rhine, Weser, Meuse, Scheldt, Seine and Elbe draining central and northern Europe. The Thames and Humber drain east England and the Göeta drains large parts of western Sweden. However, the largest fresh water source to the North Sea is the Baltic Sea (476 km³ y⁻¹).

The flushing time for the entire North Sea is estimated to be in the range of 365 to 500 days (OSPAR 2000, QSR for Region II). In periods with westerly wind, low saline water, carrying hazardous substances from the rivers in the Southern North Sea, is carried up the Jutland west coast by the Jutland Coastal Current to the Skagerrak (OSPAR, 2000, QSR for Region II).

2.4. Bay of Biscay, Iberian West Coast and Gulf of Cadiz

The catchment area that drains into the western Atlantic Ocean is 700 000 km² (Figure 2.1), with an average annual run-off of 180 km³. More than half of the run-off takes place in the Bay of Biscay. The four most important rivers responsible for over half the loading are the Loire and Gironde that drain into the Bay of Biscay, and the Miño and Douro that drain into the Atlantic at the Iberian west coast (OSPAR, 2000, QSR for Region IV). Most of the surface waters found in the area have an Atlantic origin. Deeper water masses may also be a mixture of Atlantic and Mediterranean water masses.

2.5. Celtic Seas

This area includes the Celtic Sea located south of Ireland, Saint George's Channel, the Irish Sea and North Channel between Ireland, Wales, England and Scotland, and finally the shelf areas west of Ireland and Scotland. The total catchment area is shown in Figure 2.1.

The continental slope current, also described for the Biscay area, follows the slope edge to the south-west of Ireland. On average there is a northward water movement through the Irish Sea and 30 000–100 000 m³ s⁻¹ is transported through the North Channel, but transport fluctuations are huge due to wind conditions.

Based on transport models and radionuclide distributions it is estimated that the flushing time is 150–300 days in the Bristol Channel, 1–2 years in the Irish Sea and 4.5–6 months in the Marlin Shelf area. Storm events are

known to change those rates considerably (OSPAR, 2000, QSR for Region III).

In addition to the water masses in the open areas, a number of different waters are found in fjords and other estuaries along the west coast of Scotland and the Scottish Islands. Wide ranges of water characteristics exist, depending on estuary type, residence time, freshwater input, mixing conditions, etc. (OSPAR, 2000, QSR for Region III).

2.6. Mediterranean Sea

Data on the total size of the Mediterranean catchment area were not found. The Nile has the largest catchment area of all rivers entering the Mediterranean Sea with 335 000 km² (Figure 2.1). However, because of the construction of the Aswan dam, an average of only five km³ water is discharged into the sea each year. Other important rivers are all located in the northern part of the Mediterranean. With a few exceptions, all river systems discharging into the Mediterranean Sea are small. The Rhône, Ebro and Po have catchment areas extending to 96 000, 84 000 and 69 000 km². The discharge of freshwater from the 50 main rivers is about 255 km³y⁻¹. Net inflow from the Black Sea amounts to 163 km³ a year.

Evaporation exceeds precipitation and freshwater load. As a result, there is a net inflow of 1 700 km³ water from the Atlantic Ocean each year and the overall effect is a very high salinity in the Mediterranean Sea. The actual annual inflow of Atlantic water is much higher. Chou and Wollast (1997) estimate that 53 000 km³ pass the Strait of Gibraltar each year as a surface current. This

inflow is compensated by export of high saline water from the Mediterranean Sea at the bottom. The export is estimated at 50 500 km³, giving a net water transport 800 km³ higher each year than the figure given in the Mediterranean assessment report (EEA/ UNEP, 1999).

The Mediterranean is divided into two basins separated by the Sicilian Channel about 150 km wide and with a maximum water depth of 400 m. The water depth averages 1 500 m. Shelf areas are narrow and separated from the deeper parts by steep continental shelf breaks. The inflow current of Atlantic water continues as a surface current from west to east of the Mediterranean Sea. Coastal sea level variations are generally limited to tens of centimetres. Tidal amplitudes are small in the Mediterranean.

2.7. Black Sea

The drainage basin of the Black Sea extends to an area of two million square kilometres — five times the surface area of the sea itself. About 350 cubic kilometres of river water pour into the Black Sea each year from an area covering almost a third of continental Europe and including significant areas of 17 countries: Austria, Belarus, Bosnia and Herzegovina, Bulgaria, Croatia, Czech Republic, Georgia, Germany, Hungary, Moldova, Slovakia, Slovenia, Romania, Russia, Turkey, Ukraine and Yugoslavia. Europe's second, third and fourth rivers (the Danube, Dnipro and Don) all flow to the Black Sea. Isolation from the flushing effects of the open ocean, coupled with its huge catchment, have made the Black Sea susceptible to pollution (Acar, 2001).

3. Impact of hazardous substances

Hazardous substances can be broadly divided into the following categories: heavy metals, persistent organic contaminants and degradable organic contaminants. The substances discussed in this report concern the two former categories. Substances in the latter category include organophosphates, alkylphenols and several endocrine disrupters. They are not covered here.

Marine conventions undertake biological effects monitoring of hazardous substances but, with a few exceptions (e.g. HELCOM, 2001d), data are not reported regularly and assessments have not been made yet. The information presented in this chapter on the impact of six substances is therefore based on literature review and not on data assessment.

3.1. Impacts of metals

Metals are found naturally in marine ecosystems. Human activity has increased local, regional and global fluxes of many metals, e.g. the metals included here (cadmium, mercury and lead). All three metals are non-essential (have no known biological function) and tissue levels are therefore generally not well regulated by biological functions of the bodies of marine organisms. This causes bio-accumulation. Metals generally bio-accumulate in tissues other than fat (although methyl mercury may be found in fat). In vertebrates, cadmium accumulates in kidneys and liver, whereas lead accumulates in bone.

Mercury may bio-magnify in marine food chains, whereas cadmium and lead will not. Due to bio-magnification, dietary preferences render some species at risk. Some invertebrate species, such as blue mussel, may accumulate high levels of metals with no apparent adverse effects. Eider ducks that predominantly feed on mussels in some areas are, however, thought to be at risk (secondary poisoning). A similar mechanism is thought to be relevant for seabirds that depend on specific fish species with high mercury levels.

3.1.1. Cadmium

Cadmium is widely present at low concentrations in the earth's crust, but human activities have caused a general

mobilisation of the metal in aquatic and terrestrial environments. It is not needed for any organism (it is not essential) and the metal is highly toxic. The metal affects vital biological processes such as ion exchange, energy production and protein synthesis, mainly through interaction with the metabolism of essential trace metals such as zinc and calcium. In marine ecosystems, some seabird species (eating contaminated mussels) has been identified as the possibly most sensitive component through secondary poisoning (OSPAR, 1996).

In humans, cadmium has a biological half-life of more than 20 years. Concern about long-term exposure to cadmium also intensified after recognition of a disease called 'Itai-Itai' (painful-painful) in certain areas of Japan. The disease was a combination of severe kidney damage and painful bone and joint disease. The disease occurred in areas where rice contained high levels of cadmium due to irrigation of the soil with water contaminated with cadmium from industrial sources. The toxicity also resulted from consumption of cadmium-contaminated fish taken from rivers near smelting plants.

3.1.2. Mercury

Mercury has no known biological function. It is highly toxic and is considered one of the most dangerous metals in the aquatic environment. This is due to its toxicity and potential for bio-accumulation and bio-magnification, particularly under anoxic conditions favouring the transformation of inorganic mercury into organic forms.

Organic forms of mercury affect the nervous system, while the inorganic forms affect a range of cellular processes. Mercury may be demethylated to varying extents in the tissues of aquatic organisms. In marine ecosystems, organisms at the top of food chains, mainly seabirds and marine mammals, have been identified as being most sensitive (through secondary poisoning). The metabolism and disposition of mercury is complex. Most is found as methyl mercury, which has a high affinity for sulphhydryl groups in addition to being fat-soluble. In some species it will be found associated with fat, in others associated with peptides.

Mercury, as is the case for DDT discussed below, is shown to have been a major culprit for the decline in populations of predatory birds in the 1960s and 1970s. Mercury differs from other organic metals in that it is found in one organic and three inorganic forms in the environment (methyl mercury, di- and monovalent ionic mercury and metallic mercury). There is a continuous microbial transformation of inorganic to organic mercury in the aquatic environment. Mercury in fish, for example, is nearly all organic mercury.

Methyl mercury is one of the few environmental contaminants that have been established as embryotoxic to humans. Between 1954 and 1960, at Minimata Bay and elsewhere in Japan, many infants were born with severe neurological symptoms resembling cerebral palsy and mental deficiency. This disease was traced to the consumption of mercury-contaminated fish by the mothers. By 1970, at least 107 deaths had also been attributed to mercury poisoning, and 800 cases of 'Minimata disease' were confirmed.

Apart from the known effects of mercury on birds of prey (mentioned above), lethal or harmful effects in marine and terrestrial mammals are also reported (AMAP, 1998), when concentrations exceeded from 25 to 60 mg/kg wet weight in kidneys and liver. Methyl mercury is a central nervous system toxin, while the kidneys are the organs most vulnerable to damage from inorganic mercury. Methyl mercury is also a risk to the developing foetus since it readily crosses the placental barrier (AMAP, 1998). Mercury exposure has been linked to reduced mental development in children.

The Polar Regions are affected by long-range transported mercury, and the concentration in some marine mammals seems to have increased over the previous two decades (AMAP, 1998). The levels of mercury in Arctic ringed seals and beluga whales have increased two to threefold over the last 20 years (Muir *et al.*, 1997; Wagemann *et al.*, 1996). The Arctic marine food web is often highlighted with regard to the risk of mercury to ecosystems. It is important however to acknowledge that impacts of mercury are not only restricted to the Polar Regions. In warmer waters also, predatory marine mammals may be exposed to mercury levels that are health threatening. For instance, in a study of Hong Kong's

population of hump-backed dolphins, mercury has been considered a health problem (Parsons, 1998).

3.1.3. Lead

Lead is also non-essential and toxic. Lead has high affinity for particles and is rarely found in high concentrations in seawater. Some algae are especially sensitive to lead (OSPAR, 1996), but lead may affect aquatic species at different trophic levels.

In vertebrates, lead predominantly accumulates in bone and blood. Exposure to high concentrations will cause decreased synthesis of haemoglobin and eventually anaemia. Severe exposure to inorganic lead may cause encephalopathy and mental retardation. The same mechanism is used as a biomarker (ALA-D) to monitor lead exposure in fish under the Norwegian OSPAR joint assessment and monitoring programme (JAMP).

3.2. Impacts of DDT, lindane and PCB

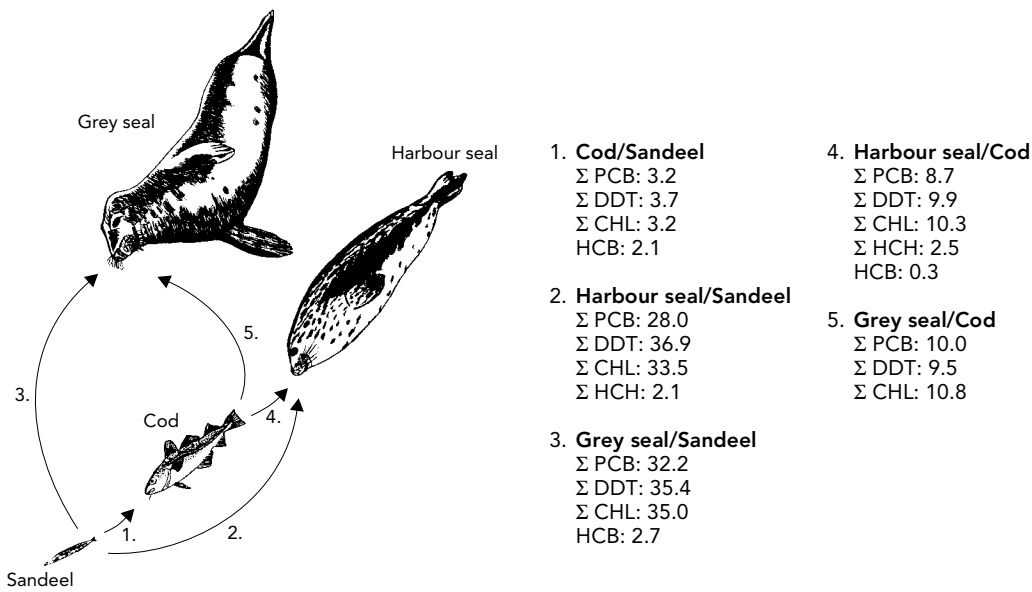
Persistent organic contaminants, such as DDT, lindane and PCB, have low water solubility, high lipophilicity and are resistant to biodegradation. These properties lead to uptake and accumulation in the fatty tissues of living organisms, in some instances causing bio-magnification through food chains (Figure 3.1). The highest concentrations of organic contaminants are therefore found in top predators, such as sea birds, marine mammals and polar bears (Bernhoft *et al.*, 1997; Ruus *et al.*, 2002). Adverse effects of these contaminants are often observed in the same species. Such effects may comprise disruption of the immune system, disruption of hormone production or transport, impairment of reproduction, embryonic damage, cancer or damage to the nerve system.

The case for seals in the Baltic Sea illustrates the impact of certain persistent organic pollutants. The seals are top predators that have been adversely affected by organic contaminants. The Baltic Sea is connected to the world oceans only through three narrow and shallow straits and it takes approximately 25 years to exchange the whole water volume. This is more than enough time for particle bound contaminants to deposit on the Baltic Sea floor. The Baltic Sea has therefore become a sink for many of the hazardous substances that have been deposited in the surrounding environment through the years.

Bio-magnification factors (BMFs) between levels in the studied food chain from Jarlfjord, Northern Norway (1989–1990)

Figure 3.1

Source: Ruus *et al.*, 1999



Seventy million people live close to the Baltic Sea and more than 250 large rivers contribute to a substantial input of contaminants to the Baltic Sea.

Populations of seals in the Baltic Sea declined towards the 1970s. It was established that this was due to lowered reproductive success. The reason was pathological changes in the uterus, rendering the female seals infertile. The severity of disease correlated with levels of organic contaminants in the seals, specifically PCB (Helle *et al.*, 1976). The high contaminant levels were likely the cause of a large disease complex with a variety of symptoms including lesions on skin, claws, intestines, kidneys and bone tissue in addition to uterus pathology. The origin for the whole disease complex was most likely the contaminants' toxic effect on

the adrenal cortex (Bergman and Olson, 1985).

3.2.1. DDT

Dichlorodiphenyltrichloroethane (DDT) is a synthetic organochlorine insecticide. A negative relationship between concentrations of p,p'-DDE (the principal DDT-metabolite) and testosterone in Dall's porpoises of the North Pacific has been shown (Subramanian *et al.*, 1987, Figure 3.2). This may have consequences for the reproduction of the animals, since testosterone plays a crucial role for normal male sexual differentiation, steroidogenesis, spermatogenesis, development of secondary male sex characteristics and sexual behaviour. In other cases, dioxin-like compounds have displayed anti-estrogenic effects, likely because the Ah-receptor (dioxin-receptor) blocks the normal function of the oestrogen-receptor.

Relationship between concentrations of testosterone and p,p'-DDE in Dall's porpoises

Figure 3.2

Source: Subramanian *et al.*, 1987.

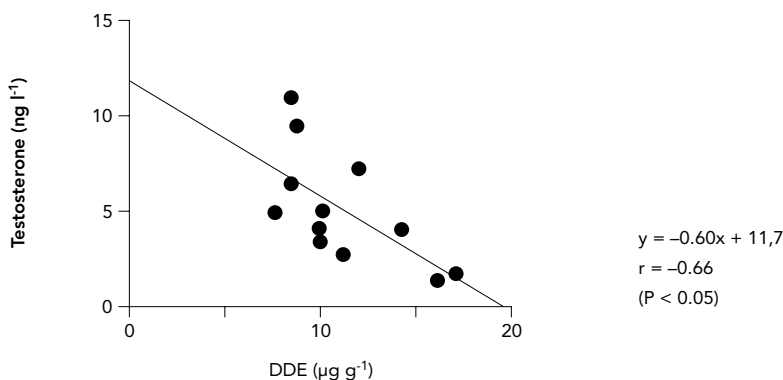
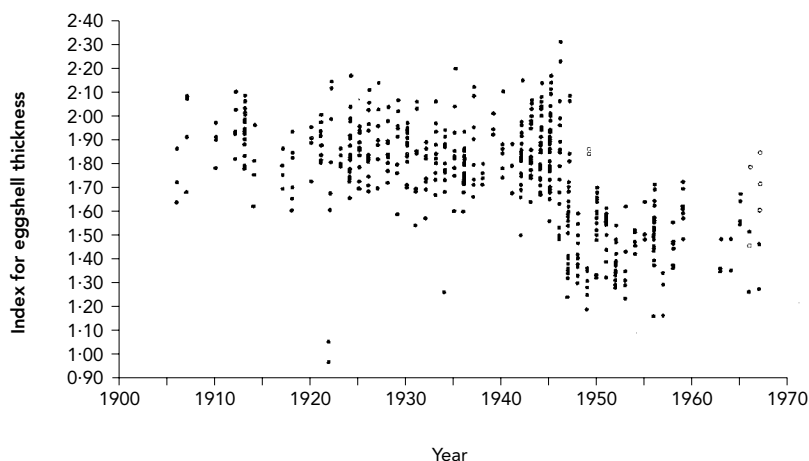


Figure 3.3

Change in eggshell thickness from 1900 to 1967, with lower levels from about 1945 corresponding to when DDT was introduced to the environment

Source: Ratcliffe, 1967.



Besides interfering with hormone homeostasis in Dall's porpoises, DDT and its derivatives were responsible for eggshell thinning and decreased reproductive success in birds, especially birds of prey (Figure 3.3). Endocrine disruption rendered the calcium-producing eggshell-gland of the female birds unable to produce sufficiently thick eggshells and they often broke before hatching (Ratcliffe, 1967; Hickey and Anderson, 1968). These hazardous substances thus contributed to a decline in many bird populations in industrialised areas of the Northern Hemisphere in the 1960s and 1970s.

3.2.2. Lindane

Lindane is also present in high concentrations in the fat of Arctic mammals (seals, polar bears). From the limited data available on acute and chronic toxicity, some crustacean species appear to be particularly sensitive to lindane (and of course insects in freshwater), whereas molluscs and algae, for example, do not appear to be very sensitive (OSPAR, 1996). Lindane is an irritant in humans and may affect mucus membranes, immune and nervous systems, following exposure.

3.2.3. PCB

Due to PCB's extreme mobility and ability to bio-accumulate and bio-magnify in marine food webs, long-lived animals at high trophic levels appear to be most at risk from PCB. Similarly to DDT, PCB are thought to be involved in the observed reproductive

problems of polar bears and possibly earlier morphological aberrations in Baltic seals.

Other endocrine disrupting effects of hazardous substances are, for instance, the binding of hydroxy-metabolites of PCB on the transport protein (transthyretin, TTR) of the thyroid hormone thyroxin. Since thyroxin is an important hormone for growth and development, this may have damaging effects on early life stages of organisms. A similar problem exists for the homeostasis of vitamin A, which is also important for the immune system, foetal development, growth and reproduction in all vertebrates. Relationships between plasma levels of organochlorines (PCB), retinol (vitamin A) and thyroid hormones in polar bears from Svalbard have been found (Skaare *et al.*, 2001), indicating that growth development of epithelial tissues and the immune system, and reproduction are (or are at risk of being) impaired. Furthermore, PCB-contaminated fish are shown to likely induce vitamin A and thyroid hormone deficiency in seals (Brouwer *et al.*, 1989).

Adverse effects of hazardous substances on the reproduction of top predators in marine ecosystems are also observed in the Wadden Sea, which receives water from the river Rhine. Reijnders (1986) observed lowered reproductive success in seals feeding on fish from the Wadden Sea (contaminated with high concentrations of PCB especially), compared with seals feeding on fish from the north-east Atlantic.

Furthermore, suppression of the immune system function (specifically natural killer cell activity) has been observed in seals that fed on Baltic Sea herring (also highly contaminated with PCB) (Ross *et al.*, 1996). During the 1988 epizootic, impaired immune system due to exposure to hazardous

substances was suspected to render the animals more susceptible to the phocine distemper virus. More recent results (Bernhoft *et al.*, 2000) now also indicate contaminant-associated suppression of antibody-mediated immunity in polar bears.

4. Sources and inputs of hazardous substances (driving forces and pressures)

4.1. Sources

In today's industrialised society, a vast number of chemicals are in use or being produced all the time. In addition, several substances are formed unintentionally as by-products in industrial processes or combustion. Human activities have thus drastically altered concentrations of many natural substances in the environment, such as metals, and added numerous new chemicals, such as DDT and PCB. Many substances have been investigated for their impact on the environment. Research has increased the awareness of their harmful effects, leading to restrictions on production, use and discharges of a number of chemicals.

Considerable attention regarding environmental pollution focuses on the marine environment. Different contaminants have reached the oceans from anthropogenic sources through direct discharge to marine waters, or indirectly through rivers and run-off from soil and atmospheric deposition. The world oceans comprise 71 % of the earth's surface and, therefore, are also an enormous recipient of pollutants from the atmosphere. After emission to the atmosphere, many pollutants are dispersed by repeated deposition and evaporation. By oceanic and atmospheric circulation, pollutants are subject to long-range transport even to relatively pristine areas, such as the Arctic.

The European Pollution Emission Register (EPER), which is currently being developed in connection with the integrated pollution prevention and control (IPPC) directive, will provide some data on a range of hazardous substances and their emissions in future. For the Baltic Sea, the pollution load compilation will provide more comprehensive data covering more point sources. The information required for identification of pressures in the water framework directive will cover a wider range of sources than EPER. For this report, information on emission sources has been compiled from literature. Only emission sources of the six substances dealt with in this report are

considered (cadmium, mercury, lead, DDT, lindane and PCB).

4.1.1. Cadmium

Cadmium is referred to as 'the lost metal' since recycling has been limited. The main sources are from general waste/disposal and industrial activities (NSC, 2002). Sources of the metal to the environment include mining and production, metal industry (including steel industry), coating/electroplating industry, production and deposition of batteries, burning of fossil fuels, the use of phosphate fertilisers, waste incineration, leaching from waste deposits and the use of cadmium salts as stabiliser and/or colouring agent (see, for example, HELCOM, 2002f).

Produced water (water produced with oil and gas) is a source of direct discharge of hazardous substances to marine waters and contains a variety of chemicals that have been dissolved from the geologic formations on the sea floor. The metals present and their concentrations in produced water from different sources vary considerably, depending on the age and geology of the formations from which the oil and gas are produced (Collins, 1975).

Cadmium is one of the metals present in North Sea produced water, and the concentrations range commonly from 0.45 to 1.0 µg/L (Neff, 2002). For comparison, typical cadmium concentrations in sea water are between 0.001 and 0.1 µg/L (Neff, 2002). However, inputs of cadmium to the North Sea from offshore activities (estimated at 0.1–0.2 tonnes a year from all sectors) are negligible in comparison with atmospheric deposition (37.5–48 tonnes a year) and riverine and direct inputs (28.6–36 tonnes a year). (See Borgvang *et al.*, 2002.)

4.1.2. Mercury

As with other metals, mercury is found naturally in the marine environment. The main anthropogenic sources are from general waste/disposal and industrial activities (NSC, 2002). It is still used in various products, e.g. batteries and electronics. Furthermore, low quantities in

fossil fuels and municipal waste ensure continued emissions of mercury into the atmosphere. (See also HELCOM, 2002g.)

Mercury differs from other metals and organic contaminants. As has been noted in section 3.1.2, mercury is found in one organic and three inorganic forms in the environment (methyl mercury, di- and monovalent ionic mercury and metallic mercury). There is a continuous microbial transformation of inorganic to organic mercury in the aquatic environment. Mercury in fish, for example, is nearly all organic mercury.

Atmospheric mercury exists mainly in the form of elemental mercury vapour (Hg⁰) (90 to 99 %), particle bound mercury (< 5 %) and gaseous divalent mercury (e.g. HgCl₂) (< 5 %) (EC, 2001). Emissions from anthropogenic sources can occur in all three forms. Atmospheric deposition occurs mainly via dry deposition or wash-out of particle bound and gaseous divalent mercury. Elemental mercury contributes to the deposition via oxidation in air or in cloud droplets. Mercury in the form of elemental vapour (Hg⁰) has a long atmospheric lifetime, which makes transport on hemispheric and global scales feasible. In the Northern Hemisphere, anthropogenic emissions have increased the background concentrations of mercury in air by a factor of 2–3 since before industrialisation.

In addition to diffuse inputs (sewage, runoff), important mercury inputs are atmospheric deposition (burning of fossil fuels, waste incineration) and industry outputs. Recent atmospheric deposition to the North Sea was estimated to be 10.5 tonnes a year, while riverine and direct inputs range between 6.8 and 8.5 tonnes a year (OSPAR, 1999; Borgvang *et al.*, 2002). According to Neff (2002), the global anthropogenic emissions of mercury may amount to 30 000 metric tonnes a year.

As mentioned, mercury undergoes transformations in the marine environment. Mercury has affinity for particles and organic material and will therefore accumulate in some sediments in coastal waters. Mercury is more volatile than other metals and atmospheric transport is an important pathway.

4.1.3. Lead

Lead is widely distributed in the crust of the earth, most commonly found with deposits of other metals (zinc, cadmium, silver, copper). The main anthropogenic sources are from general waste/disposal and industrial activities (NSC, 2002).

Lead does not occur naturally in the metallic state and has a low abundance (0.002 % of the weight of the earth's crust) (EC, 1997). In the natural cycle, lead is released to the earth's surface through several processes which include weathering of rocks, volcanic activity, and uptake and subsequent release by plants. The weathering of rocks releases lead to soils and aquatic systems. This process plays a significant role in the global lead cycle, but only rarely results in elevated concentrations in any environmental compartment. Low concentrations of lead in the air are between 0.05–0.1 µg/m³ in rural areas. Natural lead content in the atmosphere is estimated to be in the order of 0.006 µg/m³.

Lead has a great number of industrial applications, both in its elemental form and in the form of alloys and compounds. The major use of lead is in the manufacture of lead accumulators (responsible for 68 % of consumption and growing in parallel with the increase in the number of automobiles). Here it is used both in the metallic form and as lead oxide. Lead also has important applications in radiation shielding (in nuclear plants and around X-ray apparatus units), in roofing and for the sheathing of electric power cables. Its corrosion resistance makes it a useful material for tank linings, piping and other equipment used in chemical processes. Due to its density, it is also suitable for ammunition and for weighting applications (e.g. car wheel weights and yacht keels).

Lead oxide is also used in several applications. It is incorporated into glass to prevent the escape of radiation from cathode ray tubes (e.g. TV and computer monitors). Organic lead salts are added to PVC as stabilisers to protect it against degradation.

It is also important to mention tetraethyl lead, which has been used in petrol. In 1972, around 400 000 tonnes of tetraethyl lead were consumed throughout the world to improve the octane rating of petrol. Since then, this application has declined dramatically because of restrictions imposed

through environmental legislation. The tetraethyl lead market now accounts for only 1 % of lead use, most of which is used in developing countries.

4.1.4. DDT

DDT (dichlorodiphenyltrichloroethane) is a synthetic organochlorine insecticide that was first used to control insects that were vectors for human diseases at the end of World War II. After the war, it found a ready market in peacetime agricultural enterprise. All use of DDT was discontinued in western as well as central and eastern European countries around 1990, although heavy use was banned two decades earlier. However, in developing countries, the need for cheap insecticides (to control mosquitoes, and hence malaria) kept DDT in use in later years. In some areas, such as in fruit-growing areas in western Norway, there have been recent inputs of DDT, probably due to leaching from buried waste or unused canisters. In addition, there is a continued leaching from soil and river sediments in some areas. As an obsolete pesticide, DDT also poses a severe threat to marine environments when stored under unsafe conditions (HELCOM, 2001c). Over the past 50–60 years, DDT has been spread over the entire globe and is now found in all natural waters and organisms.

The original pesticide DDT (technical grade) contained up to 14 different chemicals including p,p'-DDT (1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane) and its o,p'-DDT isomer (1,1,1-trichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethane), as well as their dechlorinated analogues p,p'- and o,p'-DDD (1,1-dichloro-2,2-bis(4-chlorophenyl)ethane) and (1,1-dichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethane), respectively. p,p'-DDE (1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene) is the principal DDT-metabolite.

4.1.5. Lindane

HCH (1,2,3,4,5,6-hexachlorocyclohexane), also known as benzenehexachloride (BHC) or lindane, is a pesticide that is still used in parts of the world. In its production process, benzene and chlorine react in the presence of UV radiation, forming the crude product 'technical HCH'. This consists of 55–80 % of the a-isomer, 5–14 % of the b-isomer, 8–15 % of the g-isomer and 2–16 % of the d-isomer (Li *et al.*, 1996). Its insecticidal properties are mainly due to the g-isomer. Therefore, by selective crystallisation, g-HCH has been produced with the common name *lindane*. Non-agricultural use of lindane includes use

for wood preservation, as an insecticide, as rodenticide and for medicinal purposes (scab and louse ointments).

Lindane is not found naturally and is toxic to vertebrates as well as to insects (the target for the pesticide). The substance has been banned from use in most western European countries (SIME, 2002), but is still present in the environment due to its persistence.

Lindane was primarily used in the USA and western Europe, while technical HCH was used in the former Soviet Union, Poland, Romania, India, China, Mexico and Brazil (Fischer *et al.*, 1991). Technical HCH is still used in China as an insecticide on hardwood logs and lumber, seeds, vegetables and fruits, and on buildings (AMAP, 1998). The global usage of technical HCH and lindane is estimated to be 550 000 and 720 000 tonnes, respectively, between 1950 and 1993 (Voldner and Li, 1995).

4.1.6. PCB

Polychlorinated biphenyls (PCB) are a group of theoretically 209 different compounds (congeners) of which 150–160 are found in the environment. It should be noted that the range of PCB are very different substances, both with regard to physico-chemical properties and with regard to their biological activity. A distinction is commonly made between the generally more carcinogenic 'dioxin-like' PCB (non- and mono-*ortho* chlorinated) and the more immunotoxic 'bulky' PCB (chlorinated in = 2 *ortho* positions). All PCB are man-made, but are now found all over earth due to their persistence and relative volatility.

Since 1929 at least one million tonnes have been produced. More than half that quantity is still in the environment or in products (Tanabe 1985, 1988). Commercial PCB-products were manufactured in the USA, Japan, the former Soviet Union, and eastern and western Europe under various trade names, such as Aroclor, Clophen, Phenoclor, Santotherm and Pydraul (de Voogt and Brinkman, 1989; AMAP, 1998). The products are mixtures of several congeners with different degrees of chlorination. The PCB have unique physical and chemical properties, such as high thermal and chemical stability and high electrical resistance. Therefore, they have been used extensively in many industrial applications, such as hydraulic fluids, cooling liquids in transformers and dielectric liquids in capacitors. They have also been applied in

open systems such as plasticisers, lubricants, inks and paints. Sources of PCB to the environment are mainly diffuse and the highest levels are generally found in estuaries and close to industrialised urban centres. There is currently a ban on the open use of PCB.

4.2. Inputs

During the period 1990–1999 decreasing inputs of cadmium, mercury, lead, lindane and PCB into the North-East Atlantic were observed (Figure 4.1 and Tables 4.1–4.2). There were no available data for input of DDT. Emission reduction measures in industry have contributed to the observed reductions in the input of metals (OSPAR, 2000; HELCOM, 2001b, 2002i).

The sum of direct and riverine inputs of **cadmium** (about 109 tons in 1990 into the North-East Atlantic) was reduced by 50 % between 1990 and 1997. The UK contributed most to the input in 1990 and also strongly contributed to the total decrease in input observed in the period until 1999. High cadmium inputs in 1994 and 1995 of the Netherlands probably were due to relatively high (and not representative) concentrations of suspended solids in the samples. Data from Portugal have an irregular pattern over time. Values from Spain for 1997–1999 were relatively high compared to other countries, but were lacking prior to 1997 or questionably low. These examples of gaps or poor data inhibit adequate assessment of temporal trends.

The input of **mercury** (about 41 tonnes in 1990) was reduced by about 50 % in 1999 compared to 1990. Germany (including former GDR) and the UK contributed most to the input in 1990. Both countries also strongly contributed to the total decrease in input observed in the period until 1999.

The discharge **lead** in 1990 was about 2 200 tonnes in 1990 and was reduced to 1 330 tonnes in 1999, almost entirely due to changes in discharges from Portugal. Portugal and the UK contributed most to the 1990 input. The contributions of the other countries did not show a clear decrease over the period considered. Relatively high lead inputs in 1994 and 1995 of the Netherlands probably were due to relatively high, but not representative, concentrations of suspended solids in the samples. The results of Spain in 1997 and 1998 were high, while data on 1990–1992 were lacking. This causes a bias in

comparing total 1998 inputs with 1990 inputs. Inputs of Norway and UK in 1998 were high compared to those of other years.

The sum of direct and riverine inputs of **lindane** was about 1 800 tonnes in 1990 and reduced to about 900 tonnes by 1999. Both Norway and the UK in particular contributed to this decrease. Taking into account the change in analysis procedure in the Netherlands from 1993 onwards, the decrease in input in 1999 compared to 1990 was much larger.

The discharge of **PCB** was about 3 100 tonnes in 1990 and was reduced by 50 % by 1999. The UK contributed most to the input in 1990 and strongly contributes to the total decrease in input observed.

Atmospheric inputs were also shown to decrease. A decreasing trend in atmospheric input of cadmium, mercury and lead into the North Sea was observed for the period 1987–1995. There were no available data for atmospheric inputs of DDT, lindane or PCB. Cadmium and lead have rather short residence times in the atmosphere and were rapidly deposited. Countries bordering coastal waters (the OSPAR contracting parties) account for nearly all of the metal input. Due to the air transport characteristics, the relative contribution of atmospheric input varies between the sea regions studied. Emission reduction measures in industry and waste burning contributed to the reduction of atmospheric inputs of cadmium, mercury and lead.

In 1995, atmospheric inputs of cadmium and mercury into the North Sea were estimated at 22 tonnes and four tonnes, respectively. For lead, the atmospheric input into the greater North Sea was estimated at 670 tons. However, the uncertainty margin in these inputs was relatively large (the inputs were probably overestimated). The sum of direct and riverine inputs into the North Sea in 1995 amounted to 56 tonnes, 16 tonnes and 1 420 tonnes for cadmium, mercury and lead, respectively, showing that the contribution of atmospheric inputs was significant. According to HELCOM (2002i), the atmospheric input of cadmium and lead to the Baltic Sea has decreased by about 4 % between 1996–2000. Mercury has decreased by about 14 %. Lindane emissions have decreased by almost two orders of magnitude between 1990–1998, but deposition has only decreased by 14 %, probably due to atmospheric input from elsewhere.

Figure 4.1

Change (%) in direct riverine and atmospheric inputs of cadmium, mercury, lead, lindane and PCB to the North-East Atlantic

Source: OSPAR, HELCOM, EEA member countries.

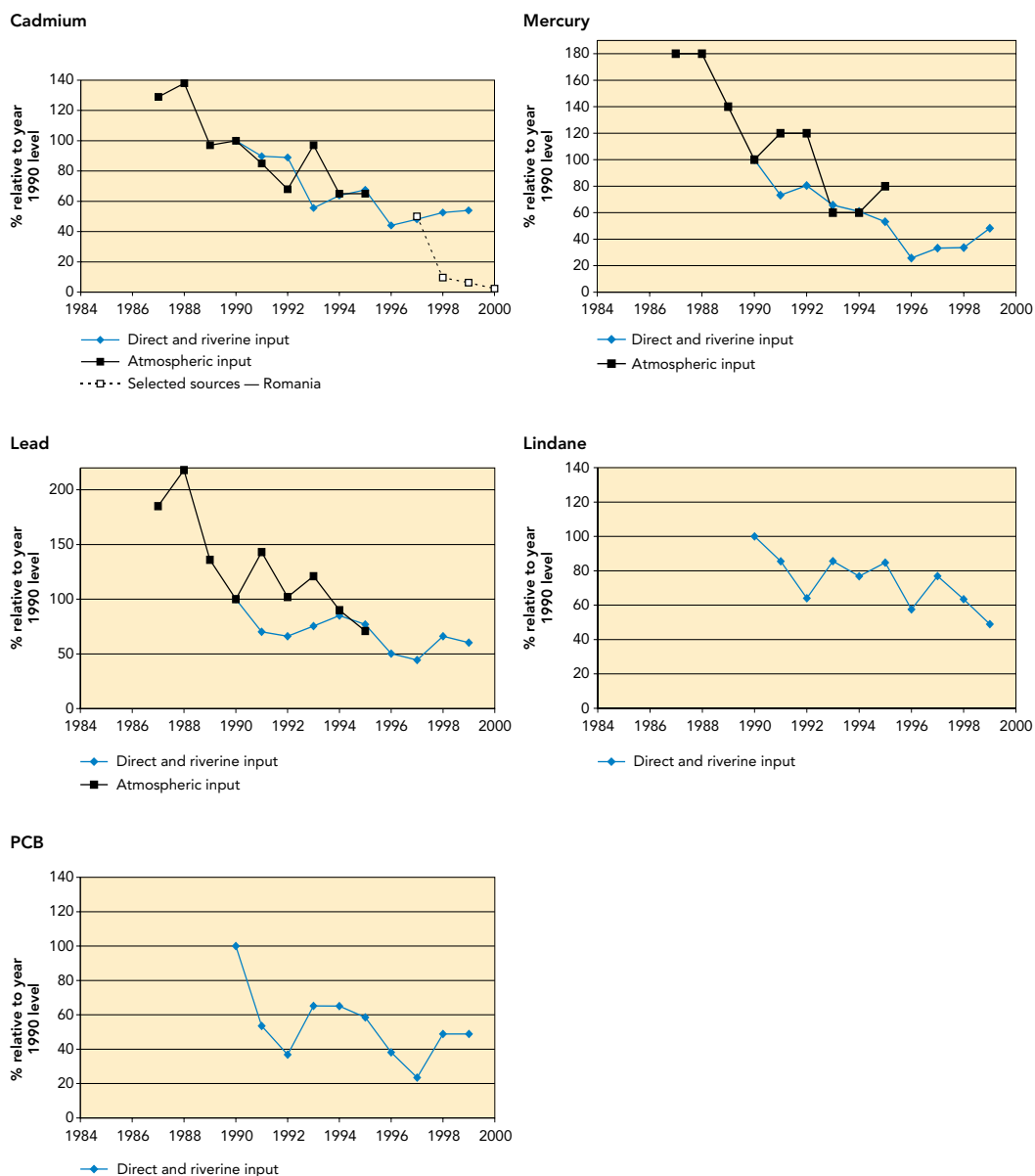


Table 4.1

Relative changes in direct and riverine inputs (DRI, as % of 1990) of cadmium, mercury, lead, lindane and PCB to the North-East Atlantic, and atmospheric inputs (AI) of cadmium, mercury and lead to the North Sea

Substance/Sea	Medium	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Cadmium																
NE Atlantic	DRI						100	90	89	56	64	68	44	48	53	54
North Sea	AI			129	138	97	100	85	68	97	65	65				
Mercury																
NE Atlantic	DRI						100	73	81	66	61	53	26	33	34	48
North Sea	AI			180	180	140	100	120	120	60	60	80				
Lead																
NE Atlantic	DRI						100	70	66	75	85	77	50	44	66	60
North Sea	AI			185	218	136	100	143	102	121	90	71				
Lindane																
NE Atlantic	DRI						100	86	64	86	77	85	58	77	64	49
PCB																
NE Atlantic	DRI						100	54	37	65	65	59	38	24	49	49

Country contributions to sum of direct and riverine inputs in tonnes per year of cadmium, mercury, lead, lindane and PCB to the North-East Atlantic

Table 4.2

Substance/ Year	B	DK	F	D	IRL	NL	NO	P	E	S	UK	SUM	%
Cadmium													
1990	4.30	0.57	4.70	8.54	3.39	10.30	16.21	9.00	n.a.	0.67	51.30	108.97	100.00
1991	3.90	n.a.	3.30	7.24	3.39	7.00	8.31	15.00	n.a.	0.34	49.35	97.83	89.80
1992	6.70	n.a.	4.50	10.55	2.31	5.70	7.90	24.00	n.a.	0.54	34.69	96.89	88.90
1993	3.42	n.a.	3.20	9.20	1.76	4.70	7.18	1.70	0.06	0.11	29.30	60.62	55.60
1994	3.44	n.a.	3.20	8.14	2.44	12.76	9.23	3.93	0.10	0.42	25.87	69.52	63.80
1995	3.81	n.a.	2.70	7.54	2.17	26.60	8.99	0.20	0.10	0.24	21.31	73.67	67.60
1996	2.90	n.a.	2.20	5.65	3.94	8.40	5.85	n.a.	n.a.	n.a.	19.11	48.05	44.10
1997	2.65	n.a.	n.a.	6.35	2.55	4.05	5.20	1.35	14.50	0.47	15.35	52.47	48.10
1998	2.45	n.a.	n.a.	6.25	3.45	9.70	7.75	0.60	6.80	0.56	19.75	57.31	52.60
1999	2.30	n.a.	n.a.	4.99	2.91	11.90	4.60	0.40	12.80	0.90	18.00	58.80	54.00
Mercury													
1990	4.05	0.13	5.70	10.54	2.45	3.18	0.85	4.90	n.a.	0.11	8.81	40.71	100.00
1991	1.15	n.a.	2.70	10.01	2.45	3.30	0.50	1.75	n.a.	0.13	7.85	29.83	73.30
1992	0.68	n.a.	8.70	11.03	2.40	3.26	0.53	0.04	n.a.	0.13	6.00	32.78	80.50
1993	2.62	n.a.	0.80	10.05	2.40	3.46	0.49	0.26	n.a.	0.11	6.55	26.73	65.70
1994	3.10	n.a.	1.10	5.65	2.40	6.41	0.38	0.85	n.a.	0.10	4.83	24.82	61.00
1995	0.60	n.a.	0.80	4.64	2.40	8.06	0.54	0.35	n.a.	0.15	4.13	21.67	53.20
1996	0.03	n.a.	0.50	2.94	n.a.	3.28	0.47	n.a.	n.a.	n.a.	3.26	10.48	25.70
1997	0.25	n.a.	n.a.	1.95	n.a.	2.70	0.40	1.15	2.60	0.09	4.41	13.54	33.30
1998	0.61	n.a.	n.a.	2.15	n.a.	2.15	2.86	0.75	0.40	0.13	4.68	13.72	33.70
1999	0.72	n.a.	n.a.	2.39	n.a.	2.33	1.46	0.56	8.60	0.20	3.31	19.60	48.10
Lead													
1990	27.5	7.2	150.0	212.8	63.4	346.5	120.9	680.0	n.a.	8.3	591.1	2207.6	100.0
1991	36.6	n.a.	150.0	297.5	63.4	230.0	102.8	80.0	7.7	5.3	575.6	1548.8	70.2
1992	32.5	n.a.	110.0	314.9	82.1	221.8	70.5	120.0	2.6	4.3	501.6	1460.2	66.1
1993	36.6	n.a.	100.0	378.1	55.5	393.8	58.5	7.3	0.9	3.2	631.4	1665.2	75.4
1994	48.2	n.a.	170.0	247.7	79.6	701.8	82.6	6.1	2.2	10.5	528.3	1877.1	85.0
1995	43.2	n.a.	55.0	185.5	63.3	871.8	82.5	2.8	1.3	8.2	384.8	1698.3	76.9
1996	44.5	n.a.	66.0	126.5	115.6	382.9	62.6	n.a.	n.a.	n.a.	311.5	1109.5	50.3
1997	40.0	n.a.	n.a.	151.5	120.5	233.0	63.7	43.0	17.0	8.7	303.3	980.7	44.4
1998	51.5	n.a.	n.a.	181.5	143.0	257.5	150.0	4.9	54.0	14.3	601.5	1458.2	66.1
1999	56.5	n.a.	n.a.	125.4	145.0	326.0	53.6	2.1	58.0	17.3	546.0	1330.0	60.2
Lindane													
1990	102.0	26.0	175.0	342.0	n.a.	16.7	536.0	n.a.	n.a.	n.a.	588.4	1786.1	100.0
1991	111.0	26.0	175.0	187.8	n.a.	7.2	261.3	n.a.	n.a.	n.a.	759.5	1527.8	85.5
1992	82.5	26.0	175.0	208.0	n.a.	10.5	97.9	0.7	n.a.	n.a.	543.0	1143.6	64.0
1993	80.0	26.0	175.0	191.5	n.a.	360.0	90.9	3.4	n.a.	n.a.	601.8	1528.6	85.6
1994	74.5	26.0	175.0	261.0	n.a.	230.0	87.7	15.0	n.a.	n.a.	501.8	1371.0	76.8
1995	55.5	26.0	175.0	291.4	n.a.	370.0	107.4	2.8	n.a.	n.a.	484.7	1512.8	84.7
1996	n.a.	n.a.	100.0	239.0	n.a.	300.0	75.9	n.a.	n.a.	n.a.	312.4	1027.3	57.5
1997	66.0	n.a.	n.a.	370.0	n.a.	300.0	78.7	n.a.	203.5	n.a.	354.5	1372.7	76.9
1998	110.5	n.a.	n.a.	261.0	n.a.	259.5	84.8	n.a.	15.0	n.a.	403.8	1134.6	63.5
1999	77.0	n.a.	n.a.	68.0	n.a.	279.0	9.4	n.a.	121.0	n.a.	324.0	877.0	49.1

Substance/ Year	B	DK	F	D	IRL	NL	NO	P	E	S	UK	SUM	%
PCB₇													
1990	30.5	30.3	100.0	143.5	n.a.	150.0	484.5	n.a.	n.a.	n.a.	2158.1	3096.8	100.0
1991	24.5	30.3	160.0	89.4	n.a.	130.0	38.0	n.a.	n.a.	n.a.	1183.5	1655.7	53.5
1992	26.5	30.3	130.0	43.6	n.a.	100.0	40.5	4.9	n.a.	n.a.	764.7	1140.5	36.8
1993	211.0	30.3	130.0	54.1	n.a.	130.0	21.6	25.0	n.a.	n.a.	1417.1	2019.0	65.2
1994	223.5	30.3	160.0	95.5	n.a.	300.0	55.0	84.0	n.a.	n.a.	1066.9	2015.2	65.1
1995	45.9	30.3	130.0	144.0	n.a.	470.0	27.8	26.0	n.a.	n.a.	938.7	1812.6	58.5
1996	n.a.	n.a.	100.0	127.5	n.a.	200.0	16.6	n.a.	n.a.	n.a.	739.6	1183.6	38.2
1997	66.0	n.a.	n.a.	109.0	n.a.	170.0	22.6	n.a.	n.a.	n.a.	360.0	727.6	23.5
1998	135.0	n.a.	n.a.	88.0	n.a.	181.0	22.4	n.a.	n.a.	n.a.	1089.4	1515.8	48.9
1999	164.0	n.a.	n.a.	123.0	n.a.	236.0	n.a.	n.a.	158.0	n.a.	835.0	1516.0	49.0

Notes: Belgium (B), Denmark (DK), France (F), Germany (D), Ireland (IRL), the Netherlands (NL), Norway (NO), Portugal (P), Spain (E), Sweden (S), United Kingdom (UK). n.a. indicates where data was not available. Rounded values from EEA Fact Sheet Y1R01HS01, zero or near zero (<1 kg) submissions assumed as data not available.

5. Spatial and temporal coverage of data

Monitoring is a prerequisite to assess the state of the environment. Hazardous substances concentrate in certain organisms and are widely distributed. Measuring concentrations in these organisms can be a useful assessment tool, provided there is adequate knowledge of how these data relate to driving forces and impacts in the ecological system (see Figure 1.1). The number and representativeness of measurements available determines the certainty of the statements in an assessment. Uncertainty surrounding these statements is related to distinguishing between natural and anthropogenic variations. This chapter provides an overview of the data material used in the present report, and assesses their spatial and temporal extent.

5.1. Riverine input and direct discharges

Many EU Member States have provided input data on hazardous substances into coastal waters for the period 1990–1999. For the North-East Atlantic, including the North Sea, annual data submitted to the OSPAR were the ‘best’ available but still incomplete. Irregular series and questionable data were evident. OSPAR input data on cadmium, mercury, lead, lindane and PCB were available for the period 1990–1999 but not from all countries. Input data for lindane and PCB were lacking from Ireland and Sweden (Table 5.1). Generally, annual high and low estimates of inputs were provided to OSPAR from Member States. Input calculations follow agreed OSPAR protocols. OSPAR (1998b) also published an integrated assessment of inputs in comparison to

biological contaminants and supplementary input data for 1997–1999.

For many countries of the North-East Atlantic the time series are short within this 1990–1999 period (e.g. one year for metals from Denmark, and one year for PCB₇ from Spain) or contain large gaps (e.g. 1997–1999 data metals were lacking for France). As mentioned, information was lacking altogether for some substances (e.g. lindane and PCB₇ from Ireland and Sweden) (see Table 4.2).

For the Baltic Sea, HELCOM (1998a) had only data on direct and riverine inputs for 1995. These were derived from the third Baltic Sea pollution load compilation but lack data on lindane. HELCOM data on cadmium, mercury, lead and PCB₇ were available for 1995. Further HELCOM heavy-metal data from Denmark, Estonia, Finland, Lithuania and Russia were not available in time for this report.

According to the recent report of EEA/UNEP (1999), no input data and time series are available for the Mediterranean Sea.

Only cadmium and lead input data from three sites along the Romanian coast for the years 1997–2000, provided by the Black Sea Commission, give some indication of the loads to the Black Sea.

Reliable data on inputs before 1990 were not available from OSPAR, HELCOM or EEA/UNEP. No data on inputs of DDT were available. Coordinates for the stations were

Number of years for which countries submitted data sets of direct and riverine inputs to OSPAR in the period 1990 to 1999 (Derived from Table 4.2)

Table 5.1

Substance/ Year	B	DK	F	D	IRL	NL	NO	P	E	S	UK
Cadmium	10	1	7	10	10	10	10	9	6	9	10
Mercury	10	1	7	10	6	10	10	9	3	9	10
Lead	10	1	7	10	10	10	10	9	8	9	10
Lindane	9	6	7	10	0	10	10	4	3	0	10
PCB ₇	9	6	7	10	0	10	9	4	1	0	10

Notes: Belgium (B), Denmark (DK), France (F), Germany (D), Ireland (IRL), the Netherlands (NL), Norway (NO), Portugal (P), Spain (E), Sweden (S), United Kingdom (UK).

not available from OSPAR to create accurate maps of inputs of cadmium, lead, lindane and PCB₇. Coordinates for inputs of mercury were only available for the Baltic Sea in 1995 (see Figure 6.10).

5.2. Atmospheric deposition

Time series on atmospheric inputs were available for this report only from OSPAR for the North Sea for the period 1987–1995 and only for three metals. Atmospheric inputs into the North Sea were estimated using Method III from the CAMP (comprehensive atmospheric monitoring programme) Principles (OSPAR, 2001). For mercury, the wet deposition was used (Method 2a). No recent data on atmospheric inputs to the North Sea are available. Information regarding the Baltic (HELCOM, 2002i) was not available in time for this report.

5.3. Concentrations in organisms

For assessment of concentrations in biota, data were collected from marine conventions and national reference centres (NRC). Most of the data were from OSPAR and HELCOM, supplied via the ICES data centre. OSPAR provided data for the North-East Atlantic and HELCOM provided data for the Baltic. France and Greece provided data on cadmium, mercury and lead for the Mediterranean Sea. France provided lindane data in mussels for the Mediterranean Sea. This contribution was generally balanced and consistent for an assessment of concentrations (1985–1999) (Table A.5). Romania provided data on cadmium, lead, DDT and lindane for the Black Sea.

The assessment of concentrations in biota (1986–1999) was based on these data. However, many time series were irregular and the contribution from countries in proportion to their respective coastlines was unbalanced. This was mainly because either investigations were not done or the supporting information to the data reported to ICES was insufficient (Tables A.1–A.6). For example, Iceland provided acceptable data from 1990 to 1992 and then again from 1996 to 1998 (Table A.1).

Data received from the Programme for the assessment and control of pollution in the Mediterranean region (MEDPOL) concerned Albania, Greece, Italy, Morocco, Spain, Turkey and Yugoslavia, but most of these data were from before 1991 and were

regarded as too old to represent current levels and trends. The later MEDPOL data concerned only two countries; Yugoslavia in 1991 and Albania in 1992. No new data have been received in 2001. The MEDPOL data have therefore been excluded from the statistical analysis. It was also apparent that MEDPOL member countries applied different guidelines for sampling which impaired comparison of data.

Data sets used for measuring concentrations of hazardous substances in mussels and fish were largely from the North-East Atlantic, and only a few from the Mediterranean and the Black Sea (Figure 5.1).

5.4. Validation of data on concentrations

Insufficient data were available on all substances to make a minimal assessment of levels and trends in sea water and sediments. However, DDT, lindane and PCB were generally not monitored in sea water because of their low water solubility.

For each data source (conventions, countries), the data have been manually inspected by the European Topic Centre on Water to clarify whether the reporting was consistent throughout the data file, regarding geographical information, measuring units, and observations below detection limits. Frequency distributions of each contaminant parameter have been inspected for each data source, with or without categorisation on locations or years, to see if there were suspicious values. These can be single values far outside the range of the other values, indicating an error. It can also be suspiciously high frequencies of a specific value in the low end of the range, or maybe of a few such values. This may indicate that the observations really were below the detection limit.

Occurrences of zero values have been checked, as possible indicators of missing observations (blank cells converted to zero by, for instance, unit conversion in Excel formulas) or inappropriately reported observations below detection limit. Time series plots for individual time series have been inspected for conspicuous outliers. This revealed a few series where the unit appears to have been changed by a factor of 1 000 at a certain time, without a corresponding change in the reported unit. Time series with suspicious data detected in this way have

been excluded from the analysis. Where appropriate, these issues were taken up with ICES or the relevant NRCs. In cases where it had not been possible to resolve such issues in time, data sets have not been included in further analyses.

Not all data received on concentrations in biota have been included in the assessment since more work on quality in the reporting

of these data needs to be done before they can be used in the assessments. The issues relate in particular to incomplete information about basis of measurements (wet/dry/lipid) and about observations below detection limit.

Details on needs for validation are given below for each country:

Sweden:

Additional data have been reported on cod for 1992, 1993 and 1999, and mussels for 1992–2000. The data includes a large number of records where observation was reported as below detection limit, but without specifying the detection limits. These should be established before the data can be included in the analysis. Also, overlap with data reported through ICES needs to be checked.

Denmark:

Supplementary data from 1985 on contaminants in mussel, but the reporting of detection limits and analysis quality was not sufficient. The frequency distribution of mercury shows a high number of the two lowest values (1 and 2 µg/kg (ppb)). This could indicate occurrence of observations below detection limit, but possibly also strong rounding of values. Data have been excluded from analysis until these issues have been clarified.

Latvia:

Data from 1998 and 1999, but of the contaminants included here, only for cadmium, and not for any of the animal species included in this analysis.

France:

Data were reported from the Mediterranean on cadmium, mercury, lead, zinc and lindane in mussels for the years 1985–1999. Data had no 'below detection limit' flags, but value frequency distributions for each substance did not indicate any large frequency of values that should have been reported as 'below detection limit', with a possible exception for lead. The dry-wet ratio was not specified but, since all values were reported on dry-weight basis, they can still be included in the statistical analysis.

United Kingdom:

Large number of parameters for the years 1993–95 and 1999. Values below detection limit were properly reported, but the basis of the measured values was not specified for any data. As this had been shown to vary over time and between stations in other data sets where it was reported, the data have been excluded from the analysis until detailed information about basis has been obtained. Better sample identification was also needed, for instance in order to calculate the required sum of PCB components. The data have uneven distribution over years and stations, limiting their use for estimating time trends.

Italy:

Data on metals in mussels for 1997–1999 were reported. The data lacked sufficient information about dry-wet basis and about values below detection limits. This information was provided on request, but arrived too late to be included in the statistical analysis for this report.

Greece:

Observations for biota, where the results were below detection limits, had been marked in the reported data, but the detection limit itself was not given, which means that a valid assessment of trends or levels may not be possible. For metals in mussels (*Mytilus galloprovincialis*), no cases of observations below detection limits were marked in the data, neither did the frequency distributions of these data indicate any problem in this respect. Until more information was available on detection limits, it was decided to include only the data on metals in mussels from the Greek data. Some apparent inconsistencies in station names have been corrected but confirmation by the NRC was not possible prior to running the analyses for this report. Some stations have been listed without coordinates, and were consequently not included in the maps. For 1997, there were some zero values for cadmium. Seen in the context of the data pattern for other contaminants, they appear to be missing values, and have consequently been deleted from the analysis. Most of the data were given on dry-weight basis, a few on fresh-weight basis, but in particular for the last two years, many data were given without information about the basis. The dry-wet ratio was not included in the data. Time trend estimates were based on only the dry-weight based data, as no sample-specific data for conversion were available. No effect of season was apparent in the data, so for the Greek data the Mann-Kendall time-series analysis was done on raw data instead of yearly averages. Current levels were estimated by station medians, using all data from 1995 and later. For this calculation, concentrations have been converted from dry- to wet-weight basis where necessary by a reported average wet:dry factor 6.7 according to specifications by the Greek NRC. The conversion factor was not quite the same as used in the maps where both dry-weight and wet-weight concentrations were classified. The values were reported as on the wet-weight concentrations. Data without specified basis were ignored in both spatial and temporal calculations.

5.5. Spatial coverage for concentrations in biota

Tables A.1–A.6 in the annex give the number of stations, by country and year, which were included in the indicator calculations (median and/or trend). Concentrations were calculated of the six hazardous substances in mussels (blue mussel — *Mytilus edulis*, Mediterranean mussel — *M. galloprovincialis*) and fish (Atlantic cod — *Gadus morhua*, herring — *Clupea harengus* or flounder — *Platichthys flesus*). Figure 5.1 gives an overview of the sampling locations.

Data on concentrations of **cadmium, mercury and lead** in the blue mussel in the North-East Atlantic were available for every country bordering this sea area except Denmark and Portugal, and only sparsely from France (Atlantic border) and the United Kingdom. Data on concentrations in the Mediterranean mussels were available for France and Greece. Data on mussels were available from 270 stations in the North-East Atlantic and 37 stations in the Mediterranean.

The data used on concentrations of **cadmium, mercury and lead** in cod, herring or flounder from the North-East Atlantic and the Mediterranean were from stations shown in Tables A.1–A.3. Cod data were from Belgium, the Netherlands, Norway, Poland, Sweden and the United Kingdom. Data for herring were from Finland, Poland and Sweden, and for flounder from Belgium, Denmark, France, Germany, the Netherlands and Sweden. Data on cod were available from 33 stations in the North-East Atlantic.

Data on concentrations of **DDT** in blue mussel in the North-East Atlantic were also available for nine countries (every country bordering this sea area except Denmark and Portugal). Data on DDT concentrations in cod from the North-East Atlantic were available from eight countries (Belgium, Denmark, Finland, Iceland, Norway, Poland, Sweden and United Kingdom). Data on concentrations in herring were available from five countries (Finland, Iceland, Poland, Sweden and the United Kingdom).

Sufficient data on **lindane** concentrations in blue mussel in the North-East Atlantic were not available and data on lindane concentrations in Mediterranean mussels were only available for France. No data for lindane in fish were available.

Data on concentrations of **PCB₇** in blue mussel in the North-East Atlantic were also available for nine countries (every country bordering this sea area except Denmark and Portugal). Data on PCB₇ concentrations in Atlantic cod from the North-East Atlantic were available from three countries (Belgium, Norway and Sweden). Data on concentrations in herring were available from three countries (Finland, Poland and Sweden); and data on concentrations in flounder from six countries (Belgium, France, Germany, the Netherlands, Norway and Sweden).

5.6. Temporal coverage for concentrations in biota

Temporal trend data for biota generally cover the period 1985–1999, but most series were far from complete (see Tables A.1–A.6). Many series cover only a small part of this period. For others, there were large gaps, up to nine years, of missing observations. In some areas, different stations were sampled in different years. The available data for each time series have been used for trend detection. The number of stations for each country and year varied from zero to 30 stations monitored annually. For the Mediterranean, the French data cover the period 1985–1999, while Greek data were from 1993–1999.

5.7. Data analysis

Part of the OSPAR data set contains high and low estimates of riverine input values as tonnes per year per region. The lower estimate in the database was calculated in such a way that any result below the detection limit was considered equal to zero. For the upper estimate, the value of the detection limit was used. For the indicator of all inputs the average was calculated of the high and low input values. Using these average values for all regions, the sum of direct and riverine input into the coastal waters of the North-East Atlantic in 1990 was calculated and set equal to 100 %. Next, yearly inputs and contributions to the inputs in the period 1990–1999 were expressed as percentages of the 1990 input. The same procedure was applied to atmospheric input. The results are shown in Figure 4.1 and Table 4.1.

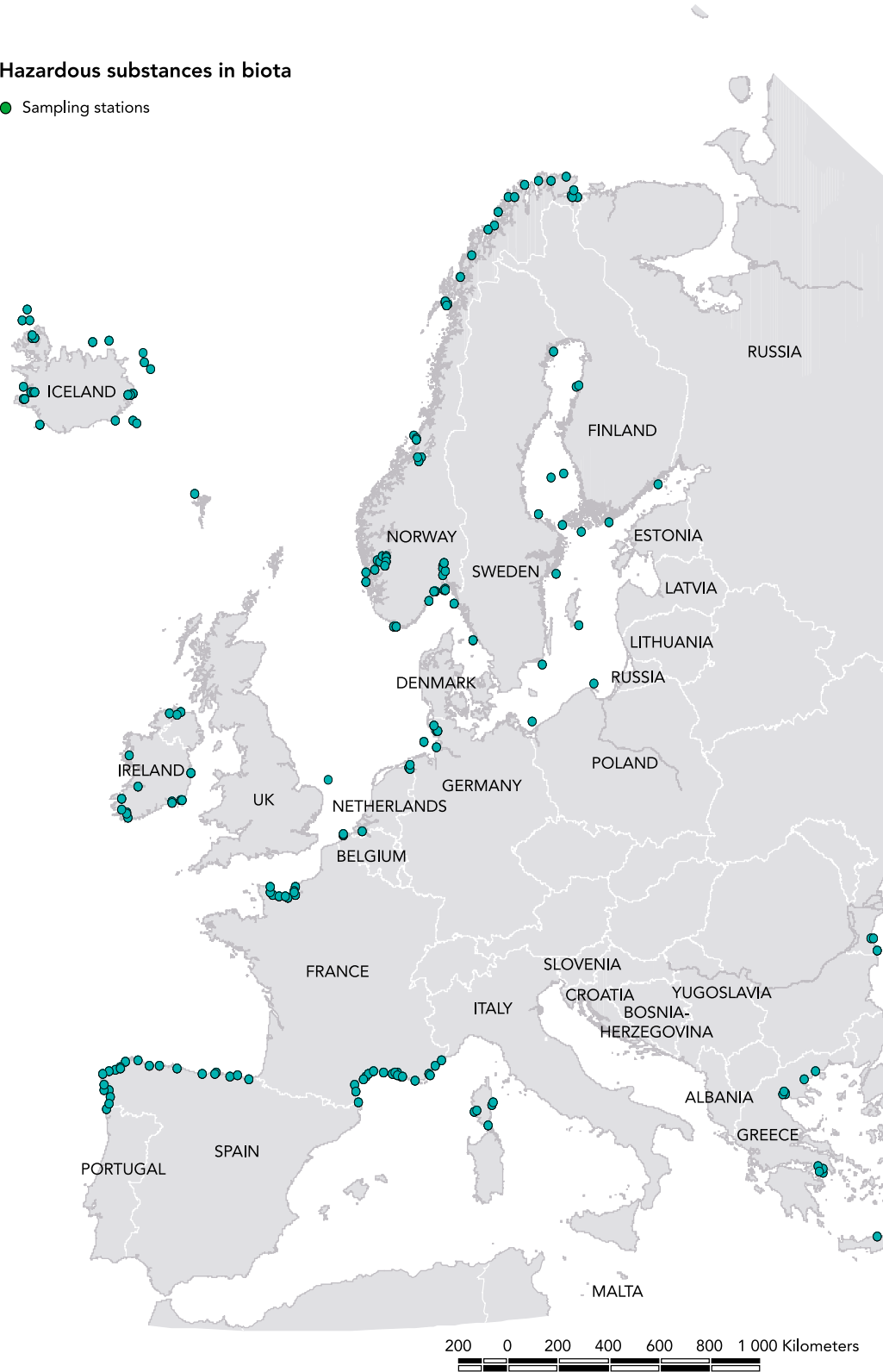
The apparent time trends may be influenced by changes in the monitoring programme or reporting practice over time, as the loads to some extent were calculated by summing the

Map showing location of stations included in indicator calculations (median and/or trend) of concentrations of cadmium, mercury, lead, DDT, lindane or PCB₇ in mussels (blue mussel — *Mytilus edulis*, Mediterranean mussel — *M. galloprovincialis*) and fish (Atlantic cod — *Gadus morhua*, herring — *Clupea harengus* or flounder — *Platichthys flesus*) 1985–1999

Figure 5.1

Hazardous substances in biota

● Sampling stations



reported values, not correcting for sources where a specific parameter had not been measured. Also, the use of average of high and low estimates means that any changes over time in detection limits will be reflected in the apparent trends. A further analysis of the load data should be done to investigate this.

Data for mussels and fish were reported on different bases (i.e., wet weight, dry weight, or lipid weight). These data could be converted to another and more appropriate basis — provided the relevant information was also available for the particular sample in question. Some of the variation in concentrations of hazardous substances can be reduced by normalising with respect to percent dry weight or percent lipid (fat) weight of the tissue measured. Concentrations were often submitted on a wet-weight basis but the water content in tissues can vary considerably. Hence, it was often appropriate to convert concentrations to a dry-weight basis, or to a lipid-weight basis, as in the case of lipophilic substances such as DDT, lindane and PCB. All ICES records were specified as dry, wet or lipid. Most data were on a wet-weight basis (124 117 records). Of these data, the dry-weight ratio was specified for 108 141 records. 33 438 records were on a lipid basis; of these data, lipid content was given for 25 883 records.

The ICES data usually contain both extracted lipid weight and total fat weight as percentages. For most of the records where both occur, they were quite similar, but there were a number of strong discrepancies, going both ways. In particular, records where total fat was much lower than extracted lipids should be considered suspect; there might have been problems with the fat determination. This problem in particular was important for data on flounder. It was necessary in some cases to convert concentrations from lipid-weight basis to dry-weight to harmonise values within time series for trend analysis. However, this was only done if extracted lipids and fat-weight agreed within 20 % when both were given, as it was not specified which of the fat determinations had been used as a basis for the fat-based concentration. If only one fat parameter had a value, that value was used to convert the concentration.

The available data were inspected, and were converted to an appropriate basis that gave the best coverage over stations and time. This

was considered separately for each species/tissue and parameter group. In the cases where the basis varied over time within a station, data that could not be converted to the chosen basis from sample-specific information were discarded in the statistical analysis. It was decided not to use a general conversion factor (e.g. 20 % wet weight for mussels) because this might introduce artificial time trends in many cases.

Some data sets were discarded because of insufficiently reported observations below the detection limit that might bias the estimates. In such cases, whole data sets were discarded and not just below detection limit values.

For all data for mussels and fish, observations reported as below a defined detection limit were handled as ranges (0-detection limit), giving low-high ranges also for sums over parameters and for median values for several observations.

Raw data were aggregated into median values of the available data for 1995–1999 in order to assess recent contaminant levels shown in the maps (e.g. Figure 6.2). Stations without any data after 1994 were not included in the maps. Medians were compared to ‘low’ and ‘high’ concentrations (see Table 6.1).

For some Swedish ICES data covering the period 1986–1999, duplicates or triplicates were found with identical sample identifiers and sample descriptors (fat content, tissue weight etc.) but different contaminant levels. Such record sets have been aggregated into ranges min-max, to account for the possibility that they represent replicate analyses on the same sample.

Local time trend analysis

Time series were identified by station code where available, otherwise by location coordinates. Because of this, some data that constitute parts of the same time series may have been separated into different data series in the analysis because coordinates may vary for a station.

To undertake a statistical analysis of time trends on a station-by-station basis, raw data for concentrations were aggregated into yearly median values within each time series (with a few exceptions). The stations have varying amounts of replicate samples per date, and varying numbers of dates per year. For each data series (species, tissue, location

and contaminant), simple median values per year were calculated. For the French Mediterranean data on *Mytilus galloprovincialis*, log-transformed concentrations were analysed by variance analysis, using Station*Year and Season (1–4) as factors. The Seasons 1–4 were: day 30–70, 120–170, 210–250 and 300–340.

Most of the samples were collected in February, May, August or September, respectively, but also from the succeeding months. Up to 1991, the sampling was more evenly distributed between all 12 months, but still concentrated within the four periods mentioned above. For all years, the sampling was distributed more or less equally over all four seasons. The extracted yearly values for the French Mediterranean data represent autumn levels. The yearly estimates from this analysis, with the effect of season removed, were used for trend analysis. For Greek data on metals in *M. galloprovincialis*, the trend tests were done using individual sample dates, as there were two to four sampling times a year, with seemingly little seasonal influence or serial correlation.

Monotone time trends were tested using Mann-Kendall statistics (ICES, 1999), modified by taking low-high ranges explicitly into account in counting concordant, discordant and tied observation pairs. Time trends were based on all available data from 1985 to 1999 for each time series. The time coverage was variable between series. However, some of the trends shown may be based on mainly older data. Significance of trends was based on a two-sided test with a nominal 5 % significance level, separately for each time series, without regard to serial correlation. Assessments of 'no trend' (i.e. no statistically significant trend) may be due both to actual lack of trend and to insufficient data.

Regional time trend analysis

The general changes in concentrations for each sea region (Figure 6.1) were based on the yearly aggregate values (averages) for each combination of location, year, species and tissue. Details of the calculation of these yearly averages have been given in the text above. The regional time trends have been extracted from these aggregates for each species, tissue and region. Only series with data for at least three years, and with data at least up to 1994 have been used. To diminish the effect on apparent time trends of changing geographical coverage between years, overall yearly average values were extracted by variance analysis (general linear model — GLM) with location and year as factors. The purpose of this was to separate the variation due to stations from the change over time, and to achieve yearly averages that were adjusted for differences in geographical coverage between years. Some apparent changes in time may still be due to changes in geographical coverage between years. The analyses were done using scaled values with the average value of each series, to get a representative average of relative variation over time, regardless of absolute levels at each station.

The indicators in Figure 6.1 show concentrations as a percentage of the regional average for the 1990. Figure 6.1 only shows the trends based on an upper estimate of the yearly average. The differences between lower limits and upper estimates were generally small or at least consistent through the time series. Some exceptions where relatively large differences were found were: cadmium in Atlantic cod in 1992–1993, and lead in blue mussels in 1991 and in Baltic herring in 1992. These deviations did not change the general picture of change in metal concentrations with time.

6. Present levels and trends (state) of hazardous substances

Concentrations of hazardous substances in blue mussels and fish constitute time integrating state indicators for coastal water quality. An advantage to using biota concentrations as indicators, as opposed to using water or sediment, is that they may be of direct ecological importance as well as having implications for human health and commercial interests in harvesting marine resources.

6.1. Regional time trend assessments

Figure 6.1 shows aggregated time trends of concentrations in mussels and fish for each region as averages of the relative variation in time over locations with sufficient data for the period 1984–1999. The charts give visual indications of the general development, without any formal assessment of statistical significance. The trends were averages over stations with very different temporal coverage, and also with very different time change patterns. No attempt to weight the stations according to representativeness was made, due to lack of information. However, a visual comparison with indicators in Figure 5.1 and the relevant maps shown in Figures 6.2–6.24 indicated that the indicators were reasonable summaries of the development over a majority of the locations included.

The aggregated results indicated a decreasing trend for cadmium, mercury, lead, DDT, lindane and PCB₇ in mussels and fish from both the North-East Atlantic and the Mediterranean Sea. Of the 178 (DDT) to 283 (cadmium) site specific time series analysed, for mussels 8–15 % showed significant trends, most of which were decreasing. Only 25 time series for lindane were measured. All of these concerned mussels from the Mediterranean and seven showed decreasing trends (Tables 6.2–6.24).

Due to the general lack of information from Denmark, France (Atlantic border), Portugal and United Kingdom (see chapter five), conclusions about the general levels and trends of the North-East Atlantic must be drawn with care.

Previous assessments of non-spatially aggregated data sets showed that most non-

aggregated and site-specific time series on concentrations in organisms in the coastal waters of the North-East Atlantic show non-significant trends. This was also found in this assessment. It should be emphasised that this may be due to lack of sufficient data. Even so, there might still be substantial long-term trends. Statistically significant trends were found in this study mainly at locations in estuaries and fjords, which were closer to the sources of pollution.

The effectiveness of using organisms as indicators for contaminant monitoring was hampered in part by limited spatial distribution for any given species. Furthermore, comparing results from different species was difficult mainly due to differences in bio-accumulation rates between species. The most comprehensive data sets available were for Atlantic cod (*Gadus morhua*).

OSPAR has addressed the difficult issue of coupling changes in concentrations found in biological indicators to changes in inputs, even when the study area is confined to a river mouth (OSPAR, 1998b, MON, 2001). Still greater precaution is needed when considering a wider geographical area.

6.2. 'Low' and 'high' concentrations of contaminants

Background concentrations

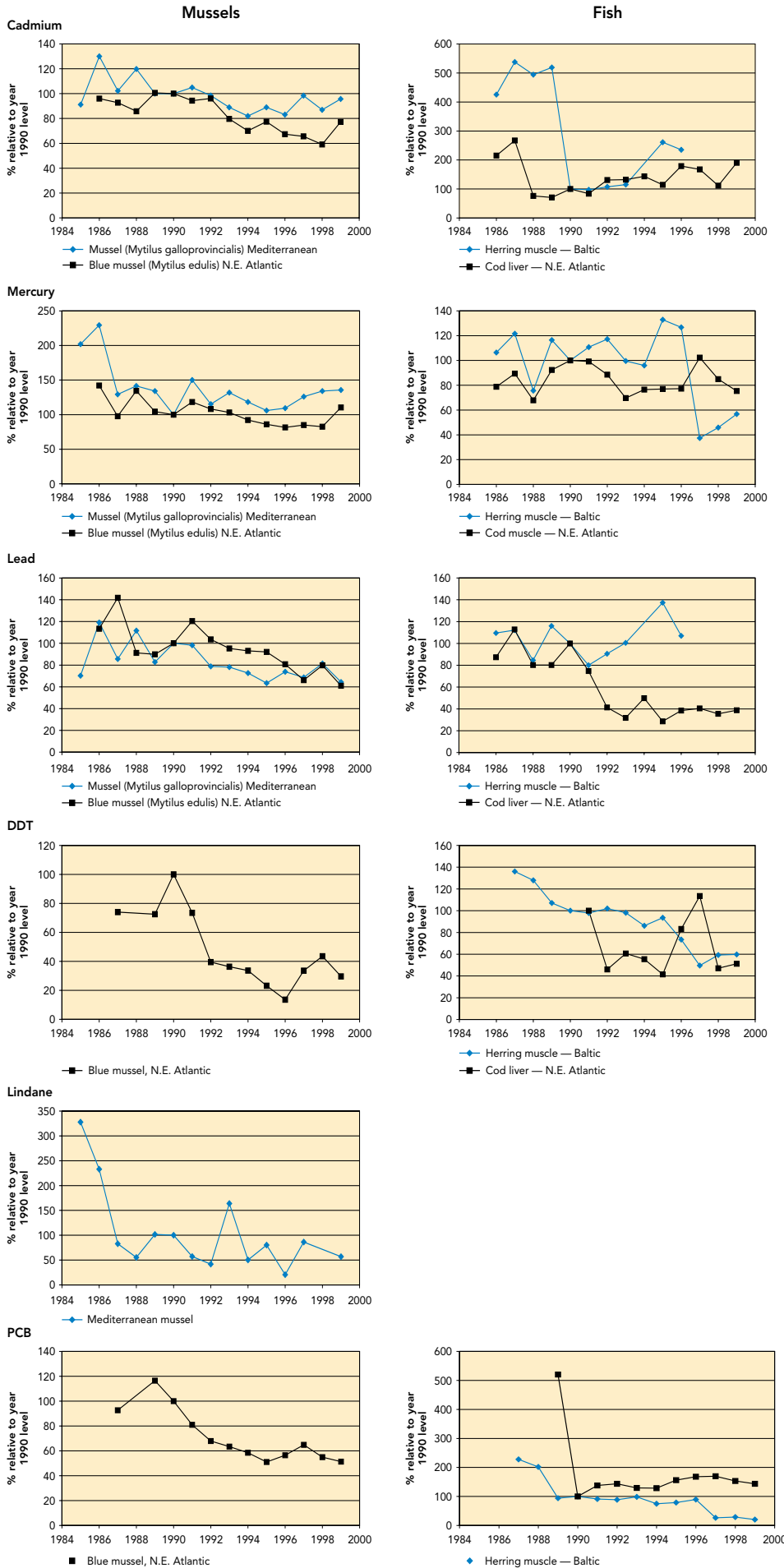
Cadmium, mercury and lead occur naturally in the environment, which means that they can be found in background concentrations in all media. Organic contaminants DDT, lindane and PCB are synthetic substances and do not occur naturally. Therefore, concentrations in the environment should be (close to) zero.

- **Cadmium** is naturally found in sea water at levels of 5–26 ng/L (OSPAR, 2000) and all marine organisms accumulate cadmium even in areas with no major local inputs. Suggested high background reference concentrations are 0.1 mg/kg for blue mussels (OSPAR, 1999) and 0.3 mg/kg wet weight for cod liver (Green and Knutzen, 2003).

Change (%) in cadmium, mercury, lead, DDT, lindane and PCB in blue mussels (*M. edulis*) in the North-East Atlantic, Mediterranean mussels (*M. galloprovincialis*) in the Mediterranean, Atlantic cod (*Gadus morhua*) in the North-East Atlantic and herring (*Clupea harengus*) in the Baltic 1985–1999

Figure 6.1

Source: OSPAR, HELCOM, EEA member countries.



- Natural concentrations of **mercury** in estuarine and coastal waters are typically around 20 ng/L. Background concentrations in mussels (whole soft part) and fish (cod muscle) are 0.01 (OSPAR, 1999) and 0.05 mg/kg wet weight, respectively (Green and Knutzen, 2003).
- The natural concentrations of **lead** in sea water vary considerably between coastal and oceanic, being more than 100 ng/L and 30 ng/L, respectively (OSPAR, 1996). Background concentrations in blue mussels and fish liver (Atlantic cod) are as low as 0.2 (OSPAR 1999) and 0.1 mg/kg wet weight (Green and Knutzen, 2003). Levels for human consumption are set at approximately 10 times that concentration.
- The pesticide **DDT** has been known as an environmental contaminant since the 1950s. Because of its physicochemical properties, it (or its derivatives) can now be found in nearly all compartments of the biosphere. Background concentrations of DDT are suggested to be 0.001 µg/g wet weight in blue mussels, 0.2 µg/g wet weight in cod liver (Green and Knutzen, 2003) and 0.05 µg/g wet weight in herring muscle (Knutzen, 1987).
- Although it is synthetic, **lindane** is found in sea water worldwide. The general background concentration is near 0.6 ng/L (Schreitmüller and Ballschmiter, 1995). In blue mussels, background concentrations of lindane are 0.01 µg/g wet weight (OSPAR, 1999).
- **PCB** are lipid soluble, have very low water solubility and are predominantly associated with particles in natural waters (SIME, 2001). Background concentrations of PCB are 0.002 µg/g wet weight in blue mussels, 0.1 µg/g wet weight in cod liver and 0.15 µg/g wet weight in herring muscle.

Classification of measured concentrations

To assess the concentration levels taken into account in this report, it was helpful to establish a 'low' and 'high' concentration to compare with (Table 6.1). No environmental quality standards are available yet under the water framework directive. Therefore, classification for this assessment had to be based on a variety of existing legislation, conventions and agreements. Limit values for concentration classes were found, in order of preference: in EU legislation, marine conventions, other international agreements, national assessments or as an arbitrary selection (10 times the 'low' value). With the

exception of the 'high' concentration designated by EU legislation, the limits have no legal application.

Natural background levels for the three metals have been used as the low concentration limit. For the three organic substances, background reference concentrations of low diffuse pollution levels in otherwise pristine areas have been used as the low concentration limit. Also used are OSPAR ecotoxicological assessment criteria, which are based on toxicity tests.

For 'high' concentrations, EU legislation was used where possible. EU legislation on foodstuffs provided 'high' values for cadmium, mercury and lead in mussels and fish (221/2002/EC). Otherwise, national limit values have been applied, or a factor of 10 above the low concentration.

Observed concentrations were classified into three classes in relation to these limit values: 1. below low limit, 2. between low and high, 3. above high.

On this basis, many of the current concentrations (median of 1995–1999) in the North-East Atlantic for cadmium (81 %), mercury (78 %) and lead (53 %) lie between 'low' and 'high'. This is unexpected considering that many stations are remote from known point sources and indicates that the OSPAR limits used are probably too low to be useful in designating problem areas. The background reference concentrations proposed by Green and Knutzen (2003) are 3–4 times higher (see Table 6.1) and might be more appropriate. The OSPAR limits for mercury and PCB in fish muscle may also be too high; 40–55 % of the stations have concentrations between 'low' and 'high'.

6.3. Local assessment of concentrations and time trends

The maps of European seas in Figures 6.2–6.24 present for each substance an:

1. overview of concentrations in mussels per sampling location,
2. overview of concentrations in fish per sampling location,
3. overview of time trend analysis in mussels per sampling location,
4. overview of time trend analysis in fish per sampling location.

Limit concentration used for classification in maps in figures 6.2–6.24 and tables 6.2–6.4

Table 6.1

Organism and tissue	Latin name	Low/High	mg/kg wet wt.	Reference	Comment
CADMIUM					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.1	OSPAR, 1999	Value rounded down from original BRC ² of 0.11 mg/kg wet weight; low compared to 0.4 mg/kg wet weight (Green and Knutzen, 2003)
Mussels	<i>Mytilus</i> sp.	High	1.0	EU, 2001	Foodstuffs limit for 'bivalve molluscs' Regulation (EC) No 466/2001
Atlantic cod, liver	<i>Gadus morhua</i>	Low	0.3	Green and Knutzen, 2003	Based on 90 percentile of 1184 individuals from reference areas
Atlantic cod, liver	<i>Gadus morhua</i>	High	0.5	TemaNord, 1994	Danish limit for 'fish liver'
Herring, muscle	<i>Clupea harengus</i>	Low	0.01	Knutzen, 1987	Upper limit to proposed range in background level
Herring, muscle	<i>Clupea harengus</i>	High	0.05	EU, 2002	Foodstuffs limit for fish muscle Regulation (EC) No 221/2002
MERCURY					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.01	OSPAR, 1999	BRC limit. A BRC of 0.04 ppm wet weight has been suggested (Green and Knutzen, 2003)
Mussels	<i>Mytilus</i> sp.	High	0.50	EU, 2001	Foodstuffs limit for 'bivalve molluscs' Regulation (EC) No 466/2001
Atlantic cod, muscle	<i>Gadus morhua</i>	Low	0.05	OSPAR, 1999	BRC limit. A BRC of 0.1 ppm wet weight has been suggested (Green and Knutzen, 2003)
Atlantic cod, muscle	<i>Gadus morhua</i>	High	0.50	EU, 2001	Foodstuffs limit for fish muscle Regulation (EC) No 466/2001
Herring, muscle	<i>Clupea harengus</i>	Low	0.05	Knutzen, 1987	Upper limit to proposed range in background level
Herring, muscle	<i>Clupea harengus</i>	High	0.50	EU, 2001	Foodstuffs limit for fish muscle Regulation (EC) No 466/2001
LEAD					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.2	OSPAR, 1999	Value rounded down from original BRC ² of 0.19 ppm w.w. A BRC of 0.6 ppm wet weight has been suggested (Green and Knutzen, 2003)
Mussels	<i>Mytilus</i> sp.	High	1.5	EU, 2002	Foodstuffs limit for 'bivalve molluscs' Regulation (EC) No 221/2002
Atlantic cod, liver	<i>Gadus morhua</i>	Low	0.1	Green and Knutzen, 2003	Based on 90 percentile of 1182 fish from reference areas
Atlantic cod, liver	<i>Gadus morhua</i>	High	1.0		Taken as 10 times 'low' value
Herring, muscle	<i>Clupea harengus</i>	Low	0.01	Knutzen, 1987	Upper limit to proposed range in background level
Herring, muscle	<i>Clupea harengus</i>	High	0.20	EU, 2002	Foodstuffs limit for fish muscle Regulation (EC) No 221/2002
DDT					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.01	OSPAR, 1999	EAC ³ limit for DDE. A BRC ² of 0.001 ppm wet weight has been suggested (Green and Knutzen, 2003)
Mussels	<i>Mytilus</i> sp.	High	0.10		Taken as 10 times 'low' value
Atlantic cod, liver	<i>Gadus morhua</i>	Low	0.5	OSPAR, 1999	EAC limit DDE. A BRC of 0.2 ppm wet weight has been suggested (Green and Knutzen, 2003)
Atlantic cod, liver	<i>Gadus morhua</i>	High	5.0		Taken as 10 times 'low' value
Herring, muscle	<i>Clupea harengus</i>	Low	0.05	Knutzen, 1987	Upper limit to proposed range in background level
Herring, muscle	<i>Clupea harengus</i>	High	0.50		Taken as 10 times low

Organism and tissue	Latin name	Low/High	mg/kg wet wt.	Reference	Comment
LINDANE					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.01	OSPAR, 1999	EAC limit. A BRC of 0.001 ppm wet weight has been suggested (Green and Knutzen, 2003)
Mussels	<i>Mytilus</i> sp.	High	0.10		Taken as 10 times 'low' value
PCB,					
Mussels	<i>Mytilus</i> sp. ¹	Low	0.002	OSPAR, 1999	Value rounded up from original BRC of 0.0017 wet weight
Mussels	<i>Mytilus</i> sp.	High	0.020		Taken as 10 times 'low' value
Atlantic cod, liver	<i>Gadus morhua</i>	Low	0.10	OSPAR, 1999	EAC limit. A BRC of 0.5 ppm wet weight has been suggested (Green and Knutzen, 2003)
Atlantic cod, liver	<i>Gadus morhua</i>	High	2.5	PNUN, 1987	Swedish PCB [total] limit for 'fish liver' times 0.5
Herring, muscle	<i>Clupea harengus</i>	Low	0.15	Knutzen, 1987	Upper limit to proposed range in background level of PCB [total] times 0.75
Herring, muscle	<i>Clupea harengus</i>	High	1.5		Taken as 10 times 'low' value

¹⁾ Blue mussel (*Mytilus edulis*) for the North-East Atlantic, Mediterranean mussel (*M. galloprovincialis*) for the Mediterranean

²⁾ Background reference concentration

³⁾ Ecotoxicological assessment criteria

Notes: Low/high concentration limits for spatial assessment of cadmium, mercury, lead, DDT, lindane and PCB. EU foodstuff limits are highlighted in grey shade. With the exception of EU legislation, the limits have no legal application.

For mercury, inputs into the Baltic Sea are also given in Figure 6.10.

6.3.1. Cadmium

The regional assessments of time trend indicated a decreasing trend for cadmium in mussels from both the North-East Atlantic and the Mediterranean Sea (Figure 6.1). No general trends were evident for concentrations in cod liver or herring muscle.

The station-by-station overview of 1995–1999 (2001 for Black Sea) levels of cadmium for mussels (both *Mytilus edulis* and *M. galloprovincialis*) indicated that concentrations were elevated in the estuaries for large rivers (e.g. the Seine, northern France), in areas with point discharges (e.g. Sør fjord, western Norway) and in some harbours (Table 6.2, Figure 6.2). In addition, blue mussels in some areas, e.g. along the Norwegian north coast, appeared to be affected by local geology (natural high background concentrations).

The only area from which mussels did not appear to be suitable for human consumption was Sør fjord, Norway and at the locations shown along the Romanian coast in the Black Sea. The high level of cadmium in

mussels from the Sør fjord was largely due to industrial activity (a zinc smelter) in Sør fjord. Although not necessarily representative for the remainder of European coasts, levels elsewhere appeared to be intermediate. Concentrations in some of these areas, e.g. along the Norwegian North coast, appeared to be affected by higher local background, possibly due to natural variations in mineralogy.

In fish, the 1995–1999 levels never exceeded suggested 'high' concentrations (Figure 6.3). The data show that cadmium levels were generally low. One area in northern Iceland was an exception, possibly due to inputs from local geology.

Of the 283 temporal trends statistically analysed on a station-by-station basis (197 for mussels, 86 for fish) only 27 were significant, 20 down and seven up (Table 6.2, maps in Figures 6.4 and 6.5). For cadmium, inputs were decreasing (OSPAR, 2000) but the regional trend for mussels and fish (see Figure 6.1) did not indicate a clear trend, although a station-by-station analysis revealed a certain dominance of decreasing trends over increasing trends. Levels in some contaminated areas, e.g. the Seine estuary and Sør fjord, appeared to be decreasing.

Subindicator summary assessment for cadmium in mussels and fish from the North-East Atlantic, Baltic Sea, Mediterranean Sea and Black Sea

Table 6.2

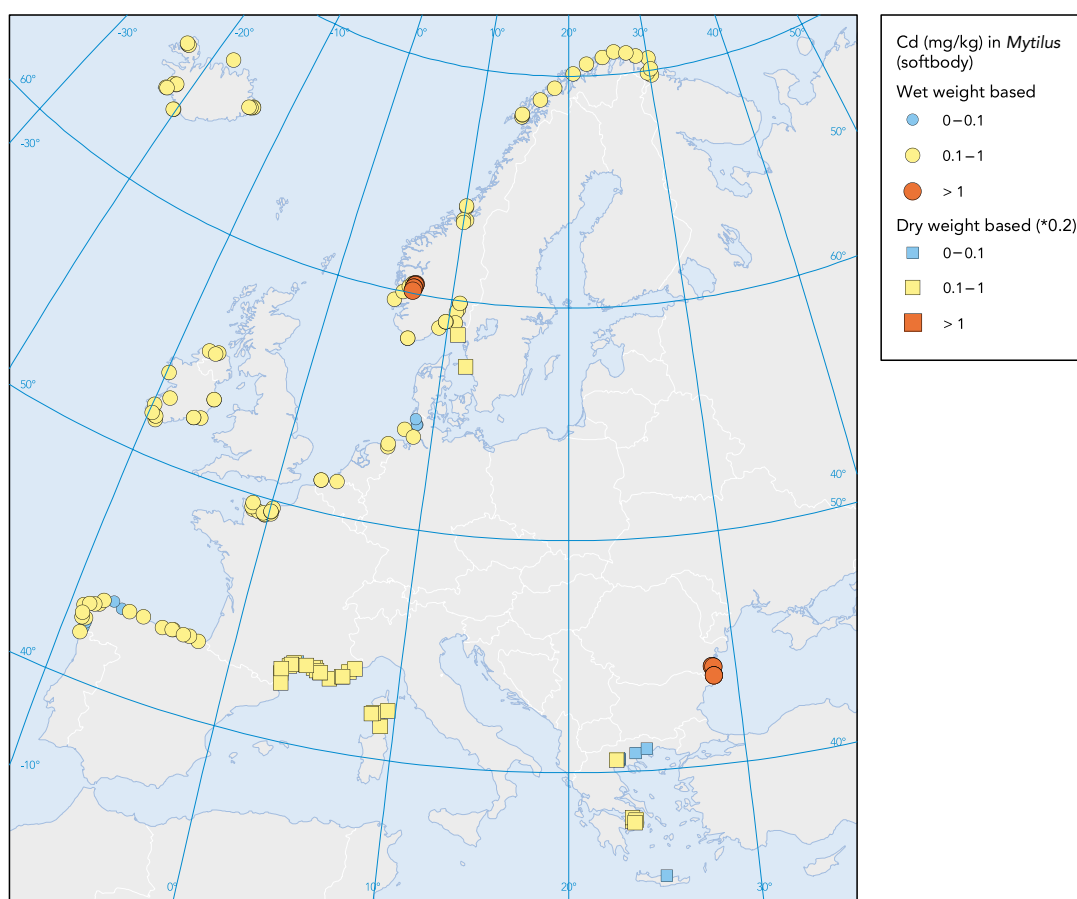
Sea region	Sub-indicator	Spatial variation			Time trends		
		Total no. of stations	Number over low	Number over high	Total no. of stations	Number down	Number up
North-East Atlantic	Mussels	119	102	6 ¹	160	10	3
	Fish	25	2	0	59	2	1
Baltic Sea	Mussels	1	1	0	1	0	0
	Fish	10	0	0	30	2	0
Mediterranean Sea	Mussels	36	26	0	36	6	3
	Fish	0			0		
Black Sea	Mussels	3	3	3	0		
	Fish	0			0		

1) Concerns only Sør fjord in western Norway

Notes: See Table 6.1 for selection of indicator species and low/high concentrations limits. See also results in maps in Figures 6.2–6.4.

Cadmium (Cd) in mussels (*Mytilus edulis* — North-East Atlantic; *M. galloprovincialis* — Mediterranean and Black Sea), median mg/kg wet weight for 1995–1999 (2001 for Black Sea).

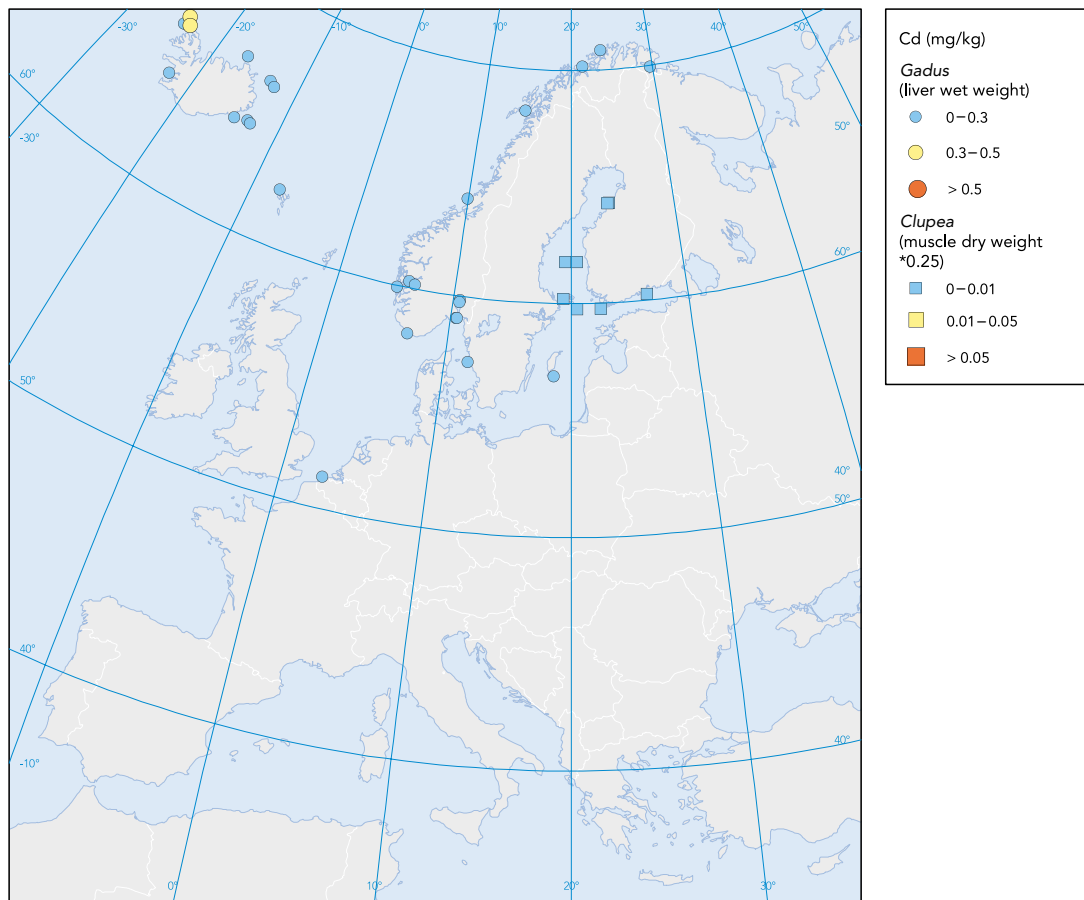
Figure 6.2



Notes: See Table 6.1 for basis of classification. EU legislation limit for cadmium in foodstuffs ‘bivalve molluscs’ is 1 mg/kg wet weight (EU, 2001b). Based on data from OSPAR and EEA member countries (Mediterranean), and data reported by Romania, see also Table 4.1. NB: larger symbols may obscure other symbols.

Figure 6.3

Cadmium (Cd) in liver of cod (*Gadus morhua*) and muscle of herring (*Clupea harrengus*), median mg/kg wet weight for 1995–1999



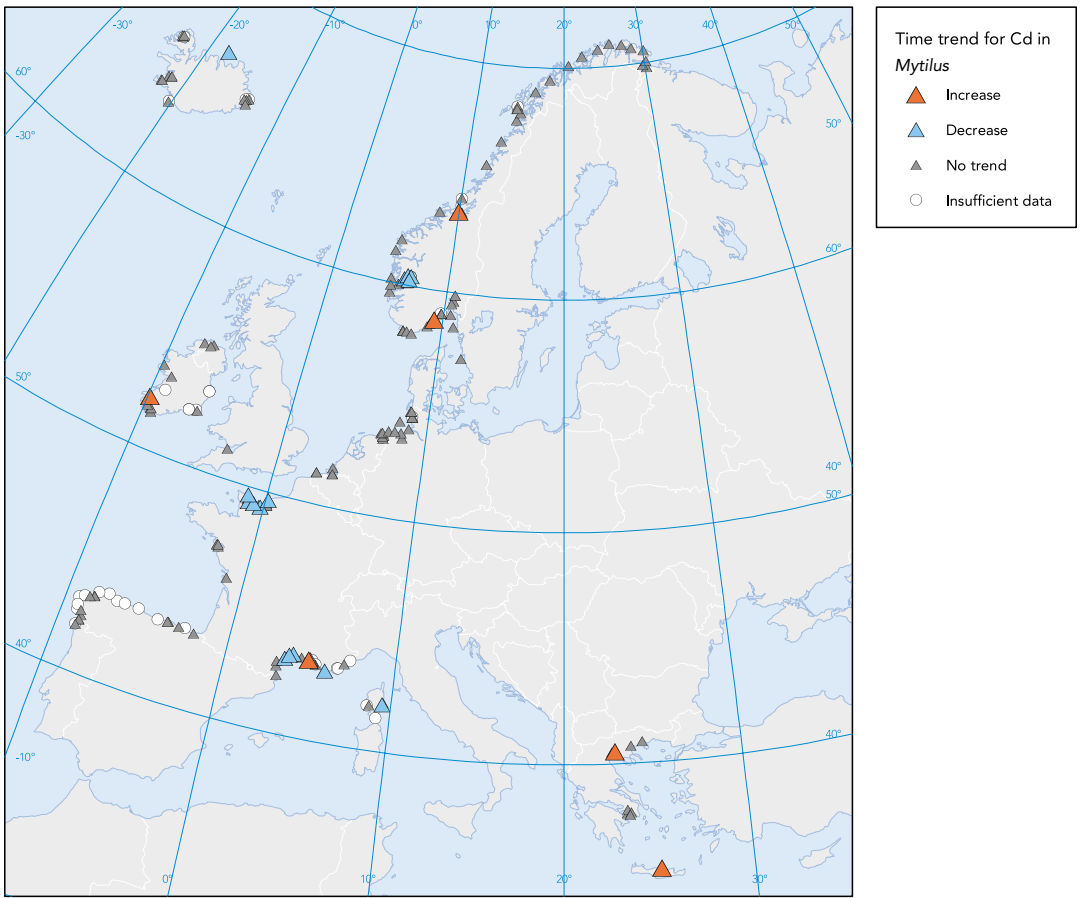
Notes: See Table 6.1 for basis of classification. EU legislation limit for cadmium in foodstuff muscle of fish is 0.05 mg/kg wet weight (EU, 2001b). Based on data from OSPAR and HELCOM, see also Table A.1. NB: larger symbols may obscure other symbols.

There also appeared to be a general decreasing trend of cadmium in mussels from the French Mediterranean coast. One exception was the Marseilles area, possibly with influence from the river Rhône, at some stations of the Aegean Sea. Increasing levels elsewhere in Europe appeared to be associated with harbour areas. In general, cadmium levels in fish from the Skagerrak (the area between Norway, Sweden and Denmark) appeared to decrease. The results from the Rhine estuary (Belgian-Dutch border) and the inner Oslofjord illustrated that stations in close proximity may have conflicting results (see Figures 6.4 and 6.5).

Previous assessments of non-spatially aggregated data sets on metals indicate that most non-aggregated and site specific time series on cadmium concentrations in organisms in the coastal waters of the North-East Atlantic show non-significant trends. This was also found in this assessment. It should be emphasised that this may be due to lack of sufficient data. There might still be substantial long-term trends. Statistically significant trends were found mainly at locations in estuaries and fjords, which were closer to the sources of land-based diffuse pollution.

Cadmium (Cd) time trend in blue mussel (*Mytilus edulis*) in the North-East Atlantic and the Mediterranean mussel (*M. galloprovincialis*) 1985–1999. (see Table 6.2)

Figure 6.4



Cadmium (Cd) time trend in cod liver, herring liver/muscle, flounder liver, 1986–1999. (see Table 6.2)

Figure 6.5

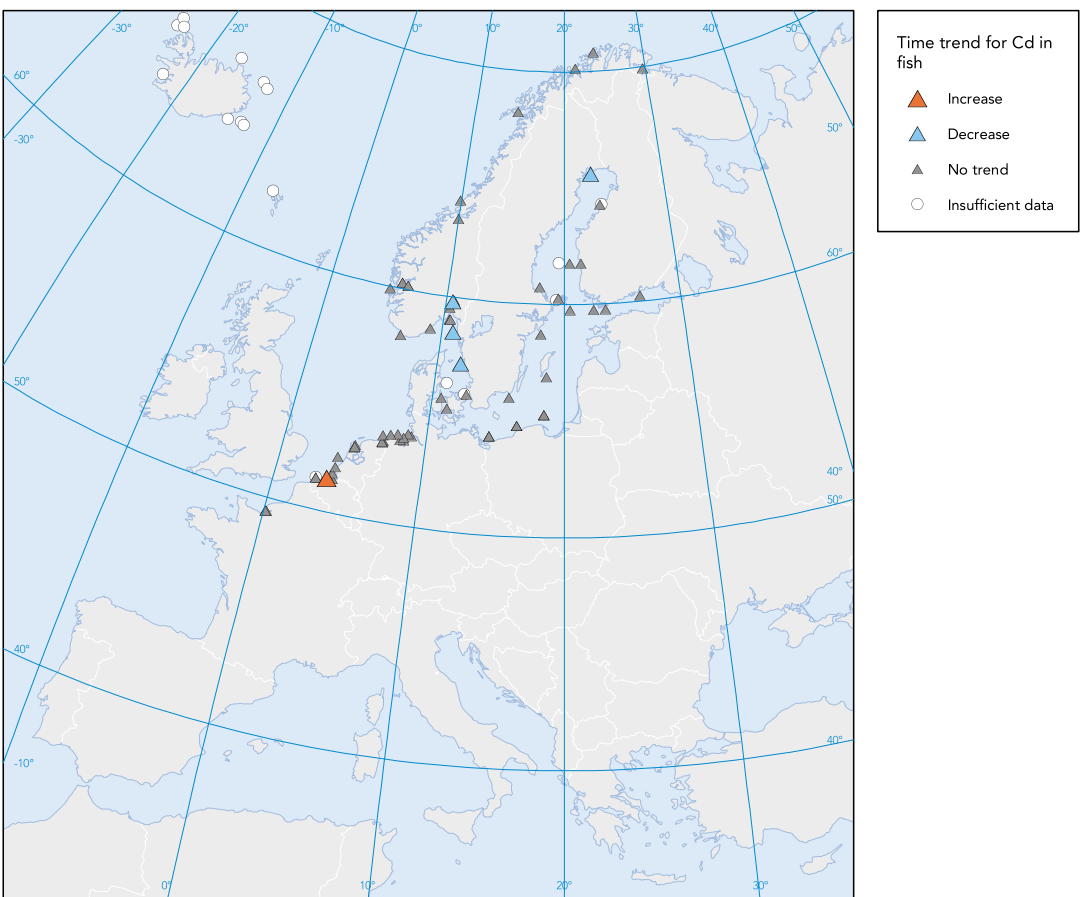


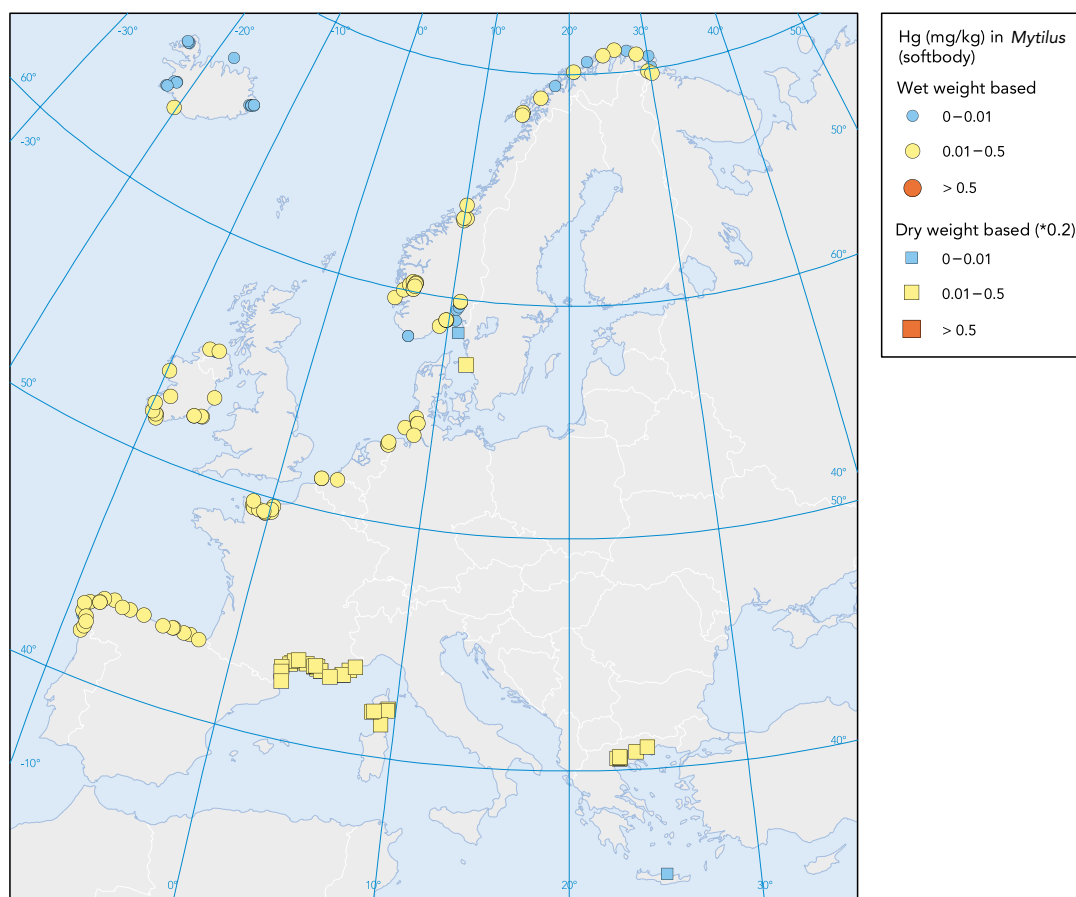
Table 6.3

Subindicator summary assessment for mercury in mussels and fish from the North-East Atlantic, Baltic Sea, Mediterranean Sea and Black Sea

Sea region	Subindicator	Spatial variation			Time trends		
		Total no. of stations	Number over low	Number over high	Total no. of stations	Number down	Number up
North-East Atlantic	Mussels	118	92	0	156	4	2
	Fish	28	12	0	63	3	0
Baltic Sea	Mussels	1	1	0	1	0	1
	Fish	16	4	0	26	2	1
Mediterranean Sea	Mussels	32	31	0	32	7	0
	Fish	0			0		
Black Sea	Mussels	0			0		
	Fish	0			0		

Notes: See Table 6.1 for selection of indicator species and low/high concentrations limits. See also results in maps in Figures 6.6–6.10.

Figure 6.6

Mercury (Hg) in mussels (*Mytilus edulis* — North-East Atlantic; *M. galloprovincialis* — Mediterranean), median mg/kg wet weight for 1995–1999

Notes: See Table 6.1 for basis of classification. EU legislation limit for mercury in foodstuffs 'bivalve molluscs' is 0.5 mg/kg wet weight (EU, 2001b). Based on data from OSPAR and EEA member countries (Mediterranean), see also Table A.2. NB: larger symbols may obscure other symbols.

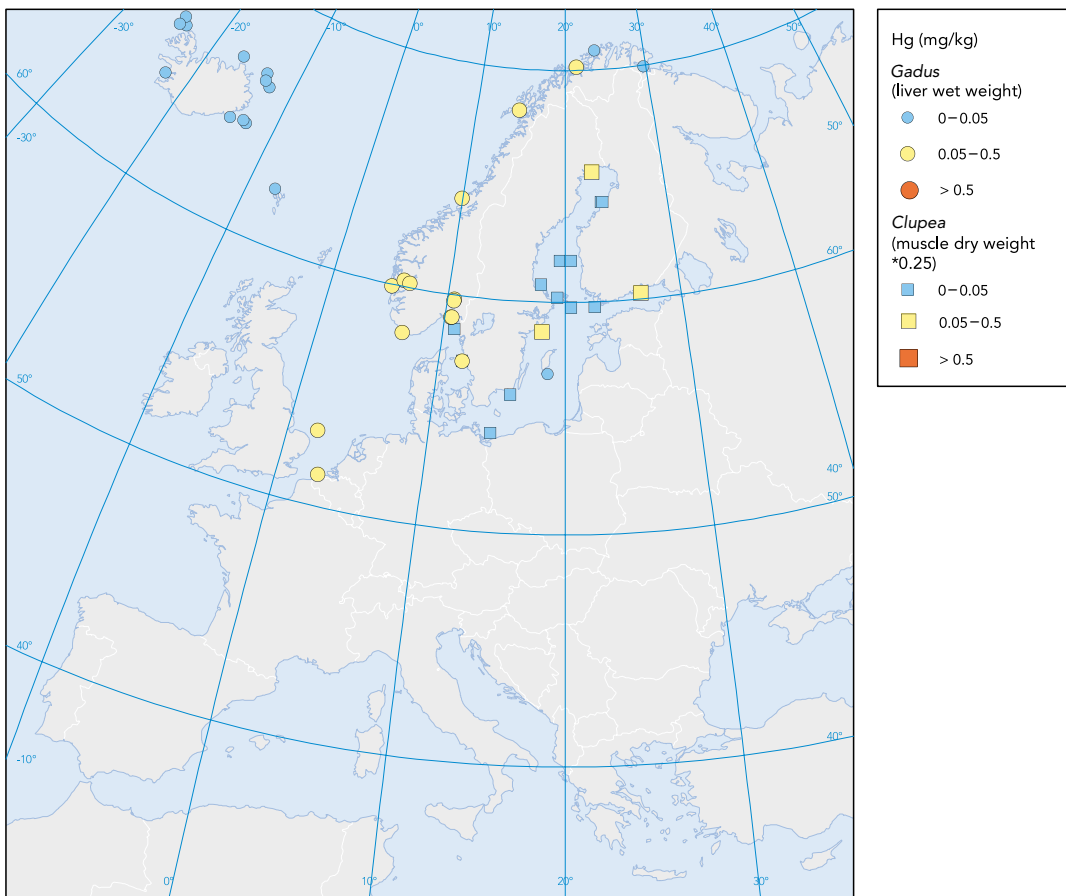
6.3.2. Mercury

The regional assessments of time trend indicated a decreasing trend for mercury in mussels from both the North-East Atlantic and the Mediterranean Sea. No general trend was evident for concentrations in cod or herring muscle (Figure 6.1).

On a station-by-station basis, the 1995–1999 levels of mercury in blue mussels in Europe were somewhat over background levels in most areas (Table 6.3, Figure 6.6). There were more locations with concentrations at background levels in remote areas than in the most industrialised regions (e.g. Iceland

Mercury (Hg) in muscle of cod (*Gadus morhua*) and muscle of herring (*Clupea harrengus*), median mg/kg wet weight for 1995–1999

Figure 6.7



Notes: See Table 6.1 for basis of classification. EU legislation limit for mercury in foodstuff muscle of fish is 0.5 mg/kg wet weight (EU, 2001b). Based on data from OSPAR and HELCOM, see also Table A.2. NB: larger symbols may obscure other symbols.

and northern Norway). The general picture indicates a diffuse exposure of coastal mussel populations to mercury, presumably mainly atmospheric, but with no real hot spots. The results must be interpreted with care, however, since not all European coastal areas were included in the survey.

The concentrations of mercury in Atlantic cod muscle (Figure 6.7) showed a similar pattern as blue mussel. On a station-by-station basis, concentrations were at background levels in cod that were collected in areas remote from known point sources. Although Atlantic cod do not store much lipid in the muscle, most of the mercury accumulates in this tissue. The other fish species monitored, Baltic herring, accumulated little mercury in muscle. Levels of mercury in the Baltic herring appeared to be low (including the comparison with cod data in one location).

Of the 278 temporal trends analysed on a station-by-station basis (189 for mussels, and

89 for fish) only 20 were significant, 16 down and four up (Table 6.3, maps in Figures 6.8 and 6.9). For mercury, inputs were decreasing (OSPAR, 2000), but no clear regional trend was registered using mussel and fish indicators (see Figure 6.1). There was some dominance of decreasing trends over increasing trends, but most of the stations show no significant trend. In mussels in most areas, notably in the Mediterranean, there were decreases in the mercury concentrations. It was not clear why there were increasing trends of mercury at Gothenburg and at the outlet of the river Ems. As for blue mussels, there were decreasing levels of mercury in fish collected at various sites, including the German Bight. The only area with an increase was at the estuary of the river Oder, near Stettin, which empties into the Baltic Sea.

Over the last centuries, mercury has been used in a wide range of human applications (chlor-alkali industry, manufacturing, dentistry, metallurgy, electronic household appliances). Both the retrieval and use of the

Figure 6.8 Mercury (Hg) time trend in blue mussel (*Mytilus edulis*) in the North-East Atlantic and the Mediterranean mussel (*M. galloprovincialis*) 1985–1999 (see Table 6.3)

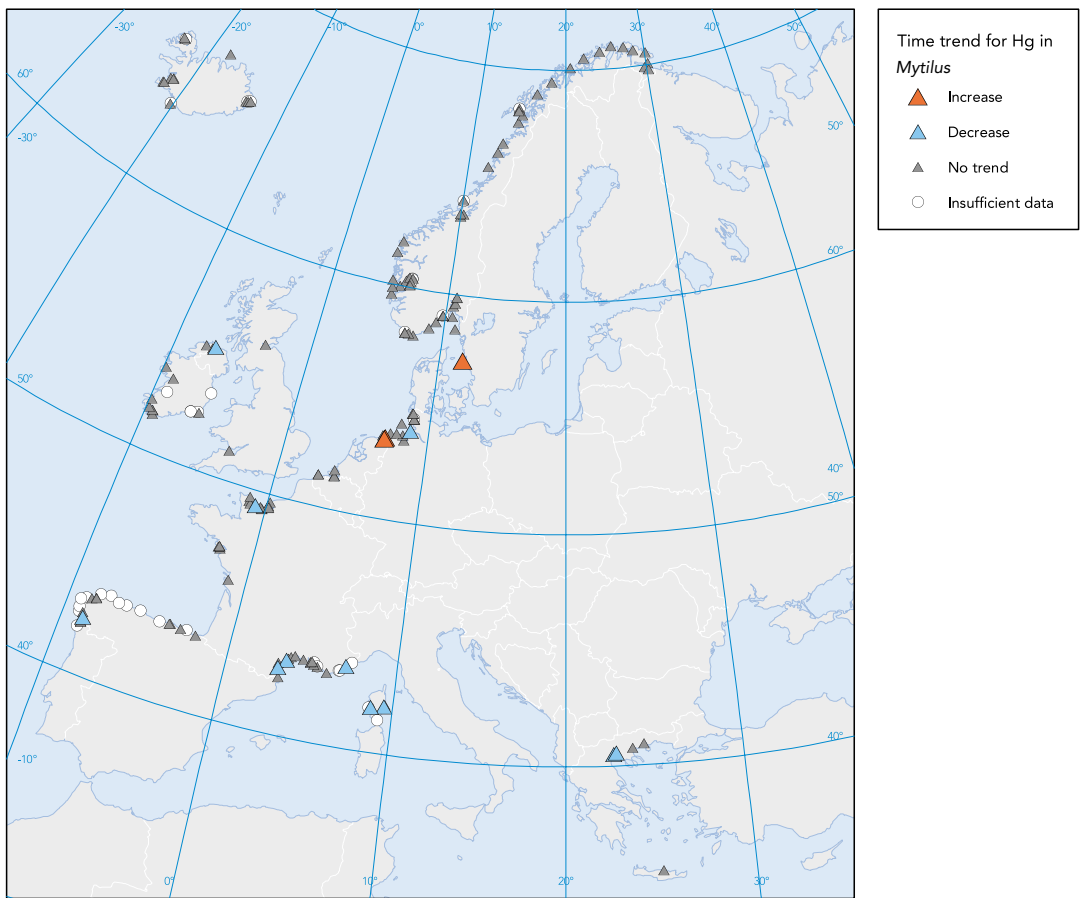
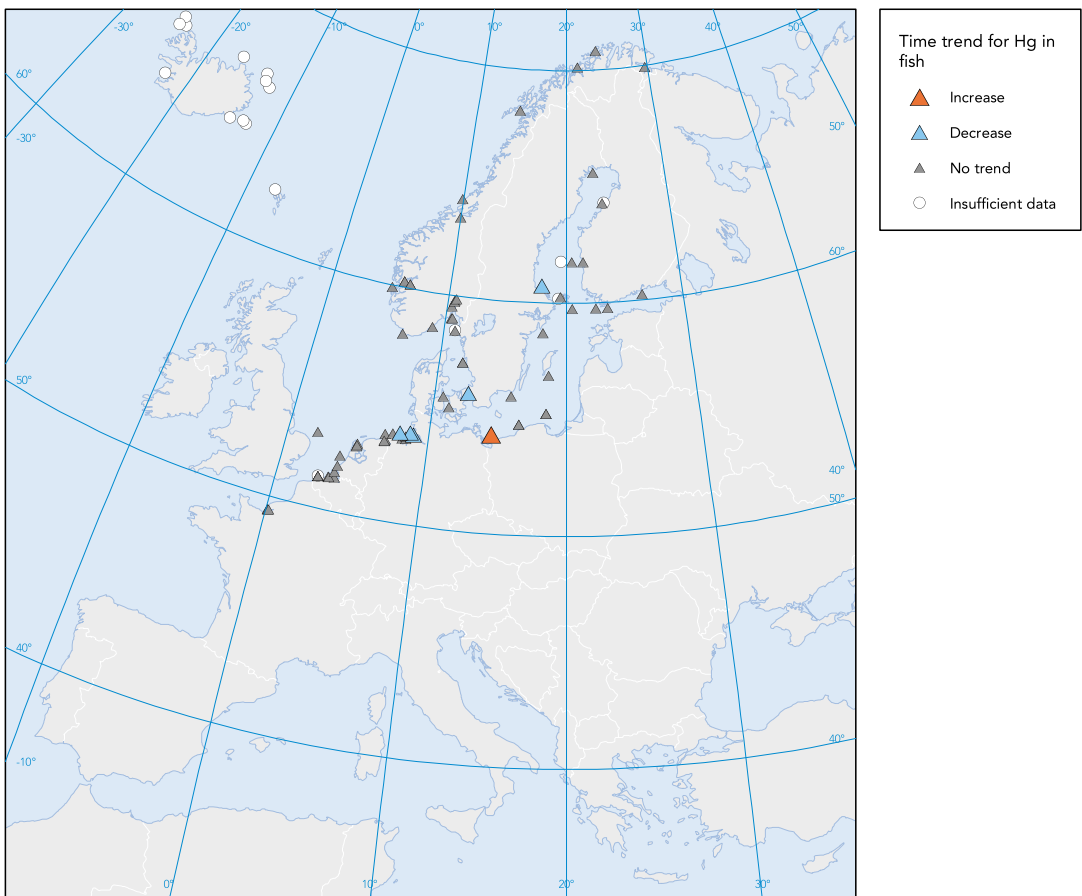
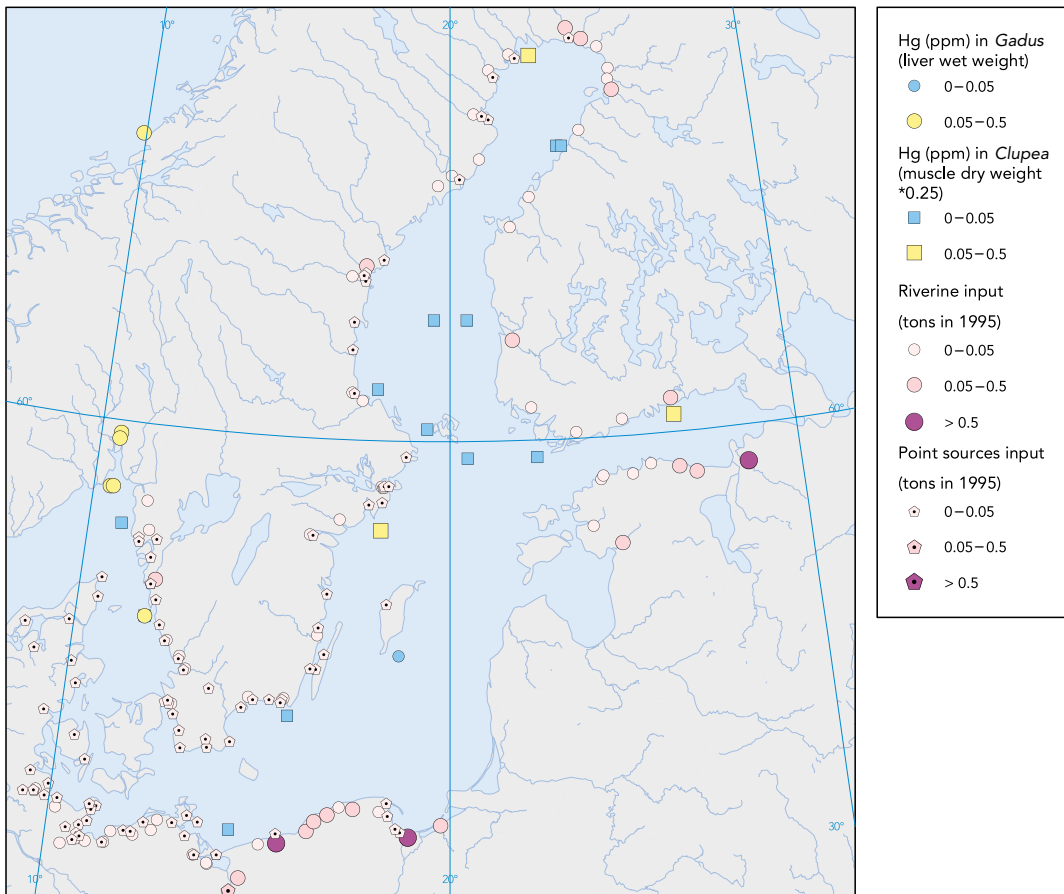


Figure 6.9 Mercury (Hg) time trend in muscle of cod, herring and flounder, 1986–1999 (see Table 6.3)



Mercury (Hg) input to the Baltic Sea, 1995 and mercury (Hg) concentrations in muscle of cod (*Gadus morhua*) and muscle of herring (*Clupea harengus*), median mg/kg wet weight for 1995–1999

Figure 6.10



Notes: See Table 6.1 for basis of classification. EU legislation limit for mercury in foodstuff muscle of fish is 0.5 mg/kg wet weight (EU, 2001b). Based on data from OSPAR and HELCOM, see also Table A.2. NB: larger symbols may obscure other symbols.

metal have caused an increased mercury load in marine, freshwater and terrestrial environments. Today the main sources of the metal to the marine environment are atmospheric deposition, industrial outfalls and municipal sewage. Overall, atmospheric deposition appears to be important except where it is outweighed by pollution near point sources and in estuaries draining industrialised regions.

6.3.3. Lead

The regional temporal trend assessment indicated a decreasing trend for lead in mussels from both the North-East Atlantic and the Mediterranean Sea and for cod in the North-East Atlantic (Figure 6.1). This indicated the effect of remedial measures on atmospheric deposition. No decreasing trend was evident for lead concentrations in herring muscle in the Baltic.

The station-by-station overview of 1995–1999 levels of lead revealed background levels in blue mussels from areas remote from local or regional sources, but concentrations of the

metal were above background levels along most of the European coasts (Table 6.4, Figure 6.11). In contrast to the other metals of concern (mercury and cadmium), levels of lead in blue mussels were above the limits for human consumption at a number of locations. The high level at the Norwegian west coast was caused by inputs from the zinc smelter in Sørkjøya. The other locations also appeared to be associated with local point sources rather than riverine inputs.

Lead concentrations in cod liver were generally low. An exception was in the inner Oslofjord where values were higher but did not exceed 10 times the background level (Figure 6.12). Elevated levels may have been due to urban influence. Concentrations were intermediate in herring muscle (data from the Baltic only).

Of the 266 temporal trends statistically analysed on a station-by-station basis (194 for mussels, 72 for fish) 39 were significant, 30 down and nine up (Table 6.4, maps in Figures 6.13 and 6.14). Inputs of lead

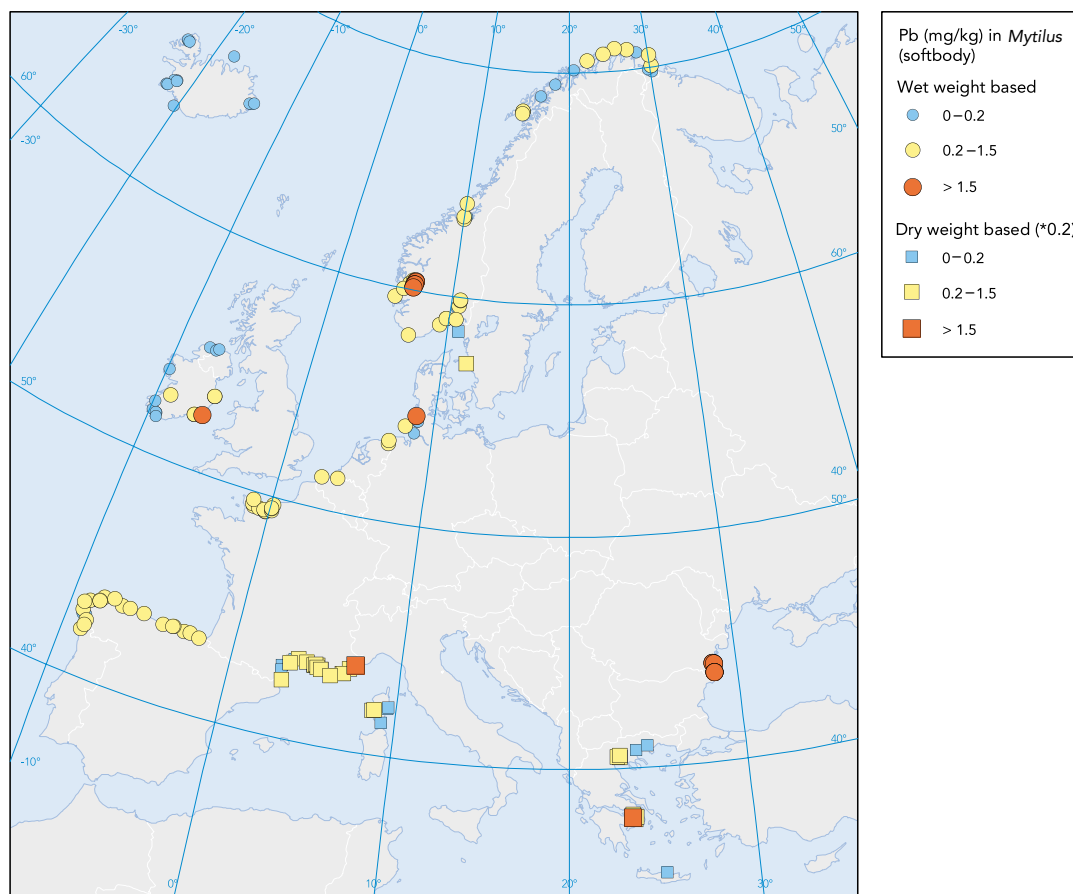
Table 6.4

Subindicator summary assessment for lead in mussels and fish from the North-East Atlantic, Baltic Sea, Mediterranean Sea and Black Sea

Sea region	Sub-indicator	Spatial variation			Time trends		
		Total no. of stations	Number over low	Number over high	Total no. of stations	Number down	Number up
North-East Atlantic	Mussels	116	61	8	156	12	2
	Fish	21	1	0	43	10	0
Baltic Sea	Mussels	1	1	0	1	0	1
	Fish	10	8	0	29	3	0
Mediterranean Sea	Mussels	36	23	2	37	5	6
	Fish	0			0		
Black Sea	Mussels	3	3	3	0		
	Fish	0			0		

Notes: See Table 6.1 for selection of indicator species and low/high concentrations limits. See also results in maps in Figures 6.11–6.14.

Figure 6.11

Lead (Pb) in mussels (*Mytilus edulis* — North-East Atlantic; *M. galloprovincialis* — Mediterranean and Black Sea), median mg/kg wet weight for 1995–1999 (2001 for Black Sea)

Notes: See Table 6.1 for basis of classification. EU legislation limit for lead in foodstuffs 'bivalve molluscs' is 1.5 mg/kg wet weight (EU, 2002). Based on data from OSPAR and EEA member countries (Mediterranean), and data reported by Romania, see also Table A.3. NB: larger symbols may obscure other symbols.

decreased (OSPAR, 2000), but no clear overall trends were found using mussel and fish state indicators. However, there was a certain dominance of decreasing trends over increasing trends on a station-by-station basis. Increasing concentrations were found in Ems and Gothenburg (as was the case for

mercury), and in the Aegean Sea (Figure 6.13). The reasons for these increases were not clear. Concentrations decreased at nearly all the Norwegian locations, the German Bight and in the Scheldt region (mouth of the Rhine). Statistically significant trends were found mainly at locations in estuaries