# Testing of indicators for the marine and coastal environment in Europe

## Part 2: Hazardous substances

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The information describing the situation during summer 2002 is partly based on non-validated monitoring data and hence should be regarded as preliminary.

# Contents

# Summary

**Background**
Indicator reporting by the European Environment Agency (EEA) will be based on yearly updated information. The use of indicators facilitates major communication on the present and future state of the environment in relation to European environmental policies. Indicator testing by the European Topic Centres is the basis for the further harmonisation of the assessment reports from the regional sea conventions and the development of a core set of indicators for EEA indicator based reporting.

This report is the second in a series of three reports on indicator testing for the marine and coastal environment and describes the process and the results of testing of potential indicators for EEA reporting by means of identification of data availability, data reliability, and data processing of a selected set of descriptive parameters for the policy issue 'hazardous substances'. The testing focused on the inputs of hazardous substances into the marine environment for potential pressure indicator, and the concentration of hazardous substances in sediment and in organisms for potential state indicator.

Due to a lack of reliable and comparable European data, the testing mainly concentrated on the method for aggregation of data from monitoring stations and the presentation of trends in hazardous substances in the north-east Atlantic including the North Sea (OSPAR data). OSPAR has not adopted an indicator-based approach nor annual assessments. Therefore this testing study aims to show, if and how OSPAR data can be used for the EEA core set of indicators on hazardous substances. The results of this testing exercise were discussed with member countries of Marine Conventions and EEA during an Inter Regional Forum workshop on indicators in June 2001 and are reflected in a separate workshop report as well as in the further development of Hazardous substance indicators as presented in the third report in this series. Hazardous substances considered in this report are those within OSPAR monitoring programmes: cadmium, mercury, lead, zinc, lindane PCB7 and TBT. However, time series on inputs and concentrations of tributyl tin (TBT) were not available, and hence TBT was excluded from further testing.

**Pressure indicator on direct and riverine input of hazardous substances**
An indicator produced from the sum of direct and riverine inputs of all six hazardous substances into the north-east Atlantic including the North Sea shows a decreasing trend of roughly 40 % in total inputs between the years 1990 and 1998. Stated at this high level of aggregation, this conclusion is rather accurate.

Since policies address reduction measures for individual substances occurring at different quantities and with different toxicities, indicators for individual substances are also necessary. At a lower level of aggregation, when looking in more detail at changes in inputs of individual substance by sea area or even by country, trend detection of inputs becomes sensitive to the missing data and the irregularities in the time series. Since OSPAR time series are incomplete, conclusions on trends must be drawn with caution at such lower level of aggregation.

The testing showed that the direct and riverine input indicator needs improvement in the quality of data, the completeness of the time series and in the aggregation methodology. Priority should be given by countries to improvement of the data on direct and riverine input aimed both at completing the data set and at quality assurance. Only then should the statistical methods and techniques to detect trends receive attention for improvement. And finally, the relation between river flow (water quantity) and riverine input loads of hazardous substances should be analysed in more detail, making it possible to determine normalised input loads independent of yearly variable river flow.

**State indicator on concentrations of hazardous substances in sediment**
The Dangerous Substances Directive as well as the OSPAR Convention require trend measurements of hazardous substances in sediments. Therefore, EEA aims at

developing a state indicator on hazardous substances in sediments within its core set. From an examination of OSPAR data on cadmium concentrations in sediment in coastal waters, it is concluded that the reliability and comparability of the data are such that using the data currently available for state indicator development cannot be justified. Extensive time series of concentrations of hazardous substances in sediment at individual locations are not available. An indicator could only be developed, when data on concentrations of hazardous substances in sediment are improved, for example complete time series for at least a 10-year period and quality assurance of the data.

**State indicator on concentrations of hazardous substances in organisms**
The Dangerous Substance Directive as well as the OSPAR Convention also require trend measurements of hazardous substances in marine organisms. Since no environmental quality standards have been set for coastal waters against which measured concentrations could be assessed, OSPAR has developed background/reference concentrations and ecotoxicological assessment criteria instead. These have been

used in the development of the proposed indicator. An ecological reference index (ERI) for concentrations of hazardous substances in organisms is determined by dividing concentrations of these substances in organisms by the background/reference concentration (BRC) or the upper limit of the ecotoxicological assessment criteria (EAC) ranges. Using OSPAR data on concentrations of hazardous substances in blue mussel in the north-east Atlantic including the North Sea, a decreasing trend was found in the ERI during the period 1990-96. The ERI appears to provide a suitable state indicator. The first testing results look promising and it is recommended to further explore the possibilities of using this indicator.

**Monitoring strategy**
It is recommended to standardise and harmonise monitoring of inputs and concentrations of hazardous substances on a European scale by building upon Inter Regional Forum initiatives. Efforts to standardise data collection and dissemination need to be encouraged and enhanced at all levels: member country, Marine Convention and European level.

# 1. Introduction

## 1.1.  Background

In 1998, the EEA presented the report Europe's environment: the second assessment (EEA, 1998). The main subjects identified as themes for concern in the marine and coastal environment chapter were: eutrophication, hazardous substances, overfishing and degradation of coastal zones.

In the EEA report *Environment in the European Union at the turn of the century* (EEA, 1999) the actual and foreseeable state of the environment in the EU and accession countries was assessed. The outlook was based on socioeconomic and environmental policies that are assumed to be implemented by 2010. The report describes the interrelations between human activities and the environment and serves to inform policy-makers on developments in environmental parameters and the effects of measures taken. As such, the report is a reference for strategic policy development. The analysis follows the DPSIR assessment framework adopted by the EEA. It starts with the **driving forces** (or human activities) that lead to **pressures** (that is, emissions) on the environment. As a result, changes in the **state** of the environment may lead to **impacts. Responses** must be defined to reduce the adverse impacts.

This EEA report also discussed the main challenges and problems in the coastal zones of four regional seas (the Atlantic, the North Sea, the Baltic, the western Mediterranean). The pressures of economic growth and spatial development differ between the regions. Due to various ecological qualities, the coastal zones of the regional seas have a different sensitivity to the pressures upon them.

In the EEA report *Environmental signals 2000* (EEA, 2000), the indicator assessment based on the DPSIR assessment framework was worked out more quantitatively, presenting eutrophication indicators etc. for coastal waters. For hazardous substances however, indicators still had to be developed and were published first in 'Environmental signals 2001' (EEA, 2001). Forthcoming EEA 'Environmental signals' reports will give a yearly overview on selected policy themes, including hazardous substances.

## 1.2.  Hazardous substances

### 1.2.1.  Introduction
Due to human activities, many hazardous substances (mineral oils, PCBs, PAHs, etc.) are emitted into the environment and reach coastal waters or are discharged directly into coastal waters. In this report, focus is placed on substances that are present mostly in relatively low concentrations in the environment. A distinction is made between inorganic substances (heavy metals) and organic substances (organochlorine compounds, etc.).

At least 100 000 substances are emitted into the environment, due to human activities. More than 100 of them are considered hazardous for the marine environment because they are toxic, persistent and bioaccumulative. As analysis techniques and information technology improve, more information will become available on emissions and on the fate of hazardous substances in the environment. In the past, data has been gathered on heavy metals and some organic substances such as PCBs and lindane. These substances are included in marine monitoring programmes. A provisional selection of hazardous substances included in these monitoring programmes has been used for the development of indicators:

- heavy metals: cadmium (Cd), mercury (Hg), lead (Pb) and zinc (Zn);

- organic substances: lindane (g-HCH), PCBs (polychlorinated biphenyls) and TBT (tributyl tin).

### 1.2.2.  Heavy metals
Heavy metals in coastal waters not only originate from human activities (anthropogenic sources) but also from natural sources. Metals do not degrade in the environment. However, only part of the metal concentrations in coastal waters is bioavailable, since metals occur in different chemical speciations and only some of these speciations can be taken up by organisms.

### 1.2.3. Lindane
Lindane is used as an insecticide. It degrades slowly in the environment and may affect hormone cycling in organisms (AMAP, 2000).

### 1.2.4. PCBs
Polychlorinated biphenyls (PCBs) are chemically stable and heat resistant. For that reason, PCBs are used as transformer and capacitor oils and in other applications where lubricants or oils are needed at higher temperatures. Due to their stability and relative volatility, PCBs have been spread worldwide. PCB concentrations have been correlated with reduced fertility in grey, harbour and ringed seals (AMAP, 2000).

The sum of seven congeners ($PCB_7$) is being monitored, and is used as indicator. $PCB_7$ consists of the congeners 28, 52, 101, 111, 138, 153 and 180.

### 1.2.5. TBT
Tributyl tin (TBT) is used as antifouling paint on ship hulls, and releases of TBT from this antifouling paint are an important source of TBT in coastal waters. TBT is degraded in water (half-life of a few days to several weeks). In sediments, decomposition takes place more slowly, especially in anaerobic conditions (half-life of many years) (AMAP, 2000). TBT has recently been identified in sperm whales stranded on the Dutch and Danish coasts, illustrating that this substance is no longer only found in harbours (OSPAR, 2000). The most well-known toxic effect of TBT is imposex, recorded in marine snails even at extremely low environmental concentrations (AMAP, 2000). Imposex is a phenomenon by which females develop male sexual characteristics and become sterile.

### 1.2.6. Policies aimed at input reduction
Measures to reduce the emission and release of hazardous substances and to protect the marine environment are being taken as a result of various initiatives on all levels (global and regional conventions, and ministerial conferences, European, national): UN Global Programme of Action for the Protection of the Marine Environment against Land-Based Activities; Mediterranean action plan (MAP); Helsinki Convention 1992 on the Protection of the Marine Environment of the Baltic Sea Area; OSPAR Convention 1998 for the Protection of the Marine Environment of the North-East Atlantic; EU water framework directive. The target is a substantial reduction in the input of specific heavy metals and organic

substances into coastal waters. In addition, the application of TBT in the marine environment is restricted under EU Directive 1999/51/EC of 26 May 1999. The use of TBT is forbidden on ships' hulls with a total length of less than 25 metres.

## 1.3. Objectives

The development of a common set of indicators to support policy framing and implementation was stressed at the fourth European conference of ministers of the environment in Aarhus in June 1998. Indicators can play a vital part in focusing and illuminating the significance of environmental change and the progress made on route to sustainable development. Indicators are items of quantified information that help to explain how the quality of the environment changes over time or varies spatially.

Indicators are linked to policy questions, which should be answered by the indicator (EEA, 2000b). The various input reduction policies are mentioned above. Policy questions addressed by this report are those listed below.

- What is the situation concerning the concentrations of heavy metals and POPs in marine waters, coastal sediments and organisms? Are standards exceeded?

- What is the trend concerning the discharge of heavy metals and POPs?

- What volumes of pollutants are deposited from the atmosphere to the sea?

Providing reliable and relevant data and information to support widely agreed key indicator sets in a consistent and timely way should be a main objective of any environmental monitoring and data-gathering programme. To achieve the development of such indicators, the EEA works as a facilitator between member countries, Community institutions and other environmental organisations and programmes.

Thus, the aim of this testing of a set of provisional indicators for the marine and coastal environment, derived from easily available data, is meant to be able to supply concise, reliable, quantitative information on a regular basis to support EEA reporting. The objective is to develop further an indicator

database that supplies basic (indicator) information on the European coastal zones, based on the DPSIR assessment framework.

In agreement with OSPAR guidelines (OSPAR/RID, 1998), OSPAR member countries monitor the hazardous substances mercury, cadmium, copper, zinc, lead and lindane. PCB7 is recommended for monitoring on a voluntary basis. Except for copper, these substances were selected for indicator testing as part of the study underpinning this report prepared by the

European Topic Centre on the Marine and Coastal Environment (ETC/MCE).

## 1.4. Scope

This report describes the findings of the testing of hazardous substance indicators. Chapter 2 deals with the inputs of the selected hazardous substances to coastal waters, constituting pressure indicators. State indicators, represented by the concentrations of the selected hazardous substances in sediment and in biota in coastal waters, are discussed in Chapter 3.

# 2. Pressure indicators of hazardous substances

## 2.1. Methodology

Pressure indicators of hazardous substances for the marine environment are emissions to water and air, riverine and direct inputs into coastal and marine waters and atmospheric deposition to coastal and marine waters. Emission indicators have not been considered here, due to the lack of available data. For riverine and direct inputs as well as deposition, indicators on a limited amount of substances were able to be tested.

The testing of pressure indicators of hazardous substances has been carried out through the following steps.

1.  Checking for data availability of input parameters with indicator potential, both quantitative and qualitative (uncertainty margins and reliability) and selection of input parameters to be used for testing. Relevant aspects of input data are:

    - sources contributing to the inputs of hazardous substances;
    - time coverage (time series);
    - geographical coverage.

2.  Development of an aggregation and presentation method. Choices are made on:

    - aggregation of substances;
    - aggregation over coastal water areas;
    - presentation of country contributions.

3.  Testing of input parameters:

    - detection of trends;
    - determining the robustness of the trend detection, taking into account the reliability of the data;
    - drawing conclusions about the usefulness of the indicator.

4.  Making recommendations for improving the input indicators with respect to:

    - geographical coverage;
    - standardisation and harmonisation of sampling and analysis procedure;

    - reporting results (including metadata and information on representativeness of samples).

These steps are described in the following paragraphs.

## 2.2. Data collection and data availability

### 2.2.1. Data sources and geographical and time coverage

Three large marine areas are distinguished (see Figure 2.1).

- The north-east Atlantic including the North Sea (OSPAR area): Arctic waters, the North Sea, Celtic seas, the Bay of Biscay and the Iberian coast. OSPAR collects data on inputs of hazardous substances into its coastal waters on a yearly and systematical basis.

- The Helcom area: the Baltic Sea. Helcom systematically collects data on inputs of hazardous substances on a five-yearly basis (annually from 2000 onwards).

- The UNEP/MAP area: the Mediterranean where it borders EU countries. Medpol collects data on inputs of hazardous substances into the Mediterranean.

Inputs of hazardous substances into coastal waters consist of the types listed below.

- Direct inputs: direct discharges in coastal water, from coastal discharges of industries and municipalities, waste discharges at sea and release at sea, for example from shipping and oil production.

- Riverine inputs: loads of hazardous substances entering coastal waters with river inflow. Both human activities as well as natural sources in the whole river basin contribute to this input.

- Atmospheric input: wet and dry deposition on coastal waters. Atmospheric transport of substances may take place over long distances. Again, human activities and natural sources may contribute to this input.

The European coastal waters | Figure 2.1.
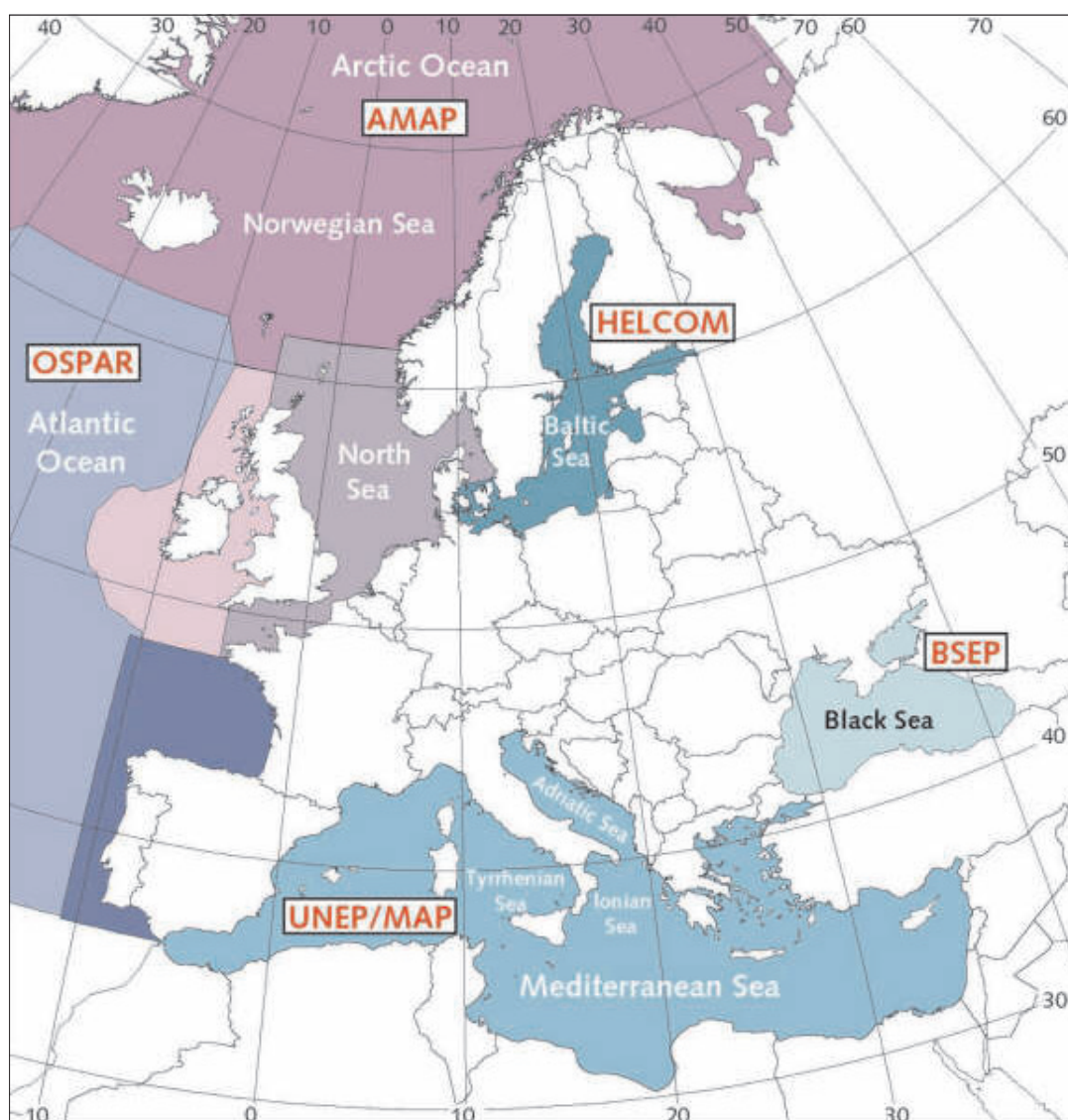


Table 2.1 presents an overview of the data on direct and riverine inputs of hazardous substances into European coastal waters that were available for the production of this report. Time coverage (completeness of time series) as well as geographical coverage (yearly country contribution to inputs) are presented.

Available data on direct and riverine inputs from OSPAR, Helcom and UNEP/MAP | Table 2.1.

| Substance | Coastal water area and country contribution ($^1$) to inputs | Time series ($^2$) available in the period 1990–98 |
|---|---|---|
| Cadmium | OSPAR area: | |
| | • Belgium, Germany, Ireland, Netherlands, Norway, UK | 1990–98 |
| | • Denmark | 1990 |
| | • France | 1990–96 |
| | • Portugal, Sweden | 1990–95, 1997–98 |
| | • Spain | 1993–95, 1997–98 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | 1995 |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |

| Substance | Coastal water area and country contribution (¹) to inputs | Time series (²) available in the period 1990–98 |
|---|---|---|
| Mercury | OSPAR area: | |
| | • Belgium, Germany, Netherlands, Norway, UK | 1990–98 |
| | • Denmark | 1990 |
| | • France | 1990–96 |
| | • Ireland | 1990–95 |
| | • Portugal, Sweden | 1990–95, 1997–98 |
| | • Spain | 1997–98 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | 1995 |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |
| Lead | OSPAR area: | |
| | • Belgium, Germany, Ireland, Netherlands, Norway, UK | 1990–98 |
| | • Denmark | 1990 |
| | • France | 1990–96 |
| | • Portugal, Sweden | 1990–95,1997–98 |
| | • Spain | 1991–92, 1994–95, 1997–98 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | 1995 |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |
| Zinc | OSPAR area: | |
| | • Belgium, Germany, Ireland, Netherlands, Norway, UK | 1990–98 |
| | • Denmark | 1990 |
| | • Portugal, Sweden | 1990–95, 1997–98 |
| | • Spain | 1991–98 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | 1995 |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |
| Lindane | OSPAR area: | |
| | • Germany, Netherlands, Norway, UK | 1990–98 |
| | • Belgium | 1990–95, 1997–98 |
| | • Denmark | 1990–95. |
| | • France | 1990–96 |
| | • Ireland, Sweden) | n.d. |
| | • Portugal | 1993–95 |
| | • Spain | 1997–98 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | n.d. |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |
| PCB$_7$ | OSPAR area: | |
| | • Germany, Netherlands, Norway, UK | 1990–98 |
| | • Belgium | 1990–95, 1997–98 |
| | • Denmark | 1990–95 |
| | • France | 1990–96 (³) |
| | • Ireland, Spain, Sweden | n.d. |
| | • Portugal | 1992–95 |
| | Baltic Sea (Denmark, Finland, Germany, Sweden) | n.d. |
| | Mediterranean (France, Greece, Italy, Spain) | n.d. |
| TBT | OSPAR area, Baltic Sea, Mediterranean | n.d. |
| (¹) Only EEA member countries are considered. (²) n.d. = no data. (³) Input data comprise only five congeners instead of seven. | | |

The OSPAR data on direct and riverine inputs is not complete. For some countries, input data is lacking, and time series of country contributions do not always cover the whole period 1990–98. Input data on TBT in the north-east Atlantic including the North Sea is lacking; only some rough estimates on TBT inputs are available. The Helcom data on direct and riverine input of heavy metals into the Baltic seas is partly estimated and incomplete, and is only available for the year 1995. Input data on lindane, $PCB_7$, and TBT is lacking. UNEP/MAP has some data available on direct and riverine inputs of hazardous substances into the Mediterranean. EEA/UNEP (1999) remarks that, in the Mediterranean, heavy metals arise mainly from natural processes and the contribution from antropogenic activities has a limited influence on local pollution problems. This finding makes monitoring of inputs less urgent in this area.

OSPAR also presents data on yearly atmospheric inputs of cadmium, mercury and lead into the North Sea (area of 525 000 km$^2$). This input data is estimated, based on deposition measurements in coastal stations surrounding the North Sea for the period 1987–95.

### 2.2.2.  Data reliability

For reasons of comparability and for making trend analysis reliable, data on inputs should be complete. However, Table 2.1 shows that this is not the case. A lot of data is missing. Besides, even if data on inputs for a coastal area is available this does not mean that all direct and all riverine inputs in that area are covered.

A close look at the data on direct and riverine inputs of OSPAR member countries, in Annex 2, reveals many irregularities in the time series, while other time series are questionably unchanging. According to a member country comment, data on direct discharges are less variable than riverine input data because they are, in most cases, estimated and not measured values. Helcom (1998) reports that many uncertainties remain, due to incomplete data sets concerning heavy metals from nearly all contracting parties. According to EEA/ UNEP (1999), quality assurance and control procedures should be further developed and implemented for the Mediterranean, to ensure data quality and reliability.

The sampling and analysis procedures and the load calculation method in rivers affect the accuracy and reliability of the input data. De Vries and Klavers (1994) showed in a case study for the Dutch parts of the rivers Rhine and Meuse that the reliability of annual load estimates is largely determined by the monitoring strategy (in particular sampling frequency) and that the reliability of the estimates can differ significantly for different substances and rivers. When the distribution of the total annual load of substances is mainly determined by flushing events (extremely high river flow due to heavy rainfall), missing samples during such flushing events will result in an underestimation of the annual load. For such rivers, load calculation improves with the frequency of sampling.

The loads of substances yearly transported with rivers into coastal waters is related to yearly river flows. In years with high river flows, input loads are higher than in years with low river flows. The relation between flow and input loads, however, is not linear. The nature of the relation depends on: the substance involved (solubility in water and adsorption to suspended solids, contribution of natural sources to the loads); river characteristics (rain-fed or glacier river, flow velocities, flow distribution within a year and frequency and extent of flushing events and suspended solids concentration); river basin characteristics (population, landscape and slopes and land use within the river basin). That means that, at present, an unambiguous relation between river flow and input loads cannot be given and that, probably, the relation will be different for each river.

When analysing results for a substance lower than the detection limit, two load estimates (upper and lower values) are used by OSPAR and Helcom. The lower estimate is calculated such that any result below the detection limit is considered equal to zero while, for the upper estimate, the value of the detection limit is used. Where this range is wide, it is an indication that most of the concentrations were below the detection limit, and previous experience has shown that in such a case the upper estimate tends to be unrealistically high (OSPAR/QSR, 2000). The detection limits depend both on the analytical method used and on the laboratory. Large differences in detection limits for the same substance are observed between countries in the Baltic Sea area (Helcom, 1998).

The difference between the high and low values in the OSPAR database for direct and riverine inputs varies between substances. The difference between high and low values in the database is large, especially for $PCB_7$. The difference between the average values calculated and the high and low values amount to about 80 % for $PCB_7$. This difference is smaller for the other substances: up to 20–30 % for cadmium, mercury and lindane; up to about 10 % or less for lead and zinc. A very high uncertainty margin of more than 90 % is observed for the contribution of the UK to $PCB_7$ inputs. This contribution therefore is highly uncertain and probably overestimated.

Air transport models and/or extrapolation methods are used to derive atmospheric inputs into coastal waters. The results differ depending on the estimation method chosen. The atmospheric input values may therefore be biased. Thus, these values should not be compared directly with direct and riverine inputs. However, time series of atmospheric input data calculated with the same estimation method may be used for trend analysis, as the relative reliability of the compared data is not sensitive to the bias.

## 2.3.   Aggregation and presentation

### 2.3.1.  Trend detection methods
Trends can be shown using absolute numbers (input loads) or relative numbers. The inputs differ largely for the six substances considered, which makes comparison difficult. Using relative numbers makes comparison between the substances easier. Moreover, in pollution abatement, targets are often fixed as percentage emission reductions.

Several methods may be used for trend detection of inputs; from simple and straightforward to more sophisticated. Dividing inputs in the last year of the period by the inputs of the first year of the investigated period is a very simple and straightforward method to detect trends. Since this method only uses the first and last year of the time series, the resulting trend detected is sensitive to the values only of these two years compared with the general pattern of the inputs over time.

More sophisticated methods use statistical techniques. For instance, trend lines may be constructed using the method of least squares, minimising the distance of individual observations to a trend line. Several trend

line curves are possible, for example linear, logarithmic, exponential. The trend line curve selected should reflect the development to be expected over time (see Figure 2.2).

Constructing trend line curves means smoothing of irregular patterns of development over time, showing possible trends more clearly. These curves do not reveal the statistical significance of the trends shown. Many statistical methods and models are available to determine the statistical significance. The trend-y-tector (see web site: http:/www.waterland.net/rikz/osparwg/ trend-y-tector) is an example.

In the OSPAR database, some input data for countries and years is lacking and some time series on country contributions are highly irregular in their input values; other time series are questionably unchanging in input values. Applying a sophisticated statistical analysis method might suggest a higher reliability of input data than that obtained in reality. Therefore, in this report, a simple and straightforward method is used to describe the trends. Some sensitivity analysis is carried out using other methods of trend detection to determine the robustness of the conclusions drawn from the results. In future, when validated input data are available, it is recommended to use a statistical based method for detection (see OSPAR/SIME, 2000).

### 2.3.2.  Trend detection applied on the inputs of hazardous substances
The atmospheric inputs and the sum of direct and riverine input are dealt with separately, since the coastal water areas for which data are available are different and the accuracy of both types of input data does not compare very well.

Though incomplete and not very accurate, the OSPAR data on direct and riverine inputs of hazardous substances in the period 1990– 98 are used for trend analysis and indicator testing. Older OSPAR data are less reliable and therefore not used. The trend detection is carried out for the four heavy metals (cadmium, mercury, lead and zinc) as well as lindane and $PCB_7$. Trend detection with TBT is not possible, as data on inputs is lacking. To derive relative numbers for each substance, the sum of loads of all direct and riverine inputs into the north-east Atlantic including the North Sea in 1990 is set at 100 %. The input percentages for other years for each substance are calculated relative to the 1990 data.
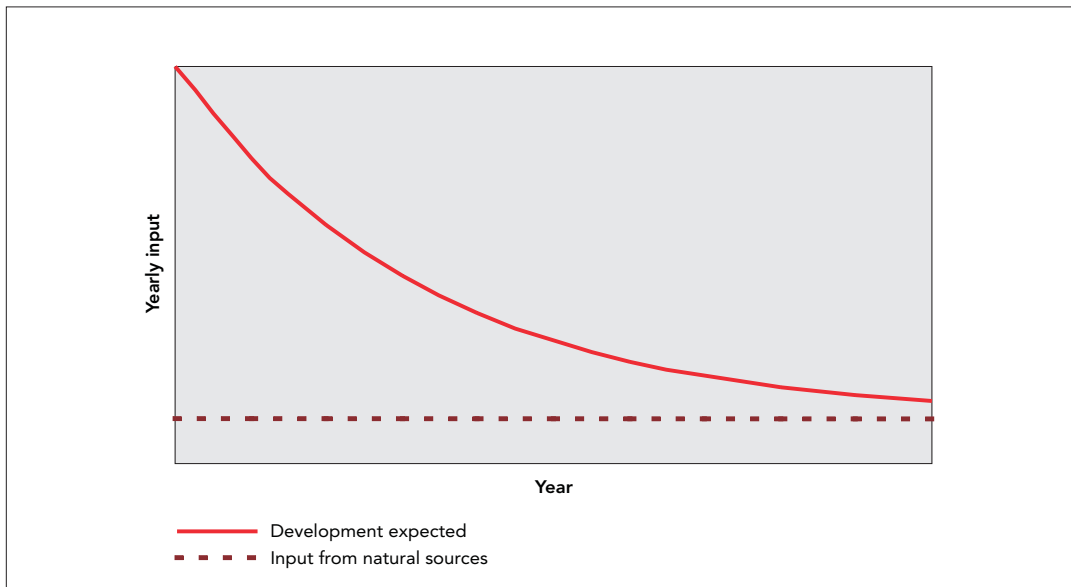
Data on atmospheric inputs is available for the North Sea for the period 1987–95 (OSPAR/ASMO, 1998). The atmospheric inputs were estimated using Method III from the CAMP (comprehensive atmospheric monitoring programme) principles. For mercury, wet deposition data is used (Method IIa).

The aggregation procedure for atmospheric inputs is similar to that for the sum of direct and riverine inputs. For each of the three metals for which data is available and for every year of the period 1987–95, an atmospheric input percentage is calculated using the 1990 atmospheric input of that substance as a base (100 %).

**2.3.3.  Levels of aggregation and presentation**
Results for direct and riverine inputs are presented at four levels of aggregation, from high (1) to low (2).

1.  Total inputs summed up for the north-east Atlantic including the North Sea, and aggregated for six substances;

2.  Total inputs for each of the six substances separately, and added up for the north-east Atlantic including the North Sea;

3.  Total inputs for each of the six substances separately, and added up for each of the four regional coastal waters of the north-east Atlantic including the North Sea;

4.  Country contributions to the input of each of the six substances separately, and added up for the north-east Atlantic including the North Sea.

## 2.4.  Results of testing

**2.4.1.  Trends in direct and riverine inputs aggregated for the north-east Atlantic including the North Sea and integrated over six substances**
Presentation on the highest aggregation level provides a general overview of the trend in inputs of hazardous substances over the period considered.

The six substances have a different fate and ecotoxicity in coastal waters. As a consequence, the six input loads cannot be simply added together; neither the absolute nor the relative numbers (percentages). To add the inputs for different substances, some kind of weighting of the inputs of each substance would be needed. Weighting factors, however, are usually subjective and often more or less arbitrary. A simple aggregation procedure is chosen using the input of each substance in a year (expressed as percentage of the input in 1990) as relative numbers. For each year, these six input percentages are averaged (see Annex 1). The yearly average percentages may be interpreted as an index showing the general development of quantities of inputs of hazardous substances over time.

Member countries recommend not to use this aggregated indicator as such, since the selected hazardous substances have levels of toxicity differing by factors of 100 and more. Concentrations would need to be converted to toxicity equivalents prior to aggregation in order to show the trend of ecotoxicological risk to the marine environment.

The results are presented in Figure 2.3. Apart from the trend line for average values, the highest and lowest value obtained for the six substances in a year are also given. Based on the 1990 and 1998 input values, a decreasing trend of 42 %, on average, aggregated inputs of hazardous substances over the period 1990–98 is calculated.

In OSPAR, procedures for river load calculation have been agreed upon (OSPAR/RID, 1998). But bias remains possible and mistakes can be made in reporting. Some examples are listed below.

- Sampling may not be representative, for example due to disturbances. In the Netherlands, suspended solids content observed in the river outflow in the mid-1990s was about three times higher compared with the period before and after (Department of Public Works in the Netherlands, 1999). Hazardous substances are strongly adsorbed on suspended solids resulting in high values for calculated riverine inputs from the Netherlands for these years (see Annex 2).

- Data on lindane input from the Netherlands in the period 1990–92 is wrong, due to a change in the analysis procedure starting in 1993 which provided more accurate data.

- Some upper values for inputs in the years 1997 and 1998 are below the lower values.

- A number of irregularities are observed in the time series of country contributions (see Annex 2):

   — cadmium inputs from Portugal over the years are very irregular;

   — the 1990 input of lead from Portugal is unbelievably high compared with the loads in the following years;

   — $PCB_7$ input from Norway in 1990 is very high compared with the loads in the following years;

   — the time series on inputs from Spain are very irregular.

- Equal loads over a number of years in time series also appear to be suspect (see zinc, lindane and PCB input from France, and lindane and $PCB_7$ input from Denmark in Annex 2).

To test the robustness of the conclusion of a 42 % decrease in inputs between 1990 and 1998, a tentative (illustrative) sensitivity analysis is carried out in Figure 2.3 for the sum of direct and riverine inputs of hazardous substances. In the data set, tentative corrections are made for missing values, for known biases and for the most suspect irregularities.

- The contributions of the Netherlands to the inputs of hazardous substances in 1994 and 1995 are reduced to 33.3 % of their original values.

| Figure 2.3. | Trend in sum of direct and riverine inputs into the north-east Atlantic including the North Sea in the period 1990–98, averaged for cadmium, mercury, lead, zinc, lindane and PCB7, and based on OSPAR data (the input of each substance in 1990 is 100 %) |
|---|---|

| | Trend in the sum of direct and riverine inputs into the north-east Atlantic including the North Sea in the period 1990–98, aggregated for cadmium, mercury, lead, zinc, lindane and PCB7 | Figure 2.4. |
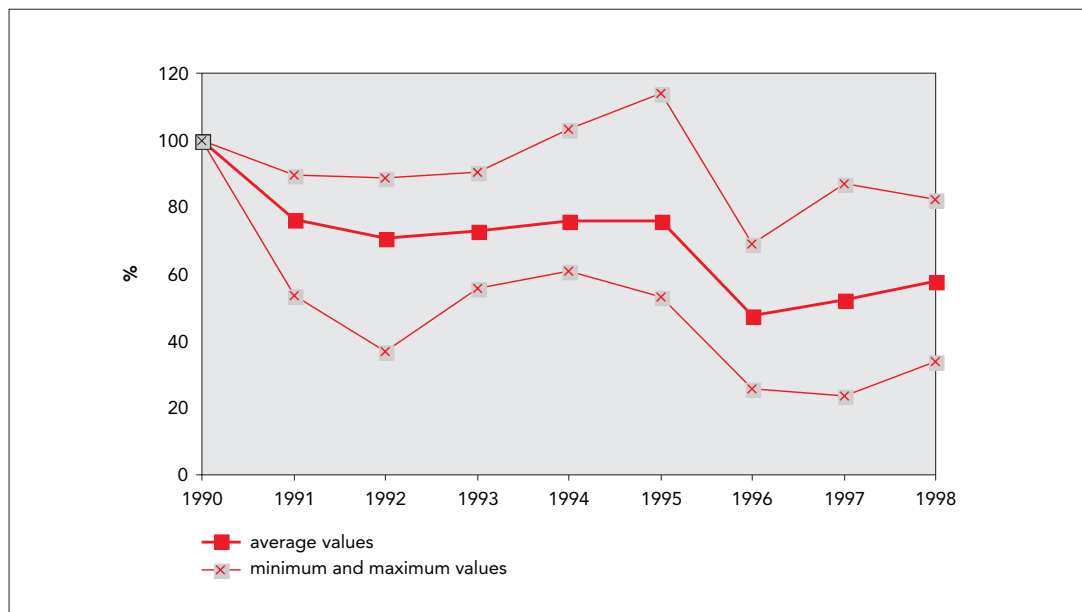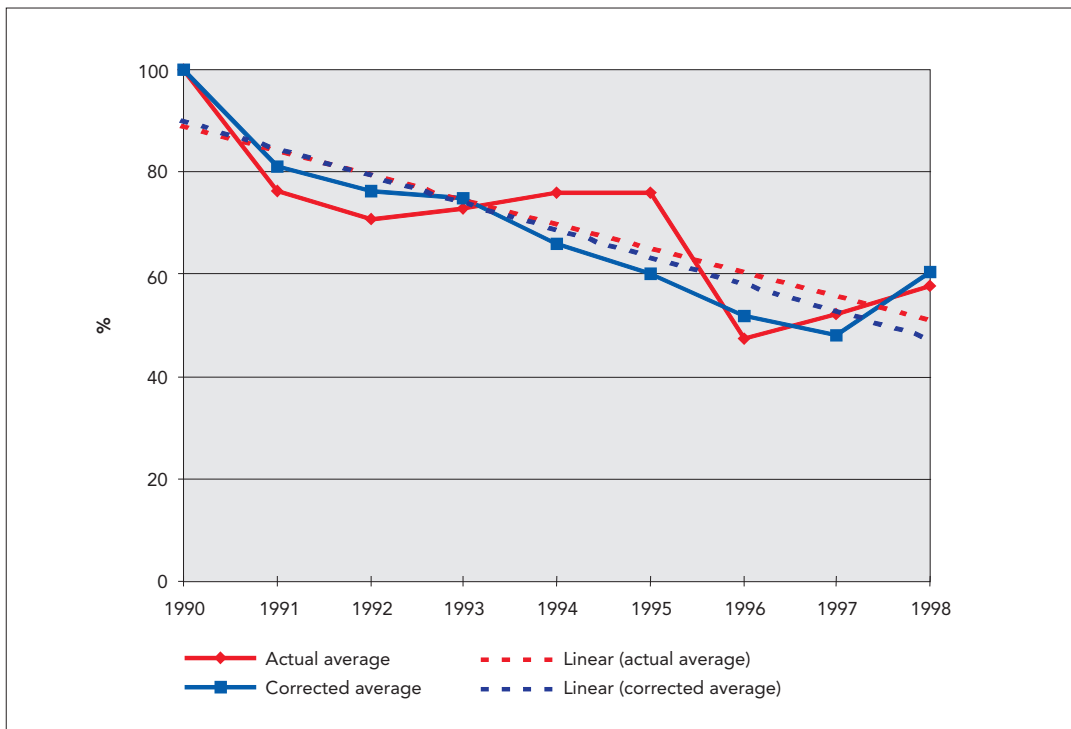


**Note:** A curve using the actual OSPAR data is presented as well as a curve with tentative corrections of the data (the input of each substance for both cases in 1990 is 100 %).

| | Trend detection for the sum of direct and riverine inputs, using actual and corrected data and using different trend detection methods | Table 2.2. |

| Method applied | Decrease using actual OSPAR data (%) | Decrease using corrected data (%) |
| --- | --- | --- |
| Simple and straightforward | 42 | 39 |
| Linear trend using method of least squares | 42 | 48 |

- The contribution of the Netherlands to the input of lindane in the period 1990–92 is corrected to the 1993 input value.

- The input of lead by Portugal in 1990 is reduced to 10 % of its reported value.

- The time series of Spain are omitted.

- Time series of country contributions lacking five or more input data are omitted.

- Missing values for country contributions to inputs are given the value of the previous year. When the missing data concerns input values at the start of the time series, values of later years are applied.

With this corrected data, a revised graph has been constructed and presented in Figure 2.4. The corrected curve is decreasing more regularly than the curve using the actual (uncorrected) data. Based on the corrected curve, a decrease in input, by

dividing 1998 input values by 1990 values, is calculated to be 39 %. With both the actual and the corrected data, linear trend lines are constructed (using the method of the least squares regression). For both trend lines, decreasing trends are calculated using the trend line values of 1990 and 1998. The results are presented in Table 2.2.

What can we learn from Figure 2.4 and Table 2.2?

- The differences in trends detected between simple and more sophisticated methods and between using the available data (data is missing and irregularities are observed in the data) and improved data are relatively small.

- At a high aggregation level, rather robust conclusions may be drawn when the trends are significant (here a large percentage decrease).

- When the trends are less significant (small percentage change), drawing conclusions from trend detection may become rather speculative.

- Improving the input data leads to an input curve closer to the linear trend line (using the method of the least squares).

### 2.4.2. Trends in total direct and riverine inputs, by substance

This aggregation level focuses on each substance separately and shows the development of inputs by substance for the whole north-east Atlantic including the North Sea, over time. The results are presented in Figure 2.5. Table 2.3 gives the results of the simple and straightforward trend detection for each substance, both for the actual OSPAR data as well as for the corrected data (see paragraph 2.4.1). From Figure 2.5 and Table 2.3 it becomes clear that, among the metals, the reduction percentage using the actual OSPAR data is largest for mercury and diminishes in the order cadmium, lead and zinc. For $PCB_7$, a larger reduction percentage is observed than for lindane. In this finding, the unreliability and probable overestimation of the UK input of $PCB_7$, as major contributor, are not accounted for. Table 2.3 also shows that the robustness of the results is greatest for the larger percentage decreases. The smaller percentage decreases (for lead, zinc and lindane) are more sensitive to data corrections.

| Table 2.3. | Decrease in the direct and riverine input of hazardous substances in the period 1990–98, based on trend detection |
|---|---|

| Substance | Decrease using actual OSPAR data (%) | Decrease using corrected data (%) |
|---|---|---|
| Cadmium | 47 | 51 |
| Mercury | 66 | 66 |
| Lead | 34 | 40 |
| Zinc | 18 | 26 |
| Lindane | 37 | 42 |
| $PCB_7$ ([1]) | 51 | 47 |
| Average | 42 | 39 |

([1]) The $PCB_7$ inputs of the UK are unreliable and probably overestimated. This may affect the results of the trend detection for this substance.

| Figure 2.5. | Trends in the sum of direct and riverine inputs of hazardous substances into the north-east Atlantic including the North Sea in the period 1990–98, based on OSPAR data (the sum of input of each substance in 1990 is 100 %) |
|---|---|
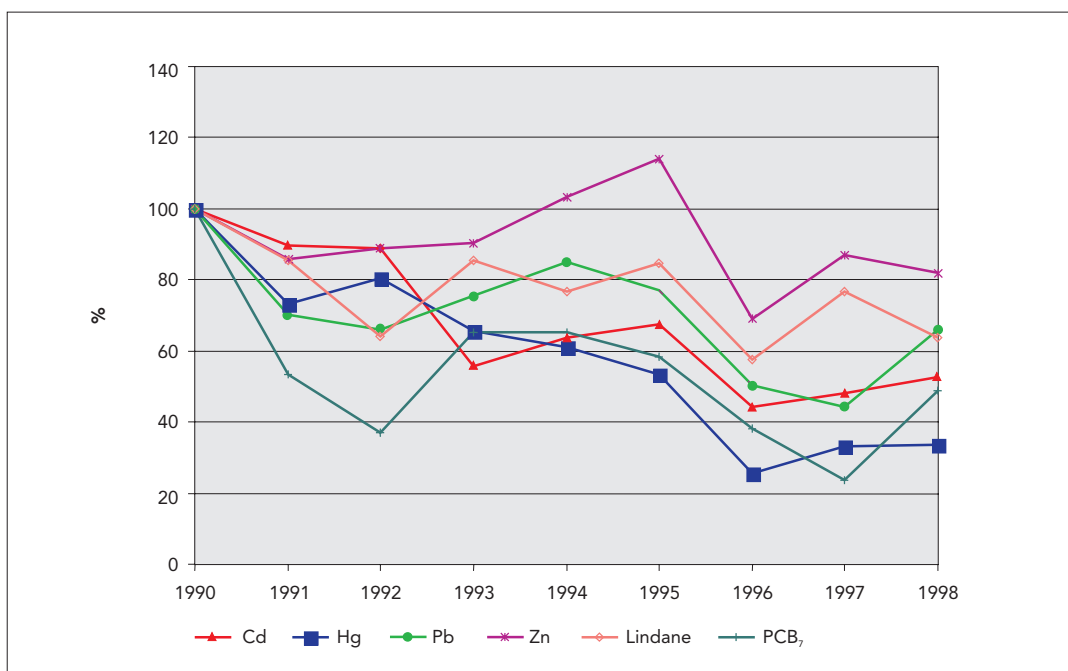
### 2.4.3. Trends in direct and riverine inputs into the four coastal waters of the north-east Atlantic including the North Sea, by substance

This aggregation level gives insight into trends for each substance in the four coastal waters of the north-east Atlantic including the North Sea, and where trends are decreasing most and where less or not decreasing.

Figure 2.6 and Table 2.4 both show — in a different way — the percentage changes of the direct and riverine inputs into the four regional coastal waters for each substance over the period 1990–98 (comparison of two years). From the figure and the table, it can be seen that the percentage change of input differs between the four regional coastal waters. For the North Sea, relatively less reduction is realised than for the other coastal water areas. At this lower level of aggregation, the influence of missing data and irregularities in the data becomes more evident. For instance, reliable percentages of change could not be determined for three substances for the Bay of Biscay and the Iberian coast.

| Percentage of change in the sum of direct and riverine input of hazardous substances into the north-east Atlantic including the North Sea, between 1990 and 1998, based on OSPAR data | | | | | Table 2.4. |
|---|---|---|---|---|---|
| Substance | Arctic waters | Greater North Sea | Celtic seas | Bay of Biscay and Iberian coast | |
| Cadmium | – 83 | – 30 | – 68 | – 18 | |
| Mercury | – 47 | – 62 | – 74 | – 77 | |
| Lead | – 46 | – 9 | – 3 | – 91 | |
| Zinc | 7 | – 31 | – 26 | n.c. | |
| Lindane | – 76 | – 39 | – 9 | n.c. | |
| $PCB_7$ | – 95 | – 40 | – 61 | n.c. | |

n.c. = not calculated, as time series for the Bay of Biscay and the Iberian coast are incomplete; **blue**: more than 40 % decrease in the period 1990–98; **green**: between 40 and 20 % decrease in the period 1990–98; **yellow**: between 20 and 0 % decrease in the period 1990–98; **red**: increase in the period 1990–98.

| Percentage of change in the sum of direct and riverine input of hazardous substances into the north-east Atlantic including the North Sea between 1990 and 1998 (comparison of two years), based on OSPAR data | Figure 2.6. |
|---|---|
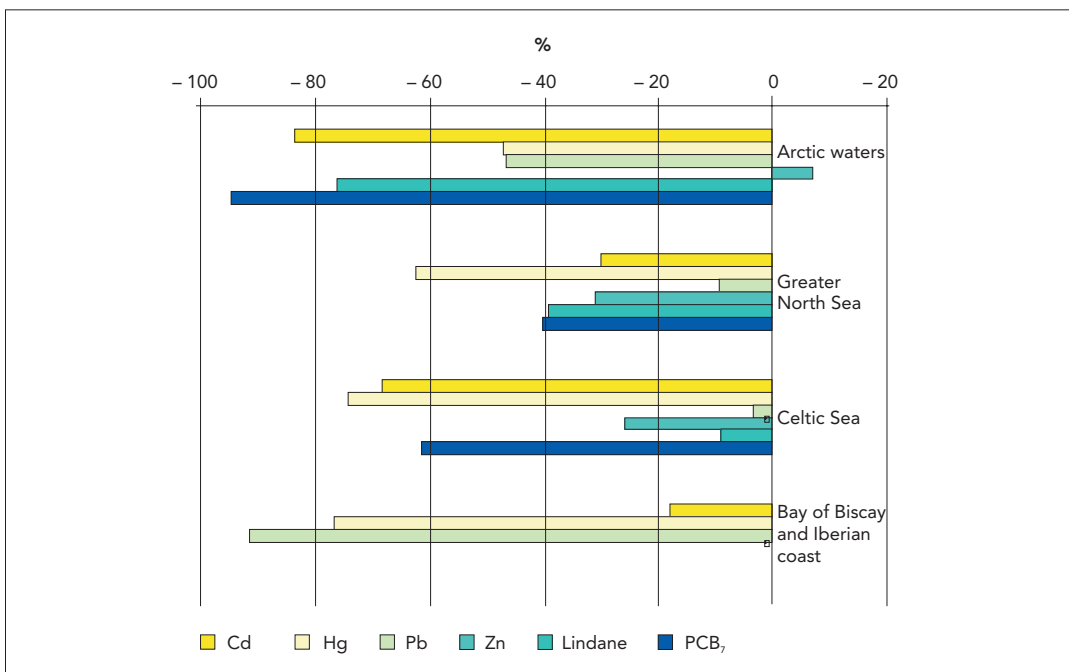


Note: Time series of the input of PCB7 into the Bay of Biscay and the Iberian coast comprises only the period 1992–95. Input data of Spain are lacking for zinc and lindane in 1990, while reported inputs in 1998 are relatively high. Due to this lack of data, no percentages of change of inputs into the Bay of Biscay and the Iberian coast are calculated for zinc, lindane and PCB7.

### 2.4.4. Trends in country contributions to direct and riverine input, by substance

This low aggregation level gives information on the development of country contributions to inputs over time. It must be remembered here that countries are not always fully responsible for these inputs, since part of the riverine input attributed to a country may originate from sources in upstream countries.

Many graphs are needed to present the results for six substances, for 11 countries (in the OSPAR data set) and for nine years (period 1990–98). In the example figure below, all country contributions to the input of zinc are presented.

Figure 2.7 shows that the contributions to the input of zinc into the north-east Atlantic including the North Sea differ largely between the countries involved. According to the data delivered to OSPAR, Belgium, France, Portugal and Sweden contribute relatively little to the input of zinc. The United Kingdom and Germany contribute most to the total decrease in input of zinc.

Apart from showing trends in country contributions to direct and riverine inputs, this aggregation level reveals the reliability of the input data. Gaps in the time series are shown, as well as irregularities and questionable regularities. The zinc input

from France is questionably unchanging. On the other hand, the input from Spain is highly irregular, and very high inputs are observed for the Netherlands for the years 1994 and 1995. Presenting the country contributions in this way should raise questions, leading to suggestions for improvements of the input database.

In Annex 2, all data is presented on country contributions to inputs, revealing the gaps in the data on inputs of hazardous substances as well as the irregularities and the questionable regularities in the data supplied to OSPAR by the countries concerned.

From Figure 2.7 and the data in Annex 2, it follows that the contributions of many countries to the direct and riverine inputs of hazardous substances into the north-east Atlantic including the North Sea decrease significantly in the period 1990–98. The United Kingdom, as the main contributor of inputs of hazardous substances in 1990, is mainly responsible for the decrease in inputs into the north-east Atlantic including the North Sea. Germany also contributes strongly to the decrease in inputs of mercury, and Norway to the decrease in inputs of lindane. Based on the data delivered to OSPAR, Portugal fully accounts for the decrease in input of lead observed between 1990 and 1998.

| Figure 2.7. | Contributions of countries to direct and riverine inputs of zinc into the north-east Atlantic including the North Sea during the period 1990–98, based on OSPAR data (total zinc input in 1990 is 100 %) |
|---|---|
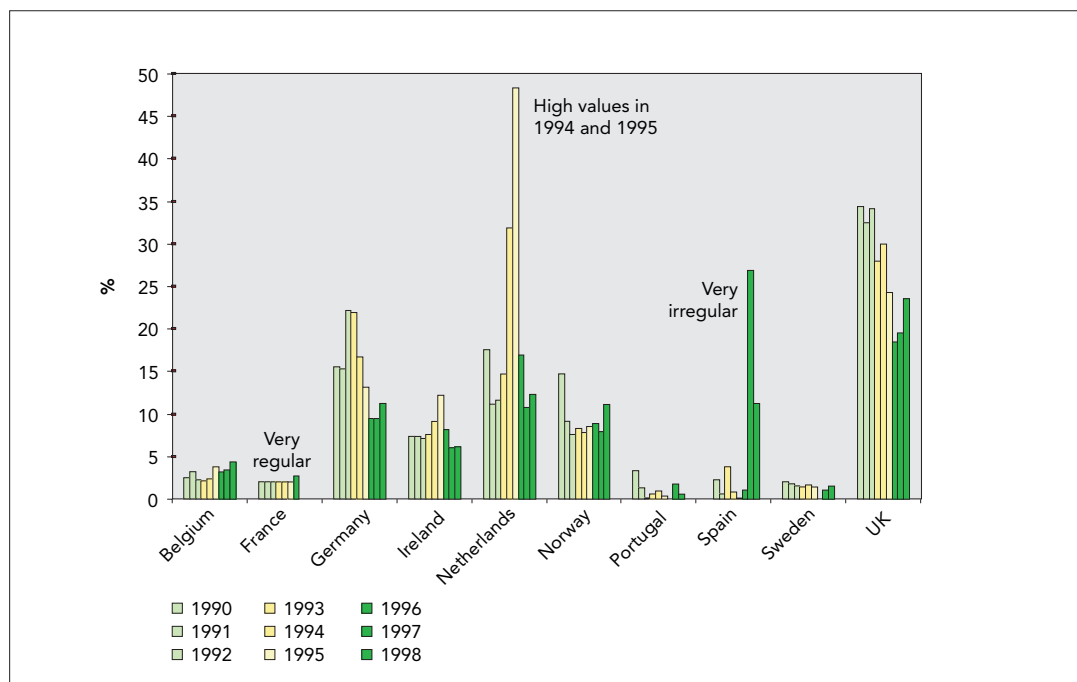


**Note:** Input data for Denmark covers only 1990 and is not presented.

The inputs from the Netherlands in the years 1994 and 1995 are high compared with inputs in other years. These high inputs are probably due to relatively high (and not representative) concentrations of suspended solids in the samples or due to high river flow. The input data on $PCB_7$ are unreliable and probably overestimated for the United Kingdom (see paragraph 2.2.2). Therefore, the results of trend detection of the UK inputs on $PCB_7$ must be considered with caution. For Denmark and Spain so much data is missing that a trend analysis is not possible. For Ireland and Sweden the same holds for the inputs of lindane and $PCB_7$.

### 2.4.5.  Trends in atmospheric inputs
Most substances have a rather short residence time in the atmosphere (of the order of a few days), and they are rapidly deposited by rain (wet deposition) or through sedimentation (dry deposition). The general pattern shows a clear decrease in deposition levels further from the coast and towards open water. The limited residence times imply that the contribution of more distant countries (not bordering seas) to atmospheric deposition to the seas is small. In contrast, hazardous substances such as PCBs exhibit very long residence times and their atmospheric

transport must be considered on a larger, even global, scale (OSPAR/QSR 2000).

Apart from some local (oil production) and diffuse (shipping) sources further away from the coasts, direct and riverine inputs into the seas become nil. Although atmospheric deposition also decreases, further away from the coasts atmospheric input becomes the major and often the only source of input.

Table 2.5 illustrates the importance of atmospheric inputs in contributing to total inputs in coastal waters. In this table, the atmospheric inputs of cadmium, mercury and lead into the North Sea in 1995 are compared to direct and riverine inputs. The contribution of atmospheric input to total inputs ranges from 20 % for mercury, to 28 % for cadmium and 32 % for lead.

Atmospheric inputs to the north-east Atlantic including the North Sea also are calculated using a long-range air transport model and air emission data. The results differ considerably from the atmospheric input data in Table 2.5, which is based on deposition measurements in coastal stations surrounding the North Sea. The differences are due to overestimation of the measured wet deposition and an underestimation of the emission data (OSPAR/ASMO 1998).

| Inputs of cadmium, mercury and lead from different sources into the North Sea (525 000 km²) in 1995, based on deposition measurements in coastal stations surrounding the North Sea | | | | | | | Table 2.5. |
|---|---|---|---|---|---|---|---|
| Input source | Cadmium | | Mercury | | Lead | | |
| | Tonnes | % | Tonnes | % | Tonnes | % | |
| Direct | 2.5 | 3 | 0.6 | 3 | 61 | 3 | |
| Riverine | 53 | 69 | 15.5 | 77 | 1 361 | 65 | |
| Atmospheric | 22 | 28 | 4 | 20 | 670 | 32 | |
| Total | 78 | 100 | 20 | 100 | 2 092 | 100 | |

**Source:** OSPAR/ASMO 1998, using average values for direct and riverine inputs.

| Figure 2.8. | Trend in atmospheric inputs of cadmium, mercury and lead into the North Sea in the period 1987–95, based on OSPAR data derived from deposition measurements in coastal stations surrounding the North Sea (the atmospheric input of each metal in 1990 is 100 %) |
|---|---|
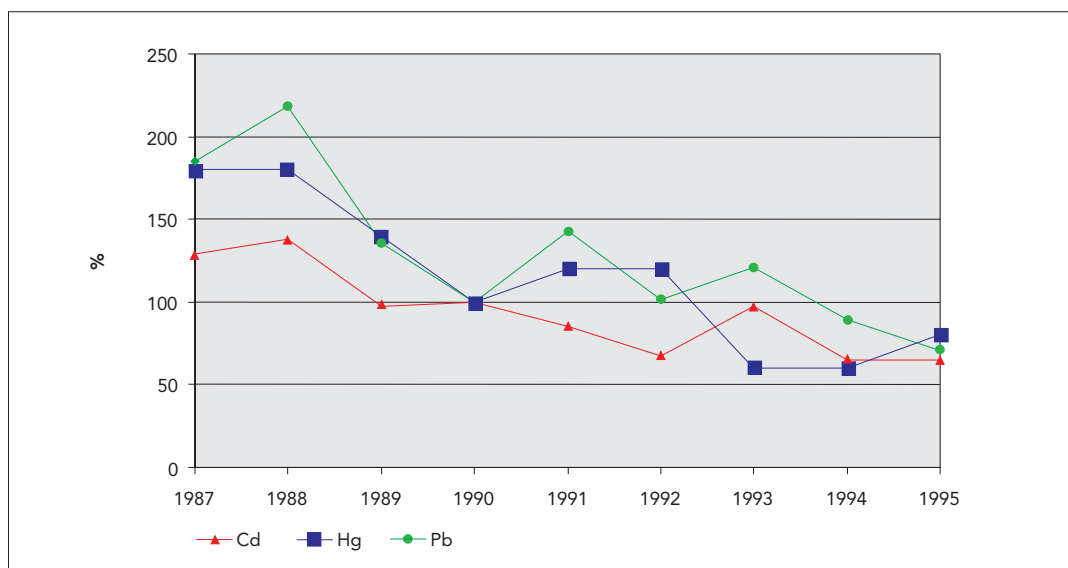


Figure 2.8 gives an overview of the trends in atmospheric inputs of the heavy metals cadmium, mercury and lead into the North Sea in the period 1987–95. From this figure, it becomes clear that the atmospheric inputs of cadmium, mercury and lead decrease significantly (on average by about 55 %) between 1987 and 1995. The inputs of mercury and lead decrease relatively more than the input of cadmium in the period considered. According to the figure, the yearly decrease in the period until 1990 is somewhat larger than the yearly decrease in the period thereafter.

Determining trends in this way is sensitive to deviations in the input values for the first and last year compared with the general pattern over time (see paragraph 2.3.1). For instance, yearly rainfall may affect wet deposition.

## 2.5.  Conclusions and recommendations

### 2.5.1.  Trend detection and robustness of results

From the results presented in the preceding paragraphs, the following conclusions can be drawn about trends in the inputs of hazardous substances into the north-east Atlantic including the North Sea.

*Sum of direct and riverine inputs of six hazardous substances into the north-east Atlantic*
☺  The inputs decrease on average by roughly 40 % in the period 1990–98.
☺  The decrease in inputs between 1990 and 1998 varies among the regional coastal waters of the north-east Atlantic

including the North Sea. The decrease is relatively less for the greater North Sea than for the other three regional coastal waters.

*Direct and riverine inputs of each hazardous substances separate*
☺  Among the metals, the direct and riverine input of mercury decreases most between 1990 and 1998. This is followed by cadmium. The decreases in inputs of lead and zinc are less substantial.
☺  The input of PCB$_7$ decreases more than the input of lindane between 1990 and 1998.

*Direct and riverine inputs by country*
☺  The contributions of many countries to the inputs of hazardous substances decrease significantly between 1990 and 1998.
☺  The United Kingdom, as the main contributor of inputs of hazardous substances in 1990, is mainly responsible for the decrease in inputs.

*Atmospheric inputs*
☺  The inputs of cadmium, mercury and lead into the North Sea decrease significantly between 1987 and 1995.
☺  The input of mercury and lead decrease relatively more than the input of cadmium between 1987 and 1995.

*Robustness of results of trend detection*
☺  At high levels of aggregation, results of trend detection may be rather robust when the trends detected are significant. In that case, the results are also less

sensitive to the method applied for trend detection. Simple and straightforward methods (directly comparing the value of the last year of the period considered with that of the first year) give results comparable to those of more sophisticated (statistical) methods.

☹ At lower levels of aggregation and/or when trends detected are less significant, the results of trend detection become sensitive to missing data and irregularities in time series, and to the method applied. In such cases, conclusions on trends might become speculative.

☹ A low reliability of input data on substances also makes trend detection less reliable. Caution is needed in drawing conclusions on trend detection.

### 2.5.2. Usefulness of the input indicator

Inputs of hazardous substances into coastal waters appear to provide a meaningful pressure indicator. The indicator makes visible the effects of European policies to abate anthropogenic emissions of hazardous substances to the environment.

The input indicator can be used at different levels of aggregation. At the highest level, the inputs of different hazardous substances are integrated and summed up over all coastal areas. This level gives general insight in (an overview of) the results of abatement policies for hazardous substances. At lower levels of aggregation, details are shown for different substances and geographical areas, which is useful, for instance, in determining 'hot spots', where inputs are high, and in determining the effects of substance-specific abatement policies.

Presentation of inputs at low levels of aggregation makes it possible to check input data, to identify missing values, and to question irregularities and suspect regularities in reported time series. As a result, the data set will become more complete and the reliability of the data will improve. The usefulness of direct and riverine inputs of hazardous substances as a pressure indicator benefits significantly from data improvement. Then, trends detected from the data become more accurate and reliable and the trends observed follow a more regular pattern (closer to reality).

### 2.5.3. Recommendations for improving the input indicator

In paragraph 2.2.2, some remarks are made on the reliability of input data reported by

countries. The usefulness of inputs of hazardous substances as an indicator improves significantly when the data set on inputs is made complete and is checked for the reliability and comparability of the data.

Determining riverine inputs is complex and laborious. For each river inflow to a coastal water this includes frequent flow measuring, frequent sampling and analysing for (often barely detectable) hazardous substances, and calculating yearly loads from this information. As a consequence, data sets are often incomplete and the reliability of data may be questioned. This also means that items of data for rivers cannot always be compared with each other.

Guidelines for monitoring exist (OSPAR/ RID, 1998; Helcom, 1998) but improvement remains possible and harmonisation on a European scale is recommended. EEA/ UNEP (1999) reports that development of an effective, common Mediterranean monitoring system of measurements of contaminants and their effects is still missing, although monitoring in the Mediterranean has been in place for a long time. That report also states that, in the Mediterranean, quality assurance and control procedures should be further developed and implemented to ensure data quality and reliability. ICES (2000) indicates that efforts to standardise data collection and dissemination need to be encouraged and enhanced and that there is a need for improved, integrated monitoring through coordination and harmonisation of existing national and international monitoring activities, as well as through implementation of new methods and technology.

Priority should be given to improving the data on input, by aiming at completing the data set and at quality assurance. Only then should attention be paid to improving statistical methods and techniques to detect trends. Finally, the relation between river flow and riverine input loads should be analysed in more detail, making it possible to determine normalised input loads standardised for yearly variable river flow.

Determining atmospheric inputs is complex too. Often, simulation models are used to determine atmospheric inputs, which makes the atmospheric input data comparable with each other but often not directly with direct and riverine inputs. Further improvement of methods to determine atmospheric inputs and to integrate these inputs with direct and riverine inputs is recommended.

# 3. State indicators of hazardous substances

## 3.1. Methodology

The proposed state indicators of hazardous substances in coastal waters concern the concentrations of these substances in sediment as well as in organisms. Each indicator is dealt with separately.

The testing of state indicators of hazardous substances has been carried out through the steps listed below.

1. Checking for data availability for concentrations of hazardous substances with indicator potential, both quantitative and qualitative (uncertainty margins and reliability) and selection of concentration parameters to be used for testing. Relevant aspects of data on concentrations are:

   - concentrations in sediment and in organism,
   - time coverage (time series),
   - geographical coverage.

2. Development of an aggregation and presentation method. Choices are made on:

   - aggregation of substances,
   - aggregation over coastal water areas.

3. Testing of concentration parameters:

   - detection of trends,
   - determining the robustness of the trend detection, taking into account the reliability of the data;
   - drawing conclusions about the usefulness of the indicator.

4. Making recommendations for improving the concentration indicators with respect to:

   - geographical coverage and spatial distribution,
   - standardisation and harmonisation of sampling and analysis procedure,
   - reporting of results (including metadata and information on representativeness of samples).

These steps are described in the following paragraphs, first for concentrations of hazardous substances in sediment and then for concentrations in organisms.

## 3.2. Availability of data on sediment concentrations

### 3.2.1. Data sources and geographical and time coverage

ETC/MCE compiled a database with data on concentrations of hazardous substances in sediment of coastal waters. To gain insight into the availability of data and the geographical and time coverage, the data dealing with cadmium has been examined in more detail.

The database contains data on sediment in the coastal waters of Greece in the Mediterranean, and in the north-east Atlantic including the North Sea, bordering Belgium, France, Germany, Ireland, the Netherlands, Norway, Spain, Sweden and the United Kingdom. For cadmium, the data covers the period 1985–97.

The data in the database comprises:

- country, coastal zone, and location of sampling;
- year of sampling and sometimes the month;
- the unit in which the results are expressed;
- the number of samples taken at the location;
- the minimum observed result, the maximum, and the median per year.

If two samples are taken at one location in a year, the median value in the database is set equal to the arithmetic mean of the two samples. Filling the database has not been carried out uniformly. For instance, data for Spain is presented separately for each sample taken at one location instead of a minimum, a maximum and a median value of all samples taken in a year. The units in which the results are expressed differ between locations, and sometimes between years at the same location.

For cadmium, time series covering more than three years at one location are rare. Two time series are available for Greece, one covering

four years and the other covering five years. One time series, covering the period 1985–98, is available for the Netherlands, but 1996 data is missing.

### 3.2.2. Data reliability

Examining the data for cadmium raised many questions about the reliability and comparability of the data. Different units are used to present results of the analysis on cadmium, and the recording of the units in the database seems questionable. The same holds for recording the analysis results in the data set. A value as high as 250 mg/kg cadmium in sediment is recorded for Greece. Belgium reports on sampling at one location with widely divergent analysis results (differing by a factor of 10 to 20).

Metadata on sampling and analysing procedures is lacking. It is known that, in analysing the samples, the choice of the liquid to extract metals from the sediment has a major effect on the analysis results. The grain size distribution in the sample also affects the results. For reasons of comparison, normalisation of results to a standard grain size distribution is needed. It is also important to know where samples are taken. The analysis results from samples taken in a highly dynamic (erosive) area may differ strongly from those in a sedimentation area in the neighbourhood of input sources.

### 3.2.3. Usefulness of sediment concentrations as indicator

Prolonged time series are needed for reliable trend analysis. Since these are lacking, no trend analysis of cadmium in sediment was performed. Furthermore, from the examination for cadmium, we conclude that the doubts about reliability and comparability of the data on concentrations of hazardous substances in sediment are such, that using the data currently available for state indicator development cannot be justified. There are no indications that the data on other substances is more reliable or more comparable. Therefore, the testing of these parameters was ended.

The usefulness of concentrations of hazardous substances in sediment as an indicator may improve when the data set on concentrations is made complete and is checked for the reliability and comparability of the data. Performing trend analysis with the improved data may then show that this parameter has indicator potential.

OSPAR (1994) has guidelines on the monitoring of sediment. However, it is recommended to develop and harmonise monitoring of concentrations of hazardous substances in sediments on a European scale (see paragraph 2.5.3). In developing a European strategy, morphodynamics of sample areas should be addressed. Also, the geographical scale of collecting data on concentrations in sediments should be agreed upon as well as the time scale (once every two or three years seems enough). The monitoring strategy must further deal with the standardisation of sampling and analysis procedures (normalisation to standard grain size distribution, extraction liquid to use for analysis).

## 3.3.  Availability of data on concentrations in organisms

### 3.3.1. Data sources and geographical and time coverage

The data set of OSPAR/MON (1998) contains data on concentrations of hazardous substances in organisms in the north-east Atlantic including the North Sea. Marinebase (Nygaard et al. 2001) also contains Medpol data on concentrations in organisms in the Mediterranean. Some data are available for the Baltic Sea but time series are missing.

The data from OSPAR/MON (1998) and Marinebase includes:

- the concentrations of the heavy metals (cadmium, mercury, lead and zinc) and of lindane and $PCB_7$ in organisms, but no data on TBT;

- the country, the station and the coastal zone of sampling;

- the organism and the tissue analysed (concentration in soft body tissue for the blue mussel, in fish muscle, in fish liver, or in all tissues, based on fat weight);

- the yearly median values for the concentration and the number of observations in a year.

Data was submitted to OSPAR/MON by Belgium, Denmark, France, Germany, Iceland, the Netherlands, Norway, Spain, Sweden and the United Kingdom. The vast majority of the data concerns the coastal waters of the North Sea. The OSPAR/MON data set contains data for the period 1978–96, but the time series are far from complete.

Data from many years is lacking. For some locations, data is available mainly from the period before 1990; for others, mainly for the period 1990–96.

In the OSPAR/MON data set, data for $PCB_7$ is limited. Seven time series covering the period 1991–96 are available for $PCB_7$ in blue mussel along the Spanish coast. Germany reports nine time series but only for three years. Time series reported by the UK cover 11 to 12 years but, instead of $PCB_7$, these time series concern another selection of congeners. Other countries did not submit data on PCBs.

France, Greece, Italy and Spain submitted data to Medpol on concentrations of hazardous substances. This Medpol data is available in Marinebase and covers the period before 1990. However, most data is from the period before 1980.

### 3.3.2. Data reliability
OSPAR/MON (1998) applied quality assurance on the data submitted by the countries rejecting data of 'poor' quality. Only recently the Medpol data has been scrutinised, cleared and classified according to its reliability (EEA/UNEP, 1999). Only data after 1987 are used in this report.

Concentrations of hazardous substances in organisms may be sensitive to seasonal variations, and to age and sex of the organism (AMAP, 2000). For example, Heesen (1995) showed that the concentrations of hazardous substances in blue mussels vary strongly over the seasons, probably due to fluctuations in body weight. He also proved that concentrations of hazardous substances in mussels decrease when body weight increases. In its guidelines for monitoring of biota, OSPAR (1994) accounts for these variations.

## 3.4.   Aggregation and presentation

### 3.4.1. Statistical trend analysis
Generally, a time lag will be observed between changing inputs of hazardous substances into coastal waters and concentrations in organisms in these waters. The time lag depends on the metabolism of substances in organisms (uptake, accumulation, degradation, excretion). Therefore prolonged time series are needed to detect significant trends following the trends in inputs.

OSPAR/MON (1998) statistically analysed the time series in the database that covered at least five years, to find trends. The results of this trend analysis for the six hazardous substances considered here are presented in Table 3.1. In total, 380 time series were analysed. Of these time series, only 65 (17 %) show a significant trend, of which 10 (3 %) are upwards and 55 (14 %) downwards. The best results are obtained for lindane: 38 % of the time series analysed show a statistically significant downward trend.

### 3.4.2. Further trend detection applied to concentrations in organisms
Quality assurance was applied to the OSPAR/MON data. Therefore, this database was used for further testing. Of the 65 significant trends observed for the six substances at different locations and for the different organisms and tissues, 39 (i.e. the majority) concern the blue mussel.

Statistically significant linear trends in concentrations of hazardous substances in blue mussels are found in the coastal waters of Belgium, Denmark, France, Germany, Iceland, Norway, Spain and Sweden, mainly at locations in estuaries and fjords.

The statistical analysis gave best results for lindane, so the next step in testing was made with lindane concentrations in blue mussel. OSPAR/MON data is available for the period 1978–96. The time series can be split into time series before and after 1990 (see paragraph 3.3.1). The period 1990–96 was chosen for further testing. This resulted in 21 time series for cadmium, 24 for mercury, 16 for lead, 26 for zinc and 35 for lindane. Roughly one third of the yearly data concentrations is lacking. The data on concentrations in blue mussel covers the coastal waters of Belgium, France (no data on lindane), Germany (no data on mercury and lead), the Netherlands, Norway and Sweden.

For the period 1990–96, the maximum and minimum of the median lindane concentrations in blue mussels observed in a year (independent of the location) were plotted. Also, the arithmetic average value of all medians in a year was calculated and plotted. The average values plotted show a clear decreasing trend in the period 1990–96. Repeating this procedure for the heavy metals gave similar results, but the trends in the average values observed are less evident. $PCB_7$ was excluded from this testing, due to the few data available.

| | Trend analysis by OSPAR/MON (1998) performed on time series for concentrations of six hazardous substances in organisms in the north-east Atlantic including the North Sea | | | | Table 3.1. |
|---|---|---|---|---|---|

| Substance | Length of time series (years) | Number analysed | Number [1] of linear trends up | Number [1] of linear trends down |
|---|---|---|---|---|
| Cadmium | 5–6<br>7+ | 34<br>36 | 0<br>1 | 2<br>3 |
| | | 70 | 1 | 5 |
| Mercury | 5–6<br>7+ | 32<br>64 | 0<br>1 | 3<br>8 |
| | | 96 | 1 | 11 |
| Lead | 5–6<br>7+ | 36<br>20 | 0<br>2 | 6<br>2 |
| | | 56 | 2 | 8 |
| Zinc | 5–6<br>7+ | 38<br>55 | 2<br>3 | 3<br>6 |
| | | 93 | 5 | 9 |
| Lindane | 5–6<br>7+ | 20<br>38 | 1<br>0 | 3<br>19 |
| | | 58 | 1 | 22 |
| $PCB_7$ | 5–6<br>7+ | 7<br>0 | 0<br>0 | 0<br>0 |
| | | 7 | 0 | 0 |
| Total of six hazardous substances | 5–6<br>7+ | 160<br>220 | 3<br>7 | 17<br>38 |
| | | 380 | 10 | 55 |

[1] Only temporal trends with a significant linear component ($P < 0.05$) and no significant non-linear component (7+ years time series) are reported.

Based on these results, a similar exercise was carried out, using the Medpol data in the Marinebase, on cadmium concentrations in the mussel in the Mediterranean in the period 1981–90. A trend could not be detected in this period. Also, since the Medpol data on concentrations of hazardous substances in organisms in the Mediterranean is rather old, and its quality is not assured, further testing on the Medpol data was not applied.

For reasons of comparison, the concentrations of substances in organisms must be translated to uniform and comparable units. This is done by dividing the concentrations in the blue mussel by the value of the background/reference concentration (BRC) or the ecotoxicological assessment criteria (EAC) for each substance. The resulting factor represents an ecological reference or a potential for environmental effects and is named ecological reference index (ERI):

ERI = measured concentration: BCR or
ERI = measured concentration: EAC

Assuming that the ecological references of the five substances considered are independent (no synergetic or antagonistic effects), the individual ERIs of substances may be added up to an aggregated or combined ERI.

The BCR or EAC values used to calculate the ERI are presented in Table 3.2. The BCR or EAC values are derived from Annex 5 of OSPAR/MON (1998). The upper limits of the BCR or EAC ranges are used here.

| Table 3.2. | Upper limit of BCR or EAC ranges for hazardous substances in blue mussel according to OSPAR/MON (1998) |
|---|---|

| Substance | Upper limit of EAC ranges (¹) | Upper limit of BCR value (¹) | Unit |
|---|---|---|---|
| Cadmium | | 550 | ng/g **dry** weight |
| Mercury | | 50 | ng/g **dry** weight |
| Lead | | 950 | ng/g **dry** weight |
| Zinc | | 150 000 | ng/g **dry** weight |
| Lindane | 40 | | ng/g **fat** weight |
| (¹) The lower limit is about 10 times smaller. | | | |

| Figure 3.1. | Ecological reference index value for cadmium in blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 (minimum, average and maximum of median values observed in a year for all locations) |
|---|---|



| Figure 3.2. | Ecological reference index value for mercury in blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 (minimum, average and maximum of median values observed in a year for all locations) |
|---|---|
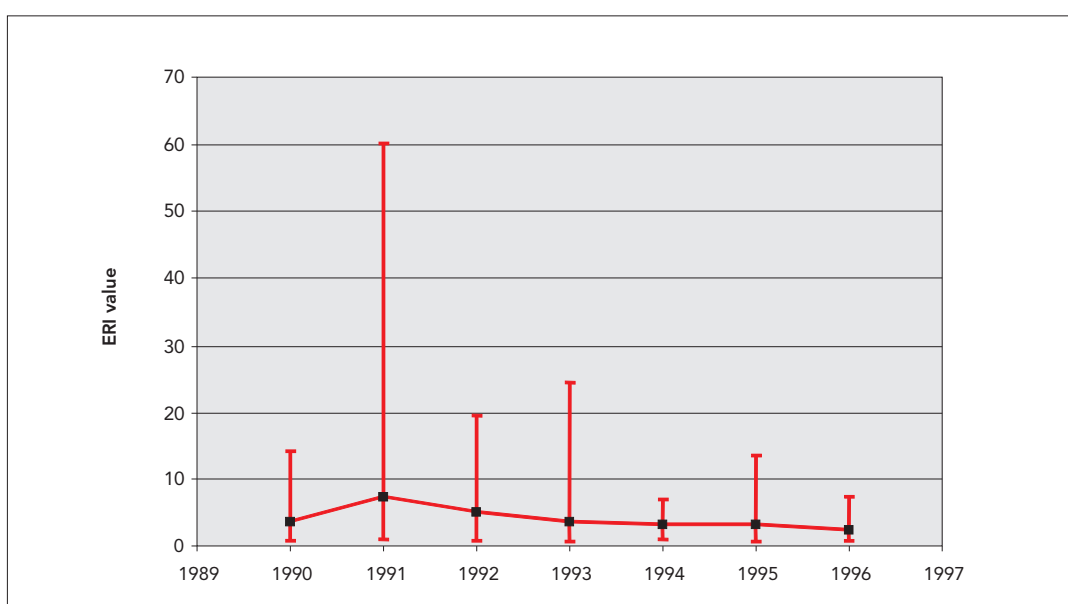
## 3.5.  Results of testing

The median concentrations of the BCR or EAC range for each hazardous substance are used to give the ERI values. Next, for each substance the minimum, the maximum and the arithmetic mean of all ERI values in a year were determined (see Annex 3). The resulting ERI values were independent of the location.

In Figures 3.1 to 3.5, the results of the trend detection are presented separately for each substance. These results are not aggregated further. Principally, assuming no synergetic or antagonistic effects of combinations of hazardous substances in organisms, it is allowed to add up the ERI values for the substances considered. The question arises about how to deal with concentrations below the upper limit of the EAC ranges (ERI < 1), which indicates no potential for environmental effects. Adding up ERI values for different substances each below a value of 1 may result in ERI > 1, suggesting that ecological risk increases significantly for the sum of these substances. We are not sure if this is true, and hence we cannot decide whether to add up fully the ERI values calculated for each substance or to add up only the parts of the ERI values above 1.

| Ecological reference index value for lead (upper limit) in blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 (minimum, average and maximum of median values observed in a year for all locations) | Figure 3.3. |
| --- | --- |



| Ecological reference index value for zinc in blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 (minimum, average and maximum of median values observed in a year for all locations) | Figure 3.4. |
| --- | --- |

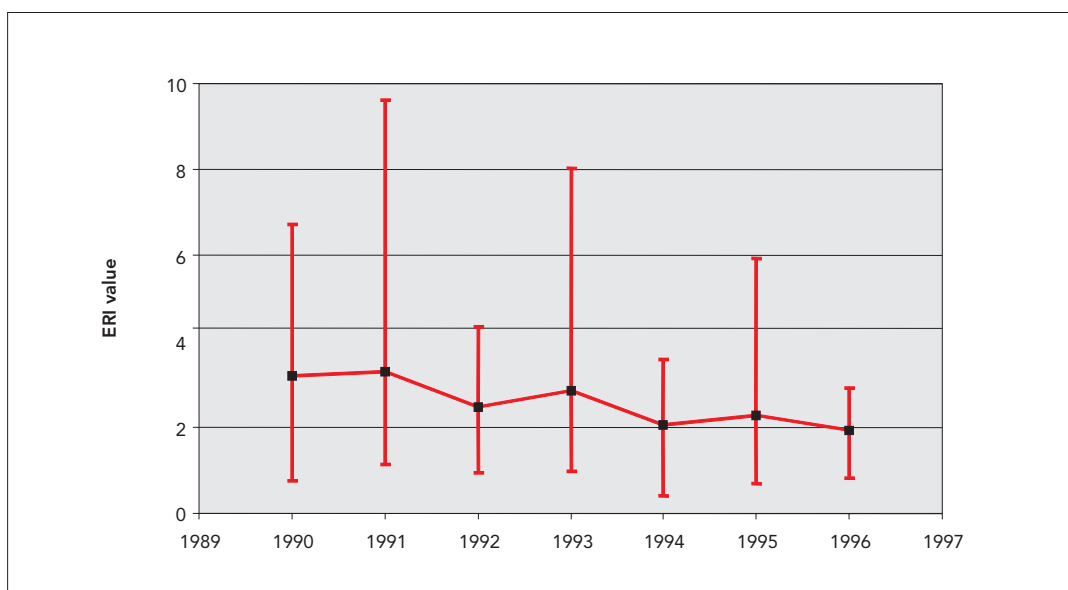| Figure 3.5. | Ecological reference index value for lindane in blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 (minimum, average and maximum of median values observed in a year for all locations) |



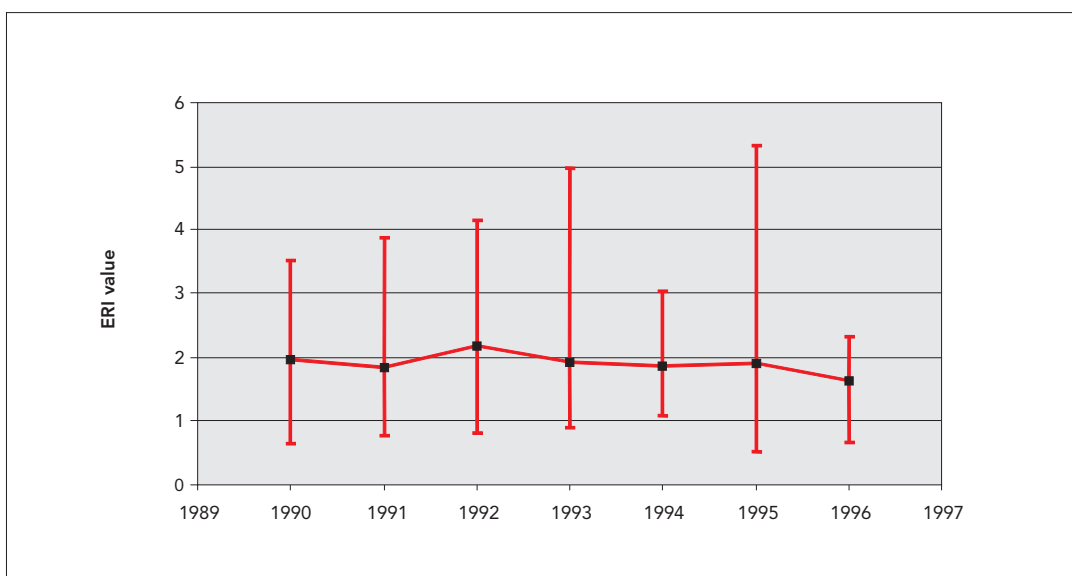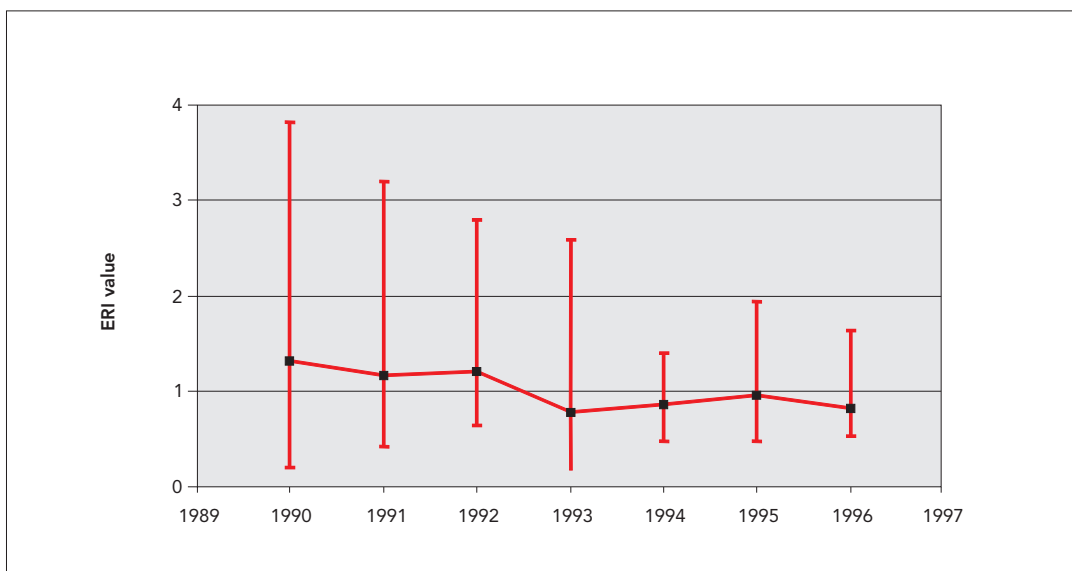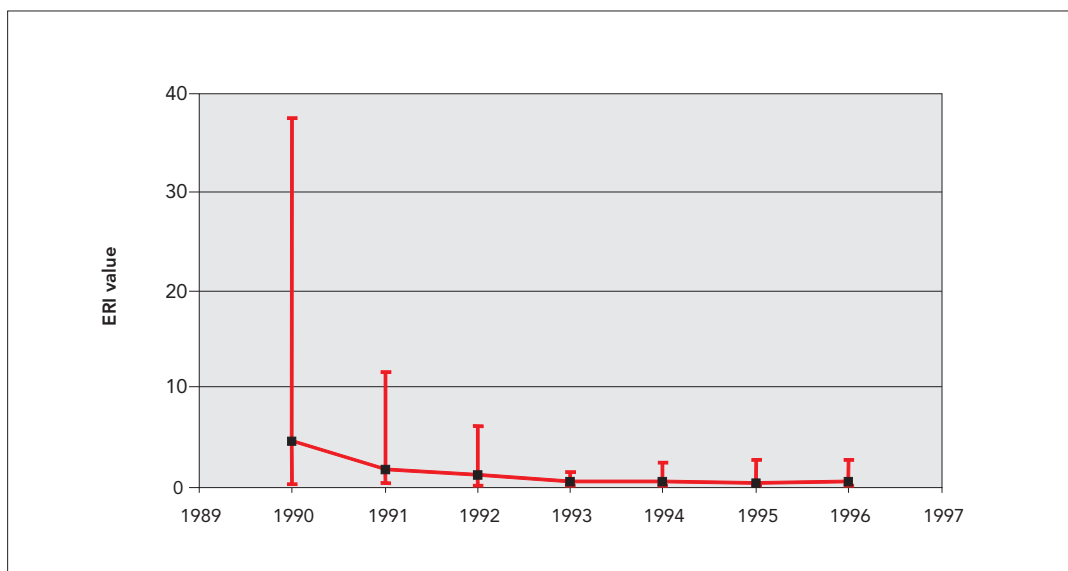Figures 3.1 to 3.5 show that the average ERI values are highest for mercury. Due to a very high maximum ERI value in 1991 (in the Soerfjord in Norway), the average ERI is about 8 in that year. The ERI value for lindane amounts to about 5 in 1990, decreasing after that. Decreasing trends are also found for cadmium and zinc in the period 1990–96, but the rate of decrease is less than for lindane. Lead does not show a clear trend.

The ecological risks of concentrations below an ERI value of 1 are small. In 1993, the average values for zinc and lindane dropped below this limit. For the other three substances, an ERI value as low as 1 was not reached in the period 1990–96.

## 3.6.  Comparison of pressure and state indicators for hazardous substances

The trends detected in concentrations of hazardous substances in the blue mussel in the north-east Atlantic including the North Sea in the period 1990–96 are not quite similar to the trends observed in the inputs of these substances into the same coastal waters (see paragraph 2.4). For instance, lead does not show a clear decreasing trend in the ERI value although the input of lead is clearly going down.

Many explanations may be found for this discrepancy. Trend detection on concentrations in organisms has been performed in a rough way using all data on concentrations of hazardous substances in blue mussels. Also, taking into account the limited reliability (see paragraph 3.3.2) and representativeness of the data (compare, for example, the effect of one extremely high value on the average for mercury), the accuracy of this trend detection is rather low. A time lag between changing inputs into coastal waters and concentrations in blue mussels may also play a role. Also the pressure–state relationship between inputs and concentrations in organisms is probably non-linear. This relationship comprises the cause–effect chain from yearly inputs of hazardous substances to concentrations in coastal waters, then to bioavailability and, finally, to yearly median concentrations in organisms.

## 3.7.  Conclusions and recommendations

### 3.7.1.  Trend detection and robustness of results

From the results presented in paragraph 3.5 we may draw the following conclusions about trends in the ERI values of hazardous substances in the blue mussel in the north-east Atlantic including the North Sea.

*Ecological risk*
☺   The ecological risk of concentrations of hazardous substances in the blue mussel in the north-east Atlantic including the North Sea appears to have decreased in the period 1990–96.

☺ The decrease is most clear for lindane. For cadmium and zinc, the rate of decrease is less than for lindane. Lead does not show a clear trend. Mercury shows a somewhat irregular pattern, due to one extremely high maximum concentration in 1991.

*Robustness of results of trend detection*

☺ The accuracy of the trend detection performed is rather low.

☺ Nevertheless, where large (decreasing) trends are found, the conclusion on the direction of the trend will be relatively firm.

☺ Where relatively small trends are detected, conclusions based on these results are highly speculative.

### 3.7.2. Usefulness of concentrations in the blue mussel as a state indicator

Concentrations of hazardous substances in the blue mussel, expressed in ERI values appear to provide a meaningful indicator. The first results with this indicator look promising and the possibilities to use this indicator should be further explored.

### 3.7.3. Recommendations for improving the concentration indicator

In paragraph 3.3.2, some remarks are made on data reliability. The usefulness of concentrations of hazardous substances in blue mussels as a state indicator may improve significantly when the data set is made complete for each location and is checked for the reliability and comparability of the data.

Much data is available on the concentrations of hazardous substances in organisms. OSPAR/MON made a statistical analysis on time series of concentrations of hazardous substances in organisms. The results were rather disappointing when viewing the number of statistically significant trends detected. From the results of these statistical analyses, we concluded that the blue mussel might be the most promising organism to use for trend detection. It is also widespread in European coastal waters.

Other organisms probably also have indicator potential. Research on conditions to be fulfilled, for example sampling locations in relation to migration patterns, is recommended. Monitoring of organisms without proven indicator potential (and with no importance for human consumption) should preferably be ended to save time and budget for monitoring of organisms that seem more rewarding, unless there are other reasons for the monitoring.

OSPAR (1994) has guidelines on monitoring of biota within its region. However, it is recommended to develop and harmonise monitoring of concentrations of hazardous substances in biota on a European scale (see paragraph 2.5.3) by building upon Inter Regional Forum activities. In developing a European strategy, the results of research on determining promising indicator organisms should be incorporated. Also the geographical scale of collecting data on concentrations in biota should be agreed upon as well as the timescale.

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Web site trend-y-tector (http://www.waterland.net/rikz/osparwg/trend-y-tector).

# Annex 1

| | Sum of direct and riverine inputs of hazardous substances into the north-east Atlantic including the North Sea in the period 1990–98 | | | | | | | | |
|------|---------|---------|------|-------|---------|---------|---------|---------|---------|
| | Average percentage ($^1$) | | | | | | Aggregated values | | |
| | Cadmium | Mercury | Lead | Zinc | Lindane | PCB$_7$ | minimum | maximum | average |
| 1990 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 |
| 1991 | 89.8 | 73.3 | 70.2 | 85.8 | 85.5 | 53.5 | 53.5 | 89.8 | 76.3 |
| 1992 | 88.9 | 80.5 | 66.1 | 88.9 | 64.0 | 36.8 | 36.8 | 88.9 | 70.9 |
| 1993 | 55.6 | 65.7 | 75.4 | 90.3 | 85.6 | 65.2 | 55.6 | 90.3 | 73.0 |
| 1994 | 63.8 | 61.0 | 85.0 | 103.4 | 76.8 | 65.1 | 61.0 | 103.4 | 75.8 |
| 1995 | 67.6 | 53.2 | 76.9 | 114.2 | 84.7 | 58.5 | 53.2 | 114.2 | 75.9 |
| 1996 | 44.1 | 25.7 | 50.3 | 68.9 | 57.5 | 38.2 | 25.7 | 68.9 | 47.5 |
| 1997 | 48.1 | 33.3 | 44.4 | 86.9 | 76.9 | 23.5 | 23.5 | 86.9 | 52.2 |
| 1998 | 52.6 | 33.7 | 66.1 | 82.2 | 63.5 | 48.9 | 33.7 | 82.2 | 57.8 |

($^1$) Total input in 1990 for each substance is 100 %.

# Annex 2

| | Country contributions to direct and riverine inputs of hazardous substances into the north-east Atlantic including the North Sea in the period 1990–98 and in the Baltic Sea in 1995<br>(as % of total input of each substance in 1990) |
|---|---|

| Cadmium | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % | Baltic | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | | | | Denmark | Finland | Germany | Sweden |
| 1990 | 3.9 | 0.5 | 4.3 | 7.8 | 3.1 | 9.5 | 14.9 | 8.3 | - | 0.6 | 47.1 | 100.0 | | | | |
| 1991 | 3.6 | - | 3.0 | 6.6 | 3.1 | 6.4 | 7.6 | 13.8 | - | 0.3 | 45.3 | 89.8 | | | | |
| 1992 | 6.1 | - | 4.1 | 9.7 | 2.1 | 5.2 | 7.3 | 22.0 | - | 0.5 | 31.8 | 88.9 | | | | |
| 1993 | 3.1 | - | 2.9 | 8.4 | 1.6 | 4.3 | 6.6 | 1.6 | 0.1 | 0.1 | 26.9 | 55.6 | | | | |
| 1994 | 3.2 | - | 2.9 | 7.5 | 2.2 | 11.7 | 8.5 | 3.6 | 0.1 | 0.4 | 23.7 | 63.8 | | | | |
| 1995 | 3.5 | - | 2.5 | 6.9 | 2.0 | 24.4 | 8.3 | 0.2 | 0.1 | 0.2 | 19.6 | 67.6 | | | | |
| 1996 | 2.7 | - | 2.0 | 5.2 | 3.6 | 7.7 | 5.4 | - | - | - | 17.5 | 44.1 | | | | |
| 1997 | 2.4 | - | - | 5.8 | 2.3 | 3.7 | 4.8 | 1.2 | 13.3 | 0.4 | 14.1 | 48.1 | | | | |
| 1998 | 2.2 | - | - | 5.7 | 3.2 | 8.9 | 7.1 | 0.6 | 6.2 | 0.5 | 18.1 | 52.6 | | | | |
| Baltic 1995 | | | | | | | | | | | | | 0.40 | 2.19 | 0.17 | 1.22 |

| Mercury | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % | Baltic | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | | | | Denmark | Finland | Germany | Sweden |
| 1990 | 9.9 | 0.3 | 14.0 | 25.9 | 6.0 | 7.8 | 2.1 | 12.0 | - | 0.3 | 21.6 | 100.0 | | | | |
| 1991 | 2.8 | - | 6.6 | 24.6 | 6.0 | 8.1 | 1.2 | 4.3 | - | 0.3 | 19.3 | 73.3 | | | | |
| 1992 | 1.7 | - | 21.4 | 27.1 | 5.9 | 8.0 | 1.3 | 0.1 | - | 0.3 | 14.7 | 80.5 | | | | |
| 1993 | 6.4 | - | 2.0 | 24.7 | 5.9 | 8.5 | 1.2 | 0.6 | - | 0.3 | 16.1 | 65.7 | | | | |
| 1994 | 7.6 | - | 2.7 | 13.9 | 5.9 | 15.7 | 0.9 | 2.1 | - | 0.2 | 11.9 | 61.0 | | | | |
| 1995 | 1.5 | - | 2.0 | 11.4 | 5.9 | 19.8 | 1.3 | 0.9 | - | 0.4 | 10.1 | 53.2 | | | | |
| 1996 | 0.1 | - | 1.2 | 7.2 | - | 8.1 | 1.2 | - | - | - | 8.0 | 25.7 | | | | |
| 1997 | 0.6 | - | - | 4.8 | - | 6.6 | 1.0 | 2.8 | 6.4 | 0.2 | 10.8 | 33.3 | | | | |
| 1998 | 1.5 | - | - | 5.3 | - | 5.3 | 7.0 | 1.8 | 1.0 | 0.3 | 11.5 | 33.7 | | | | |
| Baltic 1995 | | | | | | | | | | | | | 0.80 | 2.43 | 0.26 | 1.22 |

| Lead | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1990 | 1.2 | 0.3 | 6.8 | 9.6 | 2.9 | 15.7 | 5.5 | 30.8 | - | 0.4 | 26.8 | 100.0 |
| 1991 | 1.7 | - | 6.8 | 13.5 | 2.9 | 10.4 | 4.7 | 3.6 | 0.3 | 0.2 | 26.1 | 70.2 |
| 1992 | 1.5 | - | 5.0 | 14.3 | 3.7 | 10.0 | 3.2 | 5.4 | 0.1 | 0.2 | 22.7 | 66.1 |
| 1993 | 1.7 | - | 4.5 | 17.1 | 2.5 | 17.8 | 2.7 | 0.3 | - | 0.1 | 28.6 | 75.4 |
| 1994 | 2.2 | - | 7.7 | 11.2 | 3.6 | 31.8 | 3.7 | 0.3 | 0.1 | 0.5 | 23.9 | 85.0 |
| 1995 | 2.0 | - | 2.5 | 8.4 | 2.9 | 39.5 | 3.7 | 0.1 | 0.1 | 0.4 | 17.4 | 76.9 |
| 1996 | 2.0 | - | 3.0 | 5.7 | 5.2 | 17.3 | 2.8 | - | - | - | 14.1 | 50.3 |
| 1997 | 1.8 | - | - | 6.9 | 5.5 | 10.6 | 2.9 | 1.9 | 0.8 | 0.4 | 13.7 | 44.4 |
| 1998 | 2.3 | - | - | 8.2 | 6.5 | 11.7 | 6.8 | 0.2 | 2.4 | 0.6 | 27.2 | 66.1 |

| Zinc | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % | Baltic | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | | | | Denmark | Finland | Germany | Sweden |
| 1990 | 2.5 | 0.7 | 2.0 | 15.5 | 7.3 | 17.5 | 14.7 | 3.3 | - | 2.1 | 34.3 | 100.0 | | | | |
| 1991 | 3.2 | - | 2.0 | 15.2 | 7.3 | 11.1 | 9.1 | 1.3 | 2.3 | 1.8 | 32.5 | 85.8 | | | | |
| 1992 | 2.2 | - | 2.0 | 22.1 | 7.1 | 11.6 | 7.6 | 0.1 | 0.6 | 1.5 | 34.1 | 88.9 | | | | |
| 1993 | 2.2 | - | 2.0 | 22.0 | 7.5 | 14.6 | 8.3 | 0.6 | 3.7 | 1.4 | 28.0 | 90.3 | | | | |
| 1994 | 2.3 | - | 2.0 | 16.7 | 9.2 | 31.8 | 7.8 | 1.0 | 0.8 | 1.6 | 30.0 | 103.4 | | | | |
| 1995 | 3.8 | - | 2.0 | 13.1 | 12.2 | 48.3 | 8.5 | 0.4 | 0.1 | 1.4 | 24.3 | 114.2 | | | | |
| 1996 | 3.2 | - | 2.7 | 9.5 | 8.2 | 16.9 | 8.8 | - | 1.1 | - | 18.5 | 68.9 | | | | |
| 1997 | 3.4 | - | - | 9.4 | 6.1 | 10.8 | 8.0 | 1.8 | 26.9 | 1.0 | 19.5 | 86.9 | | | | |
| 1998 | 4.4 | - | - | 11.3 | 6.1 | 12.3 | 11.1 | 0.6 | 11.3 | 1.5 | 23.6 | 82.2 | | | | |
| Baltic 1995 | | | | | | | | | | | | | 0.16 | 5.62 | 0.17 | 11.33 |

| Lindane | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1990 | 5.7 | 1.5 | 9.8 | 19.1 | - | 0.9 | 30.0 | - | - | - | 32.9 | 100.0 |
| 1991 | 6.2 | 1.5 | 9.8 | 10.5 | - | 0.4 | 14.6 | - | - | - | 42.5 | 85.5 |
| 1992 | 4.6 | 1.5 | 9.8 | 11.6 | - | 0.6 | 5.5 | - | - | - | 30.4 | 64.0 |
| 1993 | 4.5 | 1.5 | 9.8 | 10.7 | - | 20.2 | 5.1 | 0.2 | - | - | 33.7 | 85.6 |
| 1994 | 4.2 | 1.5 | 9.8 | 14.6 | - | 12.9 | 4.9 | 0.8 | - | - | 28.1 | 76.8 |
| 1995 | 3.1 | 1.5 | 9.8 | 16.3 | - | 20.7 | 6.0 | 0.2 | - | - | 27.1 | 84.7 |
| 1996 | - | - | 5.6 | 13.4 | - | 16.8 | 4.2 | - | - | - | 17.5 | 57.5 |
| 1997 | 3.7 | - | - | 20.7 | - | 16.8 | 4.4 | - | 11.4 | - | 19.8 | 76.9 |
| 1998 | 6.2 | - | - | 14.6 | - | 14.5 | 4.7 | - | 0.8 | - | 22.6 | 63.5 |

| $PCB_7$ | Belgium | Denmark | France | Germany | Ireland | Netherlands | Norway | Portugal | Spain | Sweden | UK | % |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1990 | 1.0 | 1.0 | 3.2 | 4.6 | - | 4.8 | 15.6 | - | - | - | 69.7 | 100.0 |
| 1991 | 0.8 | 1.0 | 5.2 | 2.9 | - | 4.2 | 1.2 | - | - | - | 38.2 | 53.5 |
| 1992 | 0.9 | 1.0 | 4.2 | 1.4 | - | 3.2 | 1.3 | 0.2 | - | - | 24.7 | 36.8 |
| 1993 | 6.8 | 1.0 | 4.2 | 1.7 | - | 4.2 | 0.7 | 0.8 | - | - | 45.8 | 65.2 |
| 1994 | 7.2 | 1.0 | 5.2 | 3.1 | - | 9.7 | 1.8 | 2.7 | - | - | 34.5 | 65.1 |
| 1995 | 1.5 | 1.0 | 4.2 | 4.7 | - | 15.2 | 0.9 | 0.8 | - | - | 30.3 | 58.5 |
| 1996 | - | - | 3.2 | 4.1 | - | 6.5 | 0.5 | - | - | - | 23.9 | 38.2 |
| 1997 | 2.1 | - | - | 3.5 | - | 5.5 | 0.7 | - | - | - | 11.6 | 23.5 |
| 1998 | 4.4 | - | - | 2.8 | - | 5.8 | 0.7 | - | - | - | 35.2 | 48.9 |

# Annex 3

| ERI values of hazardous substances in the blue mussel (aggregated over all locations) | | | | | | | |
|---|---|---|---|---|---|---|---|
| **Cadmium** | **1990** | **1991** | **1992** | **1993** | **1994** | **1995** | **1996** |
| Average | 3.20 | 3.29 | 2.48 | 2.85 | 2.07 | 2.28 | 1.94 |
| Minimum | 0.76 | 1.13 | 0.95 | 1.00 | 0.40 | 0.70 | 0.84 |
| Maximum | 6.73 | 9.62 | 4.35 | 8.02 | 3.58 | 5.93 | 2.91 |
| | | | | | | | |
| **Mercury** | **1990** | **1991** | **1992** | **1993** | **1994** | **1995** | **1996** |
| Average | 3.68 | 7.45 | 5.14 | 3.75 | 3.28 | 3.38 | 2.41 |
| Minimum | 0.75 | 0.95 | 0.79 | 0.64 | 0.96 | 0.67 | 0.77 |
| Maximum | 14.20 | 60.20 | 19.52 | 24.40 | 7.04 | 13.58 | 7.30 |
| | | | | | | | |
| **Lead** | **1990** | **1991** | **1992** | **1993** | **1994** | **1995** | **1996** |
| Average | 1.96 | 1.84 | 2.18 | 1.93 | 1.86 | 1.90 | 1.64 |
| Minimum | 0.66 | 0.78 | 0.83 | 0.90 | 1.09 | 0.53 | 0.66 |
| Maximum | 3.52 | 3.87 | 4.16 | 4.98 | 3.03 | 5.33 | 2.33 |
| | | | | | | | |
| **Zinc** | **1990** | **1991** | **1992** | **1993** | **1994** | **1995** | **1996** |
| Average | 1.32 | 1.17 | 1.22 | 0.79 | 0.87 | 0.96 | 0.82 |
| Minimum | 0.21 | 0.43 | 0.64 | 0.17 | 0.48 | 0.49 | 0.54 |
| Maximum | 3.81 | 3.19 | 2.79 | 2.59 | 1.41 | 1.93 | 1.64 |
| | | | | | | | |
| **Lindane** | **1990** | **1991** | **1992** | **1993** | **1994** | **1995** | **1996** |
| Average | 4.61 | 1.76 | 1.25 | 0.59 | 0.54 | 0.46 | 0.62 |
| Minimum | 0.26 | 0.36 | 0.17 | 0.19 | 0.12 | 0.12 | 0.11 |
| Maximum | 37.50 | 11.75 | 6.25 | 1.55 | 2.50 | 2.75 | 2.75 |