chapter

4

Chemistry

4.1 Introduction

In this chapter, the status, spatial distribution and temporal trends of selected compounds and related measures are discussed.

Data were either extracted from relevant OSPAR documents, directly or indirectly from the ICES database as supplied by the Contracting Parties, or taken from the open literature. Priority was given to data produced after 1993, data relevant for establishing spatial distribution and temporal trends and to data relevant for estimating budgets and fluxes.

The mere fact that concentrations are reported at particular locations does not mean that they are of particular concern at these locations. At the same time, data are often unavailable for several areas where the substances under consideration most probably would be present. The compound(s) may simply not have been measured (or reported). This often leads to a gap in information on levels of contamination for the entire North Sea area, and consequently for the calculation of fluxes.

As a general rule, quality assurance has, over the last two decades, been of increasing concern in the process of data acquisition and screening. The improvement of the quality of data is demonstrated, for example, by the decreasing number of outliers in time trends. The possibility that the early data sets may have been ‘contaminated’ by poorer quality data can not be excluded. This cautionary remark is valid for all trace elements (for the period before 1985), and for all trace organic compounds before about 1990. This should be kept in mind when comparing data from different time periods.

4.2 Input of chemical substances in general

In the following text, ‘input’ refers to the load of substances reaching the estuarine or marine domain. Waterborne inputs are either land-based, occurring at the land/sea interface (riverine and direct inputs), or sea-based, occurring within the marine area itself (from shipping, oil/gas exploration and exploitation, etc.). Airborne inputs at the atmosphere/water interface result from both dry and wet deposition. ‘Emission’ refers to the release of substances to the atmosphere, while discharges and losses refer to releases into inland waters from point or diffuse sources.

Data from the Comprehensive Atmospheric Monitoring Programme (CAMP) have been used together with data from the Riverine Inputs and direct Discharges programme (RID), in order to establish for the first time an integrated assessment of the atmospheric and waterborne inputs to the North Sea for the period 1990–6 (OSPAR, 1998c).

4.2.1 Land based inputs

Input data such as those collected by the RID programme under the OSPAR Joint Assessment and Monitoring Programme (JAMP) are collected in order to assess, as accurately as possible on an annual basis, all riverine and direct inputs of selected substances from land-based sources to the Convention waters (OSPAR, 1998b). The commitment is to aim for monitoring coverage of at least 90% of the total waterborne input.

Riverine input is the load conveyed by a river at the point of entry to the marine area, which is usually a point of unidirectional freshwater flow immediately upstream of any tidal influence. When loads are estimated at a point situated in the tidal zone, this can lead to some inconsistencies between these estimates and the actual riverine inputs. It is also important to realise that the riverine loads reported represent the loads originating from the entire river catchment area. In the case of trans-national rivers, loads from upstream countries are ascribed to the country furthest downstream.

Direct input is any aquatic input to a river or estuary downstream of the riverine monitoring point or directly into coastal waters.

Estimates of land-based inputs are based on load calculations using river flows and concentrations measured at various (one per river) monitoring stations. When riverine inputs are calculated from measurements conducted upstream of the freshwater limit (gross inputs), processes downstream of the measuring point are not taken into account although they can change the net input considerably. These processes of a physical, chemical or biological nature (flocculation, sedimentation, evaporation, biodegradation, denitrification, etc.) will need to be more accurately described to improve understanding of the riverine inputs to the marine environment. However, though the data established under the RID programme are sometimes incomplete due to gaps in reporting, they provide a useful indication of the orders of magnitude of the inputs. *Table 4.1* provides a summary of direct and riverine inputs to the Greater North Sea.

4.2.2 Sea-based inputs

Inputs from sea-based activities have their origin in particular in shipping, the offshore oil and gas industry, mariculture, and the disposal of dredged material. Some of these inputs may also be of an accidental nature.

Of particular importance is the dumping of dredged material. Due to the huge quantities involved, the loads of contaminants (and nutrients) can be very significant even when the concentrations are low. However, dredged material from harbour areas, estuaries or navigation

Table 4.1 Summary of direct and riverine inputs to the Greater North Sea in 1996. Source of data: OSPAR (1998a).

North Sea Area	Cd (t)	Hg (t)	Cu (t)	Pb (t)	Zn (t)	γ-HCH (kg)	PCBs* (kg)	NH_4-N (10^3 t)
Kattegat								
(lower estimate)			23	7.0	70			
(upper estimate)			23	7.0	704			
Skagerrak								
(lower estimate)	3.1	0.1	104	26	511	34	0	6.5
(upper estimate)	3.2	0.1	104	26	511	34	10	6.5
North Sea (main body)								
(lower estimate)	19	7.2	778	686	4554	690	285	75
(upper estimate)	33	8.4	812	733	4679	735	959	78
Channel								
(lower estimate)	2.4	0.5	151	81	475	135	95	21
(upper estimate)	2.4	0.5	151	81	475	143	97	21

* ΣPCB_7 (sum of IUPAC Nos 28, 52, 101, 118, 153, 138, 180.)

channels may already be accounted for in the land-based inputs. When material is dredged and dumped in the marine area, it concerns merely a remobilisation and relocation of contaminants and nutrients already present. Dredged sediments with low contamination levels have limited anthropogenic influence.

4.2.3 Atmospheric inputs and long range transport and deposition

For a number of contaminants, the airborne inputs constitute an important or even the predominant contribution. This is the case for some heavy metals (especially mercury and lead), persistent organic substances and several nitrogen compounds.

Most substances have a rather short residence time in the atmosphere (in the order of a few days), and they are rapidly deposited by rain (wet deposition) or through sedimentation (dry deposition). The general pattern over the North Sea shows a clear decrease in deposition levels further from the coast and towards open water. The southern regions of the North Sea, close to the industrialised areas, are exposed to particularly high levels of atmospheric deposition. The limited residence time implies that the contribution of more distant, non-OSPAR countries within Europe to atmospheric deposition into the North Sea is small (a few % for heavy metals, 25% for total inorganic nitrogen (OSPAR, 1998c)).

In contrast, some contaminants, such as polychlorinated biphenyls (PCBs), exhibit very long residence times and their atmospheric transport must be considered on a larger, even global, scale.

4.2.4 Transport and fate of contaminants

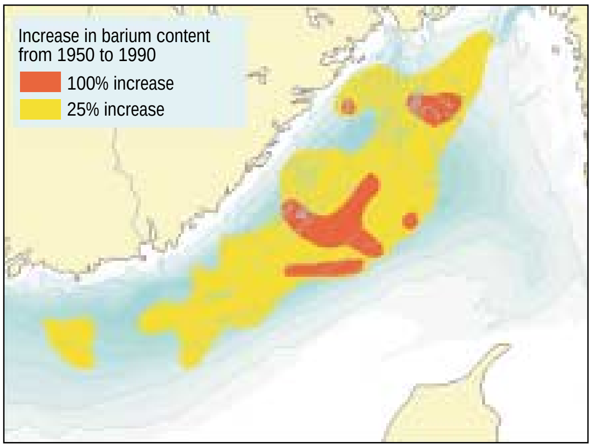
The fate of contaminants in the North Sea is closely linked to their distribution between the dissolved and the particulate phase. In general, dissolved substances will follow the movements of the respective water masses (see Chapter 2 and *Figure 2.3*).

Due to their strong association with particulate matter, the fate of many contaminants in the Greater North Sea is, however, closely related to the transport of (re-)suspended solids. In areas of deposition, sediments are loaded with contaminated particles, and it is estimated that 70% of the substances associated with particulate matter remain trapped in sedimentation areas such as estuaries, the Wadden Sea, the deepest part of the Kattegat-Skagerrak area, and the Norwegian Trench (Laane *et al*., 1998). Barium accumulation in the sediments of the Skagerrak over recent decades *(Figure 4.1)* is an example of transport and deposition of solid material over large areas (Barite, a barium mineral, is a primary component of drilling muds used in the offshore oil industry). Deposited particles will be subject to (bio-)turbation and to resuspension, processes that enhance the release of contaminants. Eventually (contaminated) particles may be subject to burial into deeper layers. There is an ongoing discussion on the importance of these processes, and how to quantify the source and sink functions of sediments.

4.2.5 Persistent contaminants in the food chain

Bioconcentration is the net result of uptake, distribution and elimination of a substance in an organism. It is a key process that influences the fate of persistent and hydrophobic organic contaminants and determines their effect on consumers higher up in the food chain. As a result, such substances (e.g. heavy metals, some organochlorines) are transferred throughout the entire food chain, thus showing highest concentrations in top predators, including man.

Figure 4.1 **Increase of barium content in Skagerrak sediments from about 1950 to about 1990. Source: Longva and Thorsnes (1997).**



NO_3-N (10^3 t)	PO_4-P (10^3 t)	Total N (10^3 t)	Total P (10^3 t)	SPM (10^3 t)
26		41	1.0	
26		41	1.0	
18	0.3	37	1.0	272
18	0.3	37	1.0	272
499	30	729	53	8376
505	32	741	54	8523
87	20	109	10	550
87	20	109	10	550

Identifying and quantifying the most important exposure pathways is rather complex since many factors are involved, including the type of chemical compound, the species of organism and its age, size, feeding habits and individual history.

4.2.6 Quantification of contaminants entering the sea

Riverine inputs are estimated from concentrations and flow rate. In cases where the results recorded are lower than the detection limit of the analytical method, two load estimates (upper and lower values) have been used. 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 limit of detection, and previous experience has shown that in such a case the upper estimate tends to be unrealistically high.

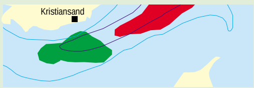
Estimates of atmospheric inputs to the North Sea were based mainly on the results of the CAMP programme. The estimates of wet and dry deposition of nitrogen compounds and heavy metals to the North Sea follow the procedure adopted by the Paris Commission in 1994. In this procedure, the data on contaminant concentrations reported by CAMP coastal measuring stations surrounding the North Sea and the results of long-range transport models are combined. Another approach makes use of models and emission data to estimate atmospheric inputs to the North Sea. For example, heavy metal deposition (excluding mercury) has been estimated using the European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990 (Berdowski *et al*., 1997) coupled to the Eutrend atmospheric transport model (Van Pul *et al*., 1998).

4.2.7 Time lag between reductions of emissions, discharges and losses and reductions observed in the field

For a number of contaminants, significant reductions in the emissions to the air and discharges to water have been achieved. However, these achievements have not produced an equivalent decrease in the riverine inputs. This situation is partly due to variations in run-off and suspended matter concentration during the period, but it also raises the following two questions: is the contribution of diffuse sources correctly accounted for, owing to the uncertainties in estimating them? to what extent is the mobilisation of contaminants, accumulated over the years in soils, sediments and other land reservoirs (such as flood plains), a possible long-term source of waterborne contaminants? It also highlights that the effectiveness of reduction measures on the actual load reaching the North Sea can only be assessed on a long-term basis.

Similarly, a reduction of contaminant concentrations in the marine system equivalent to the one achieved for emissions, discharges and losses has not been observed, particularly for sediments and biota. A variety of models have been developed to assess contaminant transport and fate in the North Sea (OSPAR, 1998d). In a model simulation based on a 50% reduction of the cadmium load from rivers and atmospheric deposition, a decrease of less than 15% in environmental concentrations was obtained at seven locations throughout the North Sea *(Table 4.2)*. The largest decreases in concentration were found in the coastal zones where river discharges are the dominant source of contaminants. Models indicate the smallest decreases (1 – 4%) occurring in the central and northern North Sea, where river influence is small and Atlantic inflow over the boundaries is the major source. In general, water flowing into the Greater North Sea from the Atlantic has a major influence on the amount of trace metals and nutrients in the North Sea because of the large volume involved.

Table 4.2 Results of reduction scenario for cadmium. Source: OSPAR (1998d).

	Location (see map)	Model 1	Model 2	Model 3	Model 4	Model 5
	UK coast (1)	6	1	4	1	6
	Dutch coast (2)	8	8	12	16	13
	South Central North Sea (3)	6	3	6	3	9
	German Bight (4)	9	4	8	6	8
	Central North Sea (5)	1	2	1	1	4
	German Bight North (6)	9	5	8	7	10
	UK Coast –Southwestern North Sea (7)	3	4	4	1	4

Model simulation of a 50% reduction of the riverine cadmium input (relative to 1989)

The figures given are in % reduction in the seawater dissolved Cd concentration

Model 1	BSHdmod.E (BSH Germany)
Model 2	NORWECOM (IMR Norway)
Model 3	NOSTRADAMUS (SOC United Kingdom)
Models 4 and 5	SCREMETOX and ZeeBOS-TOX (Rijkswaterstaat – Delft Hydraulics, The Netherlands)

4.2.8 Concentrations and Trends

Concentrations of substances in sediments are generally reported in relation to dry weight (dw) and concentrations in biota in relation to wet weight (ww), dry weight or fat weight (fw).

Trends for contaminants in biota were calculated for time series of minimum 4 years and based on a 95% level of confidence (OSPAR, 2000).

4.3 Background/reference values in water, sediments and biota and assessment tools

To help in the assessment of the ecological impact of contaminants, Background/Reference Concentrations (BRCs) and Ecotoxicological Assessment Criteria (EACs) have been developed at OSPAR workshops. In 1997 OSPAR endorsed BRCs and EACs for a number of substances together with guidance for their use.

Some of these BRC or EAC values are firm while others are still preliminary, but whenever available, they have been related to the observed concentrations.

4.3.1 Background/Reference Concentrations

Naturally occurring substances are defined as all substances produced from natural precursors by biosynthesis, geochemical or (photo) chemical processes. The BRC for naturally occurring substances is defined as the concentration that could be found in the environment in the absence of any human activity. This means that for anthropogenic substances the natural background concentration should, in theory, be zero, but due to historic contamination (e.g. as in the case of PCBs) the adopted BRC may have a (reference) value above zero. BRCs were defined preferably from data from geological or historical times and/or from pristine regions found within the Convention Area. BRCs (if available) for trace metals and trace organic compounds in water, sediment and biota (mussel tissue) are summarised in *Tables 4.3*, *4.4* and *4.5*.

Table 4.3 Ranges in the background concentrations of cadmium, mercury, lead and copper within the OSPAR maritime area. Source: OSPAR (1997b).

	Sediments relative to Al (*10^{-4})	Seawater (ng/kg)	Blue mussel (mg/kg ww)
Cd	0.007 – 0.030	8 – 25	0.07 – 0.11
Hg	0.0034 – 0.0066	0.2 – 0.5	0.005 – 0.010
Pb	1.8 – 4.0	10 – 20	0.01 – 0.19
Cu	2.2 – 5.7	50 – 360	550.76 – 1.10

Assessment of nutrient data is difficult, because few reliable data exist from times of low anthropogenic influence, i.e. before 1950. *Table 4.6* provides some of these data on winter concentrations on a local basis. However, OSPAR has not endorsed these values as BRCs.

Background concentrations of various natural radionuclides are provided in section 4.8.

4.3.2 Ecotoxicological Assessment Criteria

EACs were established for assessing chemical monitoring data from the OSPAR Convention area. EACs are the concentrations of specific substances in the marine environment below which no harm to the environment or biota is expected. Criteria for the specific contaminants were derived using all the available ecotoxicological data that passed predefined quality criteria (OSPAR, 1996).

Table 4.4 Background concentrations of selected polycyclic aromatic hydrocarbons (PAHs) in surface sediments from remote areas and in surface water for application in specific regions of the North Sea. Source: OSPAR (1997b).

	Sediments (ng/g dw)		Water (ng/l)	
	Northern North Sea/Skagerrak	Southern North Sea	Northern North Sea	Central and Southern North Sea
Benzo[*a*]pyrene	8.8 – 112	< 0.2 – 51	0.002 – 0.005	0.002 – 0.004
Fluoranthene	14 – 160	0.72 – 97	0.073 – 0.285	0.104 – 0.264
Benzo[*b* + *k*]fluoranthene	46 – 434	1.1 – 142	–	–
Benzo[*b*]fluoranthene	–	–	0.004 – 0.017	0.003 – 0.009
Indeno[*1,2,3*]pyrene	43 – 212	< 0.2 – 70	0.004 – 0.017	0.006 – 0.012

Table 4.5 Ranges of background/reference concentrations of hexachlorobenzene (HCB), pp'–dichlorodiphenyldichloroethene (DDE) and selected polychlorinated biphenyls (PCBs) in surface sediments (pg/g dw) of selected areas from the North Sea. Source: OSPAR (1997b).

	HCB	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180	DDE-pp
South Norway/Skagerrak	70	31	32	62	90	116	60	66
German Bight	910	680	260	730	1650	1200	600	510

Table 4.6 **Ranges in the pre-1950 background winter concentrations of dissolved phosphate, nitrite, nitrate and silicate (µmol/l, winter values) in different areas of the North Sea. Source of data: OSPAR (1997j).**

	Salinity	PO_4	SiO_4	NO_2	NO_3
Strait of Dover	34.5 – 35.2	0.42 – 0.57	2.5 – 7.0	0.1 – 0.9	4.8 – 11.1
Netherlands coast	31 – 33	0.48 – 0.90	9 – 21	0.05 – 0.70	19.5 – 32.2
German Bight	31 – 33	0.43 – 0.60	9 – 17	0.4 – 1.2	–
Southern Bight	> 34.5	0.25 – 0.70	2.5 – 10.5	0.03 – 15.4	1.5 – 10
Dogger Bank	–	0.35 – 0.80	2.5 – 7.1	< 0.6	–

Table 4.7 **Overview of ecotoxicological assessment criteria. Source: OSPAR (1997b).**

	Water (mg/l)	Sediment (mg/kg dw)	Mussel (mg/kg dw)
Trace metals			
As	1 – 10*	1 – 10†	nr
Cd	0.01 – 0.1*	0.1 – 1†	fc
Cr	1 – 10*	10 – 100†	nr
Cu	0.005 – 0.05*‡	5 – 50†	fc
Hg	0.005 – 0.05*	0.05 – 0.5†	fc
Ni	0.1 – 1†	5 – 50†	nr
Pb	0.5 – 5*	5 – 50†	fc
Zn	0.5 – 5*	50 – 500†	nr
Organochlorine pesticides			
DDE	nr	0.0005 – 0.005†	0.005 – 0.05*
Dieldrine	nr	0.0005 – 0.005†	0.005 – 0.05*
Lindane	0.0005 – 0.005*	nr	nr
PAHs			
Naphthalene	5 – 50*	0.05 – 0.5*	0.5 – 5†
Phenanthrene	0.5 – 5†	0.1 – 1*	5 – 50†
Anthracene	0.001 – 0.01†	0.05 – 0.5*	0.005 – 0.05†
Fluoranthene	0.01 – 0.1†	0.5 – 5†	1 – 10†
Pyrene	0.05 – 0.5†	0.05 – 0.5†	1 – 10†
Benzo[*a*]anthracene	nd	0.1 – 1†	nd
Chrysene	nd	0.1 – 1†	nd
Benzo[*a*]fluoranthene	nd	nd	nd
Benzo[*a*]pyrene	0.01 – 0.1†	0.1 – 1†	5 – 50†
Benzo[*ghi*]perylene	nd	nd	nd
Indeno[1,2,3-*cd*]pyrene	nd	nd	nd
$\sum PCB_7$	nr	0.001 – 0.01†	0.005 – 0.058
TBT	0.00001 – 0.0001*	0.000005 – 0.00005†	0.001 – 0.01*

* firm; † provisional; ‡ this bracket is within the range of background values for natural waters. This value should be compared with the bioavailable fraction in sea water; fc for future consideration; nr not relevant to the current monitoring programme; nd no data available or insufficient data available.

Cautionary note: These assessment criteria have no legal significance and should only be used for the preliminary assessment of JMP/JAMP chemical monitoring data with the aim of identifying potential areas of concern. When applied, the fact whether an EAC is firm or provisional should be taken into account.

Objectives for these assessment criteria should be, in general, to identify possible areas of concern, to indicate which substances could be considered a priority, to determine whether effects on biota are likely to occur. EACs for selected trace metals, PCBs, PAHs, tributyltin (TBT) and some organochlorine pesticides in water, sediment and biota (mussel tissue) are given in *Table 4.7*.

Although based on laboratory toxicity tests, usually employing freshwater organisms, these reference values were established for use as the best available assessment criteria. Levels below these values suggest that no harm to the marine environment should be expected. However, caution should be exercised in using generic, particularly provisional, assessment criteria in specific situations since their use does not preclude the use of common sense and expert judgement with regard to natural concentrations. Furthermore, the EACs do not take into account specific long-term biological effects such as carcinogenicity, genotoxicity and endocrine disruption, or the combined effects of substances.

4.3.3 Trend detection of data sets

Trends for contaminants in biota were calculated for time series of at least 4 years and based on a 95% level of confidence (OSPAR, 2000). It is worthwhile noting that, despite the large number of these time series available, only few data sets revealed significant trends (e.g. 5 – 10% in the case of metals). The large number of non-significant trends might originate from too rigorous statistical requirements, from the fact that most time series are still too short to reveal reliable information on trends, from a high natural variability of contaminant levels in the organisms monitored, or from insufficient sampling frequency.

4.4 **Heavy metals**

4.4.1 Fluxes and transport routes of heavy metals

Heavy metals reach the North Sea via both airborne and waterborne inputs. Inputs are also generated by some sea-based activities, such as exploitation of offshore resources and dumping of dredged materials.

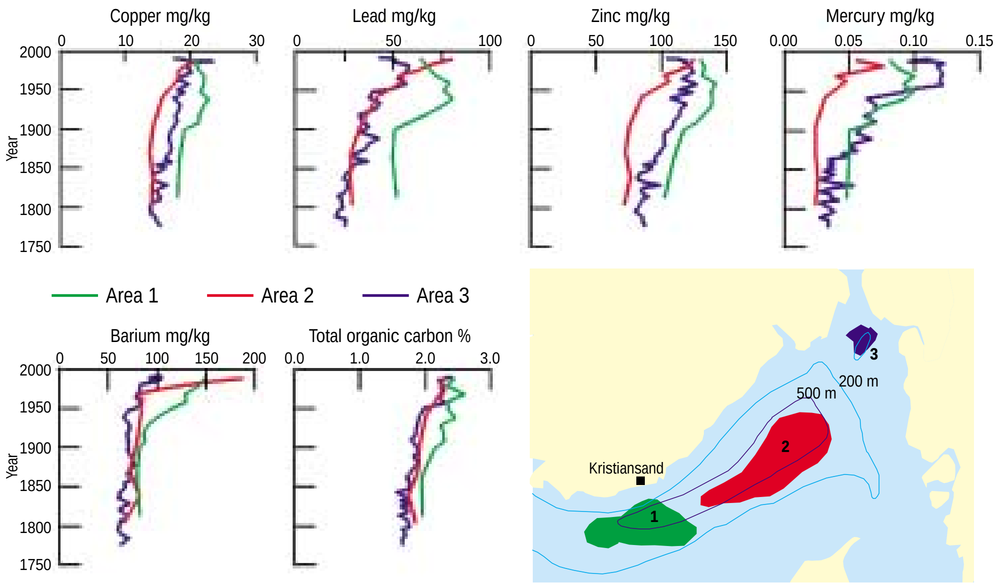
Typical atmospheric deposition levels at around 500 km from the coasts are an order of magnitude lower than deposition into coastal waters close to industrialised areas.

The riverine fluxes of metals consist of particulate and dissolved species. The particulate phase is predominant for most elements. The spatial distribution of metals in the coastal zones, where highest concentrations are found near fresh water outlets and lowest in the open sea, suggests that rivers are major sources of trace metals. However, the dumping of dredged material can disturb this pattern locally.

A decline in the concentration of various metals has generally been observed in the surface sediments in dynamic sediment zones. This is due to mixing with less contaminated suspended matter or the winnowing of contaminated particles.

The recent history of heavy metal accumulation can be observed in (net) deposition areas, such as the Skagerrak and Norwegian Trench. In the deepest part of the Skagerrak (area 2 in *Figure 4.2*), mercury concentrations in sediments have increased significantly since about 1950, while increases in lead and copper concentrations began somewhat earlier, around 1900. Apart from the changes in the contaminant load to these areas over recent decades, a number of other factors have influenced the observed pattern, including variations in sediment transport and erosion rates, and eutrophication and environmental changes in the deeper part of the Skagerrak.

Figure 4.2 **Stratigraphic plot of heavy metals concentration in sediments of the Skagerrak – Norwegian Trench Area. Source: Longva and Thorsnes (1997).**



4.4.2 Cadmium (Cd)

Sources and input estimates

During the period 1990–6, the riverine inputs of cadmium did not show a discernible change, but the direct inputs (which only contribute about 5% to the total waterborne inputs) have decreased markedly *(Figure 4.3)*. *Table 4.8* gives examples of the proportion of riverine and direct inputs, and the difference between lower and upper estimates in two different years. The total cadmium input did not follow a consistent trend *(Figure 4.3)*, mainly because of variations in runoff and suspended particulate matter concentrations. In the Skagerrak area, cadmium inputs remained in the 2 – 5 t/yr range. Inputs to the Wadden Sea by the rivers Elbe and Ems significantly decreased between 1985 and 1995 (Bakker *et al*., 1999). For 1992, the input into the German Bight was estimated to be about 10 t, with a contribution of 6.2, 3.4 and 0.33 t from the Elbe, Weser and Ems, respectively (Radach and Heyer, 1997).

Atmospheric deposition of cadmium is responsible for about one third of the total cadmium input into the Greater North Sea. Wet deposition at coastal stations has shown a decrease of around 50% between the late 1980s and the early 1990s. No significant decrease of cadmium input has been observed for dry deposition. The total atmospheric emission and deposition was halved between 1987 and 1995 (*Figure 4.4* and *Table 4.9*). Model computations indicate that the Southern Bight of the North Sea receives more than 50% of the atmospheric inputs (Van Pul *et al*., 1998). It is also estimated that 94% of the atmospheric emissions reaching the North Sea originate from OSPAR Contracting Parties.

Figure 4.3 **Total waterborne cadmium input to the Greater North Sea (t/yr). Source of data: lower estimates of riverine and direct inputs in OSPAR (1998c).**

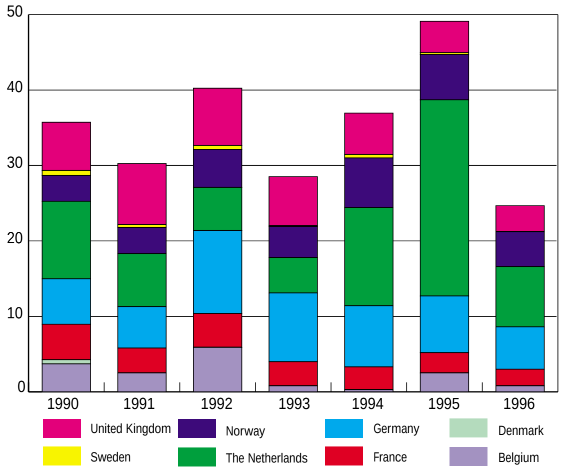


Table 4.8 Riverine and direct inputs of cadmium to the Greater North Sea in 1990 and 1996. Source of data: OSPAR (1998c).

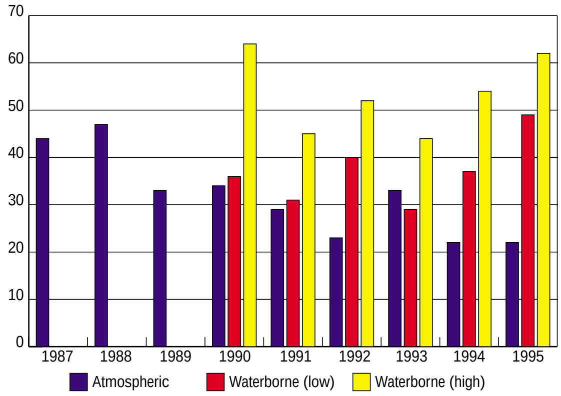
	Riverine inputs (t)		Direct inputs (t)	
	1990	1996	1990	1996
Belgium	3.7* 4.9**	0.8* 5.0**	-	-
Denmark	0.48	n.i.	0.085	n.i.
France	4.7	2.2	n.i.	n.i.
Germany	6.0*§ 11.0**§	5.6§	0.02* 0.06**	0.02* 0.07**
Netherlands	9.8§	8.2§	0.5	0.2
Norway	2.7* 15.6**	3.5* 3.8**	0.7	1.1
Sweden	0.57	0.24	0.1	0.021
United Kingdom	2.7* 7.7**	2.3* 11.4**	3.7* 7.6**	1.16* 1.46**
TOTAL‡				
Lower estimate*	31	23	5.1	2.5
Upper estimate**	55	36	9.1	2.8

§ Including loads from countries upstream.
‡ Total rounded to two significant digits.
* Lower estimate: for concentrations less than the detection limit, a value of ‘0’ was used when calculating loads.
** Upper estimate: for concentrations less than the detection limit, the value of the detection limit was used when calculating loads;
n.i. no information.

Note: Some 1990 data updated by OSPAR may differ from those reported in the 1993 QSR.

Five North Sea numerical models of the transport and fate of riverine contaminants were compared (OSPAR, 1998d). For the year 1989, model results for dissolved cadmium were compared with field measurements from the NERC North Sea Project (NSP) and total cadmium fluxes through the North Sea were calculated *(Table 4.10)*. For dissolved cadmium the modelled concentrations were quite similar (in the range of 0.01 – 0.03 µg/l) but consistently lower than the measured field concentrations, indicating possible underestimation of cadmium boundary conditions for the models. There are differences between mass balances provided by the models. However, all models calculated that the overall riverine cadmium load is of the same order of magnitude as atmospheric inputs but about one order of magnitude

Figure 4.4 **Estimates of cadmium inputs to the North Sea (t/yr). Source: OSPAR (1998c).**



less than the mass inflow from outside the region with Atlantic water.

Ranges of cadmium concentrations are summarised in *Figure 4.5*.

Concentrations in water

Scholten *et al*. (1998) compared the overall mean concentrations of dissolved cadmium in the entire North Sea for 1982–5 and 1986–90, and found a major reduction in concentrations of this metal (50%), although improvements in analytical procedures may interfere with the proper estimation of this trend. Large decreases were observed, especially in the Southern Bight, and also in the Dutch coastal zone, the Thames estuary and the Dogger Bank area.

Concentrations in sediments

In the Dutch coastal zone, the cadmium content has significantly decreased in the areas with previously highest concentrations (Laane *et al*., 1998). Between 1981 and 1996, the median concentration fell by 71% in the area north of the mouth of the Rhine and Meuse rivers. South of this mouth, the median concentration fell by 45%. No significant decrease was found 20 – 70 km offshore in the open sea.

In the Scheldt estuary, a very strong upstream gradient was still measured between Vlissingen and Antwerp, but the maximum concentration decreased by a factor of 3 between 1990 and 1995 (Vyncke *et al*., 1997). Between 1988 and 1993, cadmium concentration in the sediments of the Wadden Sea generally decreased by 10-40% (Bakker *et al*., 1996).

Concentrations in biota

Based on data up to 1996, 65 time series of up to 15 years length were assessed for the Greater North Sea

Table 4.9 Atmospheric emissions of selected heavy metals (t/yr). Source: 4NSC (1995).

Country	Cadmium			Mercury			Lead			Copper		
	1985	1990	1995(+)	1985	1990	1995(+)	1985	1990	1995(+)	1985	1990	1995(+)
Belgium (*)	12.8	6.8	5.4	14.1	9.5	8.0	1707	794	605	157	99	87
Denmark (#)	> 2.2	0.1	–	> 3.3	0.5	–	305	34	–	> 15	5	–
France		no data			no data			no data			no data	
Germany (#)	37.0	10.0	<10	37.0	28.0	<28	5004	838	<838	95	55	<55
The Netherlands (*)	4.3	–	1.9	11.5	–	5.4	1334	–	219	67	–	65
Norway(*)	1.0	0.4	0.4	1.6	0.4	0.4	417	109	10	17	2	2
Sweden (#)	1.0	0.4	0.3	0.9	0.3	0.3	110	25	40	14	5	5
Switzerland (#)	4.7	2.9	1.9		no data		680	440	260		no data	
United Kingdom (#, §)	78	73	59	29.3	27.9	12.8	8120	3190	1320	676	633	574
TOTAL (†)	140	94	79	98	67	55	18000	5400	3300	1000	800	780

* point and diffuse sources; # point sources; + projected values; § min. values; † rounded to two significant digits.

Table 4.10 Modelled mass balance for cadmium in the North Sea (t/yr). Source: OSPAR (1998d).

	Model 1	Model 2	Model 3	Model 4	Model 5
Mass In					
Straight of Dover	52	100	66	83	85
Rivers	12	16	17	13	27
Atmospheric deposition	20	10	13	10	9
Offshore		0	0	2	0
Total Mass In	104	126	96	108	121
Mass Out					
N. Boundary (net) (56°N)	70	40	90	93	119
Sedimentation (net)	0	83	9	14	19
Total Mass Out	70	123	99	106	138
NET (in - out)	34	3	-3	1	-17

Model 1	BSHdmod.E (BSH Germany)
Model 2	NORWECOM (IMR Norway)
Model 3	NOSTRADAMUS (SOC United Kingdom)
Models 4 and 5	SCREMETOX and ZeeBOS-TOX (Rijkswaterstaat – Delft Hydraulics, The Netherlands)

area (OSPAR, 2000). No upward trends were found anywhere, but four significant downward trends were found in blue mussels from the Netherlands (Western Scheldt and Ems-Dollard area) and Norway (Sørfjord and Hardangerfjord). For the remaining time series, no statistically significant trends could be determined. A rapid reduction in the concentration of cadmium in mussels was measured in the Seine estuary after phospho-gypsum discharges were prohibited in 1992. Mean levels decreased by more than 50% in a 3 year period.

Significant downward trends were found in flounder livers from the Western Scheldt and the inner Sørfjord, and in cod livers from Fladen in Sweden (OSPAR, 2000).

Reference to EAC and BRC

Dissolved cadmium concentrations in seawater are generally well below the higher EAC limit and within the range of the agreed BRC levels *(Figure 4.5)*. In some estuaries, BRC values can be exceeded by a factor 2 – 10 and the higher EAC limit may be approached (Seine) or exceeded (Scheldt) at the location of maximum cadmium concentration.

In sediments, EAC values were exceeded at some locations near the mouth of the Rhine, in the Dutch Wadden Sea and in the Scheldt estuary.

Comparing the cadmium concentrations in mussel tissue with the BRC, the means of 46 out of 58 data sets were above the BRC. In the Tay and Forth estuaries, the BRC was exceeded by a factor of 2 – 3 and in the Seine estuary by a factor of 5. Highest ratios occurred in Norwegian fjords (Sørfjord: 95-fold, Hardangerfjord: 20-fold), due to the presence of smelting industries. In the Sørfjord there is still advice against consumption of blue mussels due to high cadmium (and lead) levels.

4.4.3 Mercury (Hg)

Sources and input estimates

Both riverine and direct inputs of mercury show a decreasing trend between 1990 and 1996. *Table 4.11* gives examples of the proportion of riverine and direct inputs, and the difference between lower and upper estimates in two different years. Inputs to the Skagerrak and Kattegat are low compared to inputs to the North Sea region. From 1990–3 aquatic inputs were dominated by the Elbe whereas from 1993–6 inputs from the Rhine-Meuse dominated (OSPAR, 1998b).

Since the mid-1980s, technical improvements in production technology have lead to a significant decrease in mercury emissions *(Table 4.9)*. This is also due to important mercury emitters in the former German Democratic Republic being closed down.

Calculated atmospheric input showed a decrease from 9 to 4 t/yr between 1987 and 1995 *(Figure 4.6)*. Dry deposition of mercury was not monitored but is estimated to account for 10% of the total deposition onto the sea (Petersen *et al*., 1995).

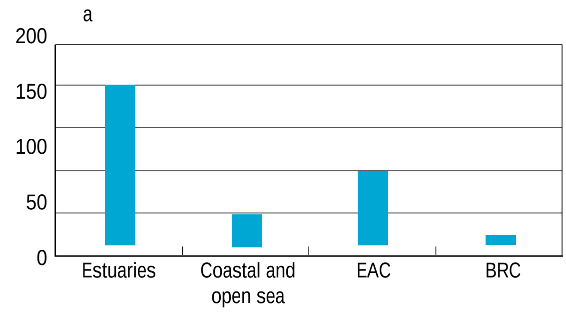
In the sea, mercury ions can be converted into volatile mercury and emitted to the atmosphere. The resulting flux of mercury is estimated to be 4 – 10 t/yr, comparable to wet and dry deposition (Baeyens and Leermakers, 1998).

Mercury concentrations are given in *Figure 4.7*.

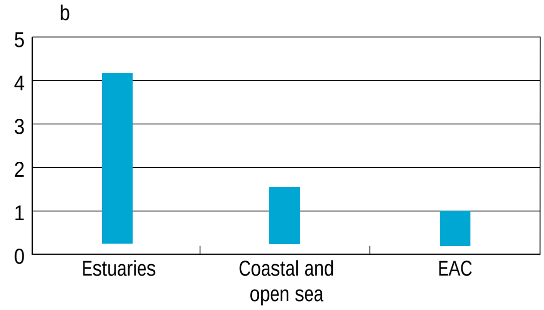
Concentrations in water

Mercury concentrations range from more than 1 ng/l at the mouth of estuaries to 0.1 ng/l in open seawater. Particulate mercury accounts for over 90% of the total mercury in coastal waters and decreases to less than 10% in the less turbid open sea.

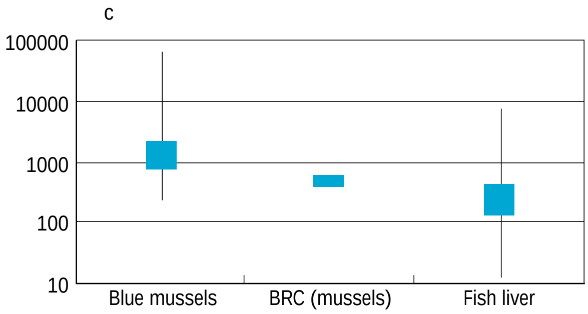
Figure 4.5 **Cadmium concentrations: (a) in water (ng/l). Concentration ranges, EAC (firm) and BRC; (b) in sediments (mg/kg). Concentration ranges and EAC (provisional); (c) in biota (ng/g dw). Concentration ranges and upper and lower quartiles. Source: OSPAR (2000).**



Based on reported values for the period 1993–6



Based on reported values for the period 1993–6



Based on 189 median yearly values for the period 1993–6. The bar indicates the data range and the box the data between the 25 and 75 percentile. The BRC range is also shown as a box.

Table 4.11 Riverine and direct inputs of mercury to the Greater North Sea in 1990 and 1996. Source of data: OSPAR (1998c).

	Riverine inputs (t)		Direct inputs (t)	
	1990	1996	1990	1996
Belgium	3.6* 4.5**	0.02* 0.03**	n.i.	0.00
Denmark	0.042	n.i.	0.091	0.001
France	5.7	0.5	n.i.	n.i.
Germany	10*§ 11*§	2.9§	0.02* 0.05**	0.01* 0.07**
Netherlands	3.1§	3.2§	0.08	0.08
Norway	0.04	0.13	0.5	0.09
Sweden	0.072	n.i.	0.033	0.023
United Kingdom	0.9* 3.4**	0.5* 1.7**	0.8* 0.9**	0.2* 0.3**
TOTAL				
Lower estimate	19*	7.25*	1.5*	0.40*
Upper estimate	49**	8.46**	1.7**	0.56**

§ Including loads from countries upstream.
* Lower estimate: for concentrations less than the detection limit, a value of ‘0’ was used for calculating load.
** Upper estimate: for concentrations less than the detection limit, the value of the detection limit was used.
n.i. no information.

In coastal areas, particulate mercury concentrations are highly variable, ranging from 0.05 – 1.0 mg/kg. In the open sea, they generally range from 0.02 – 0.20 mg/kg.

Important decreases in particulate mercury concentrations were observed in the river Elbe, from 20 – 80 mg/kg prior to 1990 to 5 – 10 mg/kg after 1992 (Wilken and Wallschläger, 1996), and were comparable to those in the Seine and the Scheldt estuaries.

A fraction of the mercury may be present in the more toxic methylated forms. Dissolved monomethyl-mercury (MMHg) concentrations range from up to 600 pg/l in estuaries to less than 10 – 60 pg/l in the open sea. Particulate MMHg concentrations range from less than 0.001 – 0.010 mg/kg (Baeyens, 1998). On the basis of the available data, no significant regional variability can be found.

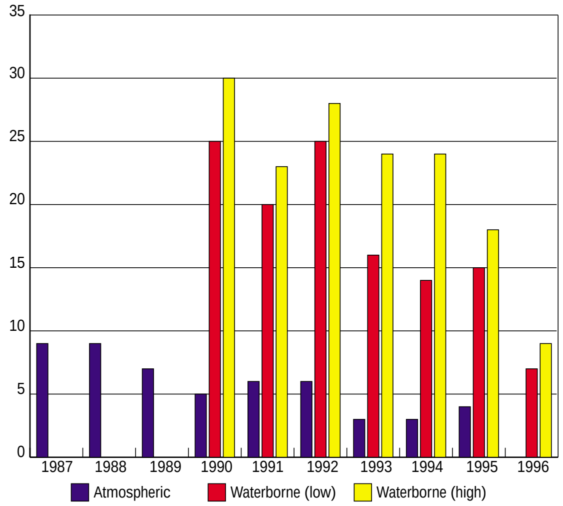
Due to major improvements in the methods applied, monitoring results of the 1980s are inconsistent with those of recent years. Hence, trends cannot be established.

Concentrations in sediment

Mercury concentrations in sediments typically range from 0.01 – 0.5 mg/kg. Higher concentrations are found in the estuaries of the Scheldt, Forth, Elbe and Thames as well as at near-shore stations and dredged spoil disposal sites.

Mercury concentrations at former sewage sludge disposal sites in the inner German Bight show a clearly decreasing trend. In the Belgian coastal area, a downward trend in mercury concentrations at the disposal sites was noted in the period 1979–95 (a 5% annual decrease), whereas for the other coastal stations and the Scheldt estuary decreases were insignificant.

Figure 4.6 **Estimates of mercury inputs to the North Sea (t/yr). Source: OSPAR (1998c).**



Atmospheric inputs are based on wet deposition measurements at coastal stations and calculated for an area of 525 000 km^2. Lower and higher estimates are reported for total waterborne inputs (riverine inputs + direct discharges).

Concentrations in biota

Eighty-six time series of mercury in blue mussels and fish covering the period 1978–96 were assessed (OSPAR, 2000). After having passed through severe statistical tests only eight significant time trends were detected, all downward except for one from mussels in the Sørfjord. Significant downward trends were found in time series for flounder from the Belgian coast, the Ems-Dollard, the Wadden Sea and the Elbe, as well as in plaice from the Southern Bight of the North Sea.

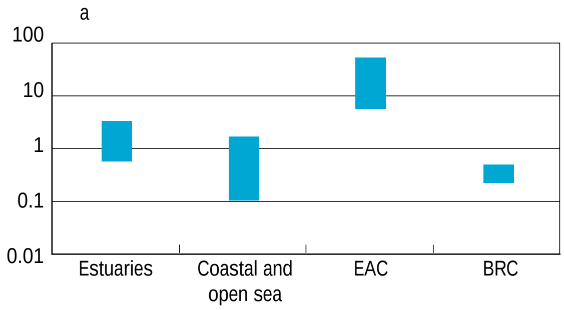
In general, blue mussel data show elevated concentrations in the vicinity of industrial and densely populated areas. In Norway, relatively higher concentrations are found in the industrial areas of the Sørfjord, in the inner parts of the Hardangerfjord and the Oslofjord. Low concentrations are found along the Swedish and the German North Frisian coasts. The Elbe estuary is severely influenced by discharges from industrial areas in the former GDR and the Czech Republic. The southern coasts of the North Sea, from the mouth of the river Elbe as far as France, show comparable concentrations, but with elevated concentrations in the Seine estuary.

Mercury in fish tissue occurs mostly in the MMHg form. However, juvenile fish, shrimp, mussels and sea urchins have lower MMHg fractions (e.g. up to 40% in mussels).

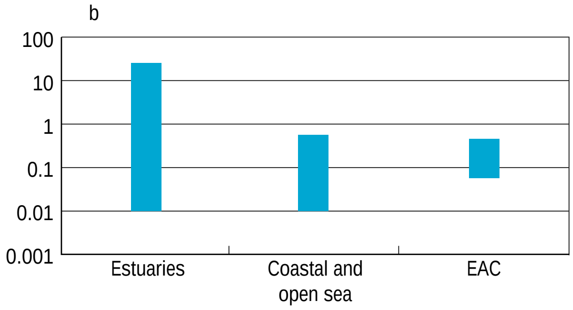
Reference to EAC and BRC

Dissolved mercury concentrations in seawater and estuaries seldom exceed lower EAC limits. In offshore stations, they are comparable to the BRC. In coastal zones and estuaries, they can exceed the BRC by a factor 2 – 10.

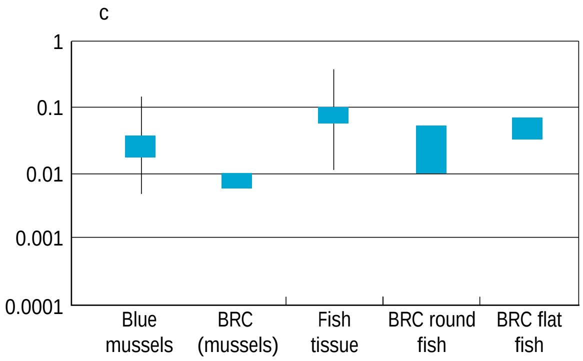
Figure 4.7 **Mercury concentrations: (a) dissolved mercury in water (ng/l). Concentration ranges, EAC (firm) and BRC values; (b) in sediments (mg/kg dw). Concentration ranges and EAC (provisional); (c) in biota (mg/kg ww). Concentration ranges and upper and lower quartiles. Source: OSPAR (2000).**



Based on reported values for the period 1993–6



Based on reported values for the period 1993–6



Based on median yearly values for the period 1993–6.
The bar indicates the data range and the box the data between the 25 and 75 percentile. The BRC range is also shown as a box.

Particulate mercury concentrations can exceed upper EAC limits in high turbidity areas, such as in estuaries.

The upper EAC limits were exceeded in a number of sediments from the estuaries of the Scheldt, Elbe and Forth and lower limits were exceeded at the disposal sites for dredged spoil. Enrichment factors (mean concentration at site divided by the BRC) range from 1 – 7 in coastal and offshore sediments and from 10 – 50 in contaminated estuarine stations.

Blue mussel data sets show systematic elevated concentrations with respect to BRCs. Highest enrichment factors in Norway occur in the industrial areas of Sørfjord (up to 11) and at one site in the Oslofjord (up to 4.5). Along the coast, from the mouth of the river Elbe to France, enrichment factors range from 2 – 7. Low ratios (1 – 3) occur all along the Swedish coast and in the North Frisian area.

In general, data sets for fish muscle tissue show values close to BRCs (enrichment factor less than 2). For Norwegian data series, enrichment factors are less than 3 except for a time series in dab from Borøy (5), and for one in cod from the inner Sørfjord (3). Enrichment factors are in many cases lower along the Swedish and Danish coasts (less than 2). German time series show enrichment factors up to 3. Dutch time series range close to the BRC. For Belgium and France, enrichment factors of less than 4 are observed.

4.4.4 Lead (Pb)

Sources and input estimates

During the period 1990–6, the direct inputs of lead to the Greater North Sea were halved. However, they only contributed about 10% of the total input. *Table 4.12* gives examples of the proportion of riverine and direct inputs, and the difference between lower and upper estimates in two different years. The total annual loads decreased slightly (from 1000 to 800 tonnes), but the mean value over the period remained high (1 200 tonnes) *(Figure 4.8)*. In the Skagerrak area, lead inputs which varied in this period between 25 and 37 t/yr showed an increasing trend. Inputs to the Wadden Sea by the Elbe have strongly decreased since 1993, but in the Weser there has been a significant increase in inputs since the beginning of the 1990s (Bakker *et al*., 1999).

Measurements of wet, dry and total atmospheric deposition at coastal stations showed a decrease of 50 – 65% between 1987 and 1995 *(Figure 4.9)* This confirms a major decrease in emissions *(Table 4.9)*, with most of the reduction having occurred before 1990 (OSPAR, 1998c). The values for the mid 1990s are in the range 700 – 900 t/yr, indicating that the atmospheric inputs are not larger than the riverine inputs, as they were before 1990. Model computations indicate that 98 – 99% of the airborne lead reaching the North Sea originates from OSPAR Contracting Parties and that the Southern Bight receives more than 50% of the atmospheric inputs (Van Pul *et al*., 1998).

Lead concentration ranges are summarised in *Figure 4.10*.

Concentrations in water

Scholten *et al*. (1998) found an overall reduction of 38% in concentrations of dissolved lead in time series for the entire North Sea between the periods 1982–5 and 1986–90. Large decreases were observed around the Skagerrak, Dogger Bank, Dutch coast and Thames estuary.

Table 4.12 Riverine and direct inputs of lead to the Greater North Sea in 1990 and 1996. Source of data: OSPAR (1998c).

	Riverine inputs (t)		Direct inputs (t)	
	1990	1996	1990	1996
Belgium	24* 31*	29* 60**	n.i.	0.009
Denmark	5.1	n.i.	2.12	n.i.
France	150	66	n.i.	n.i.
Germany	212§	125§	0.5* 1.0**	1.2* 1.8**
Netherlands	340§	380§	6.5	2.9
Norway	45* 91**	33	10	4.1
Sweden	7.8	7.5	0.5	0.5
United Kingdom	109* 131**	100* 120**	80.6* 107**	49.5* 50.5**
TOTAL‡				
Lower estimate*	890	740	100	58
Upper estimate**	970	790	130	60

§ Including loads from countries upstream.
‡ Total rounded to two significant digits.
* Lower estimate: for concentrations less than the detection limit, the value ‘0’ was used when calculating loads.
** Upper estimate: for concentrations less than the detection limit, the value of the detection limit was used when calculating loads.
n.i. no information.

Note: Some 1990 data updated by OSPAR may differ from those reported in the 1993 QSR.

Concentrations in sediments

Between 1981 and 1996, the median concentration of lead dropped by 53% in the Dutch coastal zone north of the mouth of the Rhine (Laane *et al*., 1998). Outside the Rhine plume, no significant decrease has been found.

In the Belgian coastal zone, the lead concentration was about 25% less in 1995 than the mean value observed in 1990. In the Scheldt estuary, a strong downward gradient was measured from Antwerp to Vlissingen. Between 1990 and 1995, the concentrations decreased by a factor of 1.5 – 3 (Vyncke *et al*., 1997). In

Figure 4.8 **Total waterborne lead input to the Greater North Sea (t/yr). Source of data: lower estimates of riverine and direct inputs in OSPAR (1998c).**

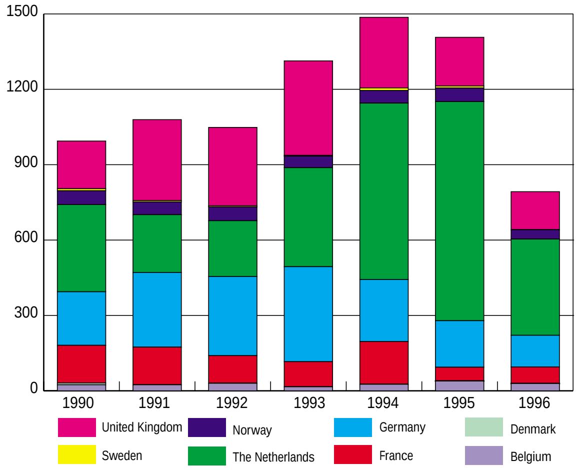
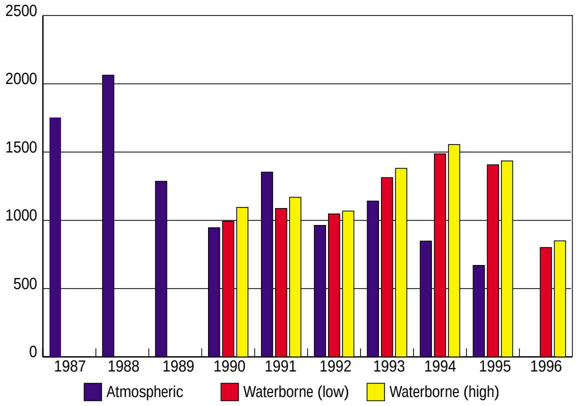


Figure 4.9 **Estimates of lead inputs to the North Sea (t/yr). Source: OSPAR (1998c).**



Atmospheric inputs are based on deposition measurements at coastal stations and calculated for an area of 525 000 km^2. Lower and higher estimates are reported for total waterborne inputs (riverine inputs + direct discharges).

the Wadden Sea, lead levels decreased by about 10% between 1988 and 1993 (Bakker *et al*., 1996).

Concentrations in biota

Most of the time series analysed showed no significant trend, except downward linear trends observed in blue mussels from Germany (Borkum), the Belgian coast, the Dogger Bank and Norway (Sørfjord). In the Belgian coastal area, a decrease in lead has been noticed in benthic organisms found in dredged spoil disposal sites.

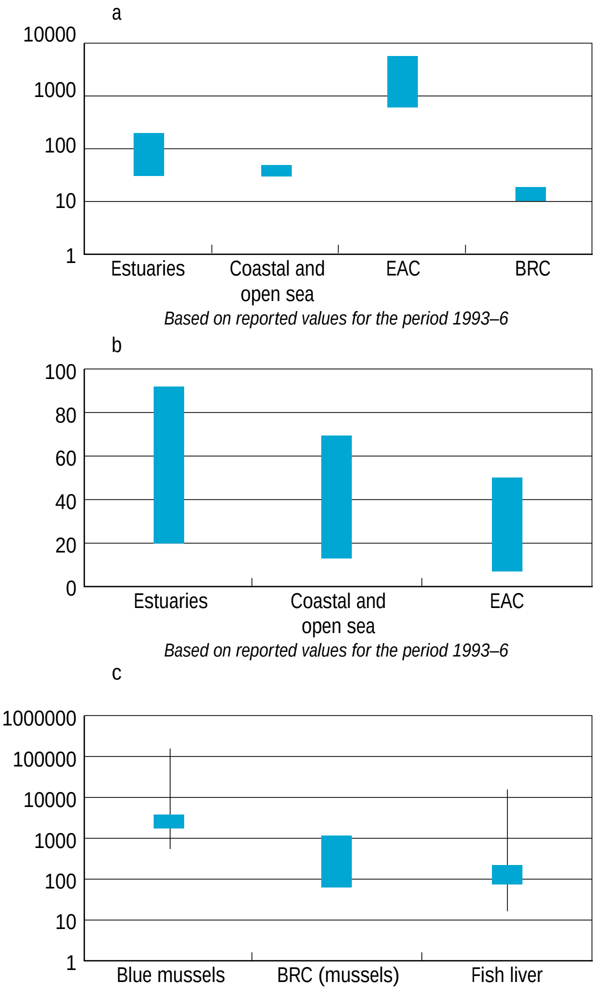
Reference to EAC and BRC

Dissolved lead concentrations in seawater and estuaries were higher than the BRC, but all below the lower EAC. The BRC was exceeded by a factor 2 in the north-east area off the English coast and in the Forth estuary. The south-east area off the English coast and the Channel were just above the BRC. Dissolved lead concentrations in the Scheldt estuary exceeded background values by a factor of 10 (upstream) – 2.5 (near the mouth).

In sediments of the Belgian coastal zone, the upper EAC limits were not exceeded, but lower limits were exceeded at four sites, including two dredged spoil disposal sites. In the Scheldt estuary, enrichment factors in sediments varied from 1.4 – 24 with a median value of 2.4 (Vyncke *et al*., 1997).

For blue mussels, 27 of 31 time series were above the BRC. The data sets for the Seine area were up to five

Figure 4.10 **Lead concentrations: (a) in water (ng/l). Concentration ranges, EAC (firm); (b) in sediments (mg/kg). Concentration ranges and EAC (provisional); (c) in biota (ng/g dw). Concentration ranges and upper and lower quartiles. Source: OSPAR (2000).**



Based on reported values for the period 1993–6 (a)

Based on reported values for the period 1993–6 (b)

Based on 158 median yearly values for the period 1993–6. The bar indicates the data range and the box the data between the 25 and 75 percentile. The BRC range is also shown as a box.

times higher than the BRC. The German coastal data sets were just above the BRC, with the exception of one location in the German Bight exceeding the BRC by a factor of 4. The Western Scheldt and Ems-Dollard were above the BRC by a factor of 3 and 4 respectively. In Norway, a number of locations were approximately ten times (Hardangerfjord) and up to forty times (Sørfjord) higher than the BRC. The Swedish Fladen and Väderöarna sites were just above the BRC.

4.4.5 Copper (Cu)

Sources and input estimates

Waterborne inputs did not show a clear trend between 1990 and 1996 *(Figure 4.11)*. *Table 4.13* gives examples of the proportion of riverine and direct inputs, and the difference between lower and upper estimates in two different years. This might be explained by the relative importance of diffuse sources.

The decrease of atmospheric emissions has been less pronounced for copper than for other heavy metals (Table 4.9.). Deposition data from coastal stations have not been assessed by OSPAR in recent years, but model calculations give an estimate of 56 t/yr reaching the Greater North Sea, 96% originating from OSPAR Contracting Parties (Van Pul *et al*., 1998). This is, however, a minor contribution when compared to the waterborne inputs.

Copper concentration ranges are summarised in *Figure 4.12*.

Concentrations in water

Very little information concerning the concentration of copper in water has been made available for the period under investigation. On the other hand, copper is a micronutrient, and its concentration is influenced by biological activity in the water column.

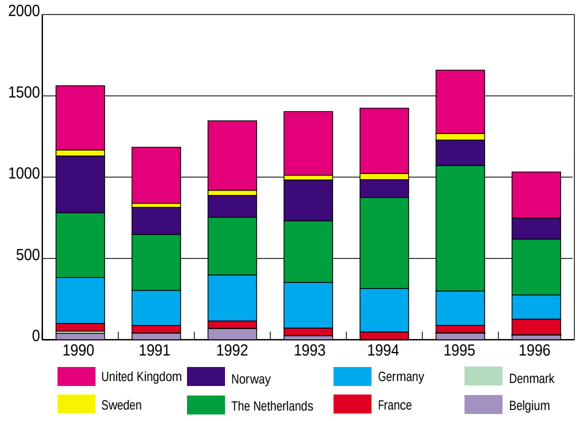
Table 4.13 Riverine and direct inputs of copper to the Greater North Sea in 1990 and 1996. Source of data: OSPAR (1998c).

	Riverine inputs (t)		Direct inputs (t)	
	1990	1996	1990	1996
Belgium	39* 51**	28* 61**	n.i.	0.002
Denmark	10.5	n.i.	3.13	1.17
France	47* 49**	97	n.i.	n.i.
Germany	282§	147§	0.9* 1.9**	2.2* 3.0**
Netherlands	390§	340§	8.3	2.6
Norway	270	94	79	35
Sweden	36.7	22.1	0.48	2.1
United Kingdom	187* 202**	171*	209* 219**	113
TOTAL‡				
Lower estimate*	1 200*	900*	300*	160*
Upper estimate**	1 300	1 000	310**	160*

§ Including loads from countries upstream.
‡ Total rounded to two significant digits.
* Lower estimate: for concentrations less than the detection limit, the value ‘0’ was used when calculating loads.
** Upper estimate: for concentrations less than the detection limit, the value of the detection limit was used when calculating loads.
n.i. no information

Note: Some 1990 data updated by OSPAR may differ from those reported in the 1993 QSR.

Figure 4.11 **Total waterborne copper input to the Greater North Sea (t/yr). Source of data: lower estimates of riverine and direct inputs in OSPAR (1998c).**



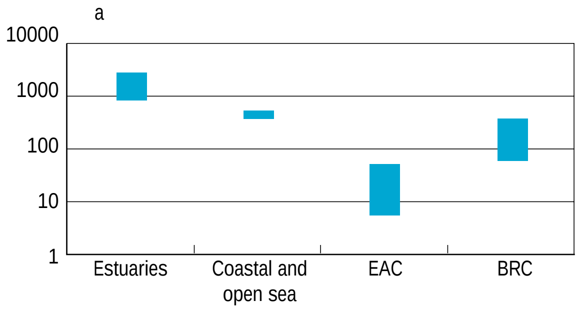
Concentrations in sediments

In the Dutch coastal zone, copper concentrations decreased between 1981 and 1996 in the areas north (40%) and south (30%) of the Rhine/Meuse mouth, and in the offshore area (35%) (Laane *et al*., 1998). Major decreases were noticed for sediments along the Belgian coast, including in the dredged spoil dumping zone, with concentrations dropping by 65% between 1990 and 1996. In the Wadden Sea, a reduction of about 20% was observed in 1993 as compared to 1988 (Bakker *et al*., 1996).

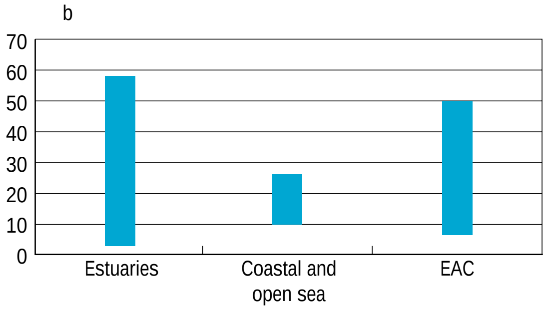
Concentrations in biota

Trend analysis conducted on 71 time series of up to 15

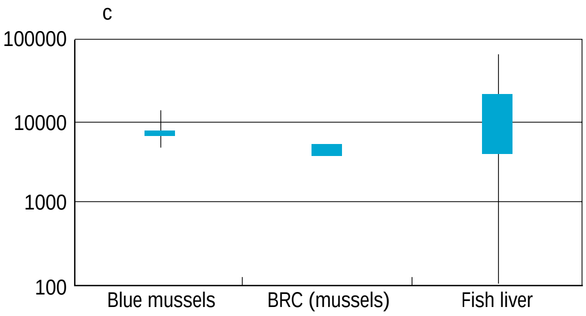
Figure 4.12 **Copper concentrations: (a) in water (ng/l). Concentration ranges, EAC (firm); (b) in sediments (mg/kg). Concentration ranges and EAC (provisional); (c) in biota (ng/g dw). Concentration ranges and upper and lower quartiles. Source: OSPAR (2000).**



Based on reported values for the period 1993–6



Based on reported values for the period 1993–6



Based on 185 median yearly values for the period 1993–6. The bar indicates the data range and the box the data between the 25 and 75 percentile. The BRC range is also shown as a box.

years duration in mussel and fish tissue (OSPAR, 2000) revealed eight linear downward trends: in Denmark (Hvide Sande), Germany (Jade Bay, outer Weser, Borkum), The Netherlands (Terschelling) and Norway (Oslofjord, Sande and Sørfjord). Sites in the Elbe estuary showed decreasing patterns, a trend that is attributed to the decline of chemical industries in the former GDR. Nevertheless, concentrations are still high, possibly due to releases from the Czech Republic. Two upward trends were detected, in France and Norway.

Reference to EAC and BRC

Dissolved copper concentrations in seawater were generally higher than the upper EAC. However, copper is an essential element. The toxicological value is slightly higher than the value needed to avoid biological deficiencies. In these cases, the EAC tends to be lower than the BRC, and is therefore less useful as a quality criterion. Discussion is ongoing internationally to solve this problem. Concentrations were in the range of the BRC along the north-east coast of England and in the Channel, and just above the BRC in the south-eastern area off the English coast. The BRC was exceeded by a factor 2 – 5 in the Tay estuary and 3 – 7 times in the Forth estuary. In the Scheldt estuary, dissolved Cu concentration exceeded background values by a factor 2 – 4.

In sediments of the Belgian coastal zone, the EAC was reached at only one dredged spoil disposal site. In the Scheldt estuary, sediments were slightly enriched.

Comparing the mean concentrations in blue mussel with the BRC, 53 of 61 blue mussel data sets were above the BRC, but because copper is known to be bio-regulated by blue mussels, interpretation is more difficult for this species.

4.5 Persistent organic contaminants

4.5.1 Organo-tin compounds

Sources and input estimates

Tributyl tin (TBT) is widely used as an anti-fouling agent in paint for ships. Vessels with a length of more than 25 m (its use has been forbidden for smaller vessels since 1990) are the major input source. Apart from harbours, dry docks and TBT-related industrial effluents are important sources. Denmark estimates their TBT input for 1997 at 0.6 – 4.9 tonnes (1/3 of the estimated use) while the Norwegian input ranged between 17.8 and 57.3 t/yr over the period 1985–95. The UK estimates annual TBT inputs to be 6 t/yr (OSPAR, 1997c).

Fluxes and transport routes

Few laboratories are able to measure the low concentrations of TBT and its degradation products (monobutyltin (MBT) and dibutyltin (DBT), often present in comparable concentrations, phenyl-tin and methyl-tin).

Concentrations in water

TBT concentrations in offshore waters are generally less than 1 ng/l (expressed as TBT), whereas values of the order of 100 ng/l are found in frequently used waterways. Marina areas are highly contaminated with TBT due to historic inputs, but in countries where effective regulations have been introduced, concentrations have substantially decreased in the past decade. In addition to statutory controls, the rate of leaching of TBT from large vessels has been reduced by the use of co-polymer rather than free-association paints.

Only a very limited amount of data is available from special surveys (OSPAR 1997i). In water, the highest concentrations have been observed in Germany (0.243 µg/l (Dornumer Siel) to 0.35 µg/l (Bremerhaven) in harbours), the Netherlands (up to 0.14 µg/l (Rotterdam)), France (in marinas) and Denmark (up to 0.12 µg/l in harbours).

Concentrations in sediment

In sediments near sources values up to 16.9 mg/kg have been observed in Denmark. Concentrations in UK samples ranged from less than 0.002 mg/kg at offshore sites to over 10 mg/kg in some harbour areas. In Norway all sediment values ranged from less than 0.01 – 1 mg/kg. In Belgium, values up to 0.081 mg/kg were found in the Scheldt. Concentrations in the range 0.0036 – 0.046 mg/kg were reported in the Dutch Western Scheldt and from 0.050 – 0.070 mg/kg in the harbour of Rotterdam.

Concentrations in biota

In biota, values from Norway are up to 3 mg/kg in blue mussels and 0.43 mg/kg in dog whelks (*Nucella lapillus*) (OSPAR, 1997c).

A number of studies point out that, even in remote areas with low concentrations of TBT in the water, accumulation occurs in the liver and kidneys of seabirds.

Reference to EAC and BRC

Since the EACs are close to or even below the detection limit of most laboratories, particular concern should be attributed to ambient concentrations, and it is impossible to judge even the lowest observed concentrations. Generally, offshore water and sediments show no detectable concentrations, but concentrations of up to 3 500, 1 200 and 300 times the EAC for water have been recorded in harbours, marinas and rivers respectively.

For sediments, the measured concentrations are up to 30 million times the upper EAC in harbours, up to 1600 times the upper EAC in rivers and less than 30 times the upper EAC in frequently used offshore waterways. In mussels, values are usually lower than the upper EAC in the coastal zone, but up to 300 times the EAC in harbours.

4.5.2 Polychlorinated biphenyls (PCBs)

The term ‘PCBs’ refers to analyses conducted on a total basis, whereas the individual congeners are referred to as ‘CB’ (chlorinated biphenyl) followed by the IUPAC number. Among the many CB congeners it is possible to measure, the sum of 7 congeners (ΣPCB_7: CBs 28, 52, 101, 118, 138, 153 and 180) is usually reported to OSPAR. CB153 is often used as a CB representative of all the others, and in biota it constitutes around 10% of the total amount.

Sources and input estimates

In coastal waters, waterborne inputs of PCBs are dominant, whereas atmospheric deposition is more important in the open sea. The atmospheric input by wet deposition to the OSPAR Convention area is estimated to be 3 – 7 t/yr for the period 1992–4 (dry deposition is not known) (OSPAR, 1997d). The main source is electrical equipment (OSPAR, 1998f). More than 90% of the total release of PCBs occurred before 1980. The estimated inputs to the Greater North Sea by direct and riverine inputs ranged from 530 – 1 500 kg/yr between 1990 and 1996 ***(Table 4.14)***.

Fluxes and transport routes of PCBs

The semi-volatile nature of PCBs is the reason for their effective transport over long distances in the atmosphere. No data are available about the fluxes to the North Sea area. Total deposition fluxes for PCBs in the Skagerrak were estimated to be 2 – 6 x 10^{-3} µg/m^2/day (OSPAR, 1997d). In water, PCBs are mainly adsorbed to particles and thus transported by materials in suspension.

Concentrations in water

Due to their low solubility, PCB concentrations in water are usually extremely low and hence difficult to detect. From a 1990 survey in Scottish waters (SOAEFD, 1996), it was concluded that values at most places were less than 1 ng/l for the sum of all the congeners. However in the Elbe plume, CB 153 concentrations were found up to 30 ng/l. At other locations in the German Bight, concentrations were below 10 ng/l.

Concentrations in sediments

Individual CB congeners are commonly detected in sediments but levels are generally low in the western North Sea (less than 2 µg/kg), with the highest levels measured in the Forth estuary (ΣPCB_7 6.8 – 11.3 µg/kg). Concentrations in the open sea were less than 1 µg/kg.

Relatively high concentrations were measured in the Ems, Elbe and Scheldt estuaries (Kallo, ΣPCB_7 382 µg/kg in 1993; OSPAR, 1998g). ΣPCB_7 concentrations greater than 20 µg/kg were found in the Dutch coastal zone. The concentrations of PCBs dropped by 70% close to the Rhine/Meuse mouth and by 80% outside of the plume in the period 1986–96.

Concentrations in biota

Concentrations of CBs in dab liver off UK coasts were found to be less than 0.1 µg/kg ww, except in the Thames estuary (ΣPCB_7: 0.19 µg/kg ww in liver) and the Forth estuary (ΣPCB_7: 0.355 µg/kg ww). These are the highest values for the western North Sea. High levels of non-ortho and mono-ortho PCBs in cod liver have resulted in advice against consumption of liver from the inner part of several Norwegian fjords (Oslofjord, Drammensfjord, Sandefjordsfjord, Kristiansandsfjord and fjords around

Table 4.14 Riverine inputs (kg) and direct discharges (kg) to the maritime area by country. Source: OSPAR (1990–95 data: 1998b; 1996 data: 1998a).

Country	Way	1990		1991		1992		1993	
		γ-HCH	∑PCB	γ-HCH	∑PCB	γ-HCH	∑PCB	γ-HCH	∑PCB
Belgium	Direct	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
	Riverine	102	31	100–122	22–28	77–88	14–40	62–98	3.5–419
Denmark	Direct	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3
	Riverine	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3	0–30.3
France	Direct	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
	Riverine	175	171–242	175	171–242	175	171–242	175	171–242
Germany	Direct	4.8	0.0–1.0	4.8	0.08–1.6	0.0–0.3	0.04–1.10	0.0–0.3	0.04–1.10
	Riverine	342	68–218	183	3.2–174	207–209	6–80	193–194	20–87
Netherlands	Direct	5.7	5.7	5.0	0.8	3.9	0.0–0.9	3.9	n.i.
	Riverine	11	150	2.4	130	6.6	96	360	130
Norway	Direct	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
	Riverine	420	74–530	216	1.5–40	70	0.04–41	61	0.1–21
Sweden							n.i.		
UK	Direct	150–152	77–142	128–154	2.5–101	103–104	2.1–10.0	110–111	0.7–62
	Riverine	147–234	31–1800	224–351	4.1–690	142–239	10–360	245–298	13–710

n.i. no information

Bergen). High levels were also observed in blue mussel samples at Borkum and Mellum (0.710 mg/kg dw). However, there is no information on time trends since most available data has not been quality assured.

Reference to EAC and BRC

For blue mussel samples collected along the coast of Norway, mean concentrations were generally about twice the BRC. The high value observed in dab near Haakonsvern exceeded the BRCs by a factor of 20. The mean concentrations of the CB153 congener in blue mussel tissue from the coasts of Germany and Belgium are on average about twelve times the BRC values. $\sum PCB_7$ levels off the German coast vary from 1.5 – 15 times the BRC, meaning that the EAC was exceeded by a factor of 2 – 3. Along the coast of Norway, the mean concentrations of CB153 were about twice as high as the BRCs for blue mussels, in all but a few sites.

4.5.3 Polycyclic aromatic hydrocarbons (PAHs)

In some cases only 6 PAHs (defined by Borneff) have been measured, whereas in other cases a more extended series of PAHs (such as the 16 of EPA, which incorporate the 9 PAH compounds agreed by OSPAR, see *Table 4.15*) have been reported as ‘Total PAHs’. As a result, it is difficult to compare data from different sources.

So far, little is known about the concentrations of the degradation products of PAHs in the North Sea area, like their sulphur-, hydroxylated- and nitro-analogues, which often have a greater persistence than their precursors (Bakker *et al*., 1996).

Sources and input estimates/trends

Annual atmospheric emissions of PAHs from mainly land based activities in North Sea riparian states, were estimated to be of the order of 7 000 t in 1990 *(Table 4.16)*. This value is an underestimate because harmonised data were not available for all of the main activities. Other emissions occur at sea, from shipping and from the offshore oil and gas industry. The respective proportions of emissions of PAHs deposited on sea and on land are unknown.

Waterborne inputs of PAHs enter the North Sea via rivers and direct discharges from land. Coal tar containing coating systems are an important source. Reliable estimates are not available, but an indication of the relative importance of waterborne PAHs is given by the concentrations found in sediments (see below). Spillage and operational discharges of oil from shipping and the offshore oil and gas sector also contribute a direct input of PAHs into the marine environment. *Table 4.20* gives the inputs of PAHs from offshore oil and gas installations in

1994		1995		1996	
γ-HCH	ΣPCB	γ-HCH	ΣPCB	γ-HCH	ΣPCB
n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
52–97	3.5–444	54–57	9.2–83	n.i.	n.i.
0–30.3	0–30.3	0–30.3	0–30.3	n.i.	n.i.
0–30.3	0–30.3	0–30.3	0–30.3	n.i.	n.i.
n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
175	171–242	175	171–242	120	95
0.0–0.3	0.01–1.00	0.4	0.05–1.10	0.04–0.05	0.02–1.00
250–272	40–150	291	58–230	234–243	55–200
3.9	0.9	3.9	0.9	n.i.	n.i.
150–310	300	352–385	470	300	200
n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
61	36–54	70	0.1–25	48	0.1–16.8
85–86	0.01–60	90–92	0.3–71	61–66	0.4–140
196–265	8.4–697	154–251	0.3–646	96–140	30.0–410

Table 4.15 Overview of PAH compounds.

PAH	Borneff	EPA	OSPAR
Indeno[1,2,3-*cd*]pyrene	●	●	●
Benzo[*b*]fluoranthene	●	●	
Benzo[*k*]fluoranthene	●	●	
Benzo[*a*]pyrene	●	●	●
Benzo[*ghi*]perylene	●	●	●
Fluoranthene	●	●	●
Phenanthrene		●	●
Anthracene		●	●
Pyrene		●	●
Naphtalene		●	
Acenaphthylene		●	
Benz[*a*]anthracene		●	●
Chrysene		●	●
Acenaphthene		●	
Dibenz[*a,h*]anthracene		●	
Fluorene		●	

the Norwegian, Dutch and Danish sectors as 28 t in 1996. Assuming that there is a correlation between the discharges of PAHs and produced water, this indicates that the waterborne contribution of PAHs from all North Sea offshore installations would be of the order 100 t/yr.

Concentrations in water

Higher concentrations were generally found in coastal and estuarine samples with total PAH concentrations up to 8.5 µg/l. Total PAH concentrations higher than 1 µg/l were found in the estuaries of the rivers Tees, Humber, Great Ouse, and Thames. On the east coast of England, of 39 samples taken in 1993 at offshore locations only one (off the river Tyne) showed an elevated concentration of naphthalene (0.263 µg/l). In the German Bight, the concentration of the sum of 7 PAHs was between 0.0018 and 0.09 µg/l.

Concentrations in sediments

The DIFFCHEM survey (OSPAR, 1997a), which described the presence of PAHs in 22 estuaries in western Europe, revealed that fluoranthene was the most prominent PAH (values ranged between 33.9 and 1 022 mg/kg dw), but benzo[*b*]fluoranthene, pyrene and benzo[*e*]pyrene were also abundant. Values for the sum of all PAHs were between 0.218 mg/kg (Wadden Sea) and 6.08 mg/kg (Scheldt estuary). In a pilot study (River Glomma, Norway) the total concentration of 21 PAHs was between 0.073 and 0.370 mg/kg, while the concentration of the carcino-

Table 4.16 Main sources and total annual emissions of PAH to air in 1990 for North Sea countries.

Country	Main sources (the two major sources are given) (*)	Total annual emissions of PAH (t/yr)	Reference
Belgium	Wood preservation, road transport	818	OSPAR (1997k)
Denmark	Wood preservation, stationary combustion/road transport	76.7	van den Hout (1993)
France	Stationary combustion/road transport	3479	OSPAR (1997k)
Germany	Stationary combustion/road transport	420	van den Hout (1993), OSPAR (1997k)
Netherlands	Solvent use (incl. wood preservation), stationary combustion	184	OSPAR (1997k)
Norway	Aluminium industry, wood preservation	140	van den Hout (1993)
Sweden	Stationary combustion/road transport	282	OSPAR (1997k)
UK	Wood preservation, stationary combustion	1437	van den Hout (1993), OSPAR (1997k)

(*) Wood preservation: assumed to include both production and use of creosote treated timber.
Stationary combustion: including both diffuse and point sources.
Road transport: not including emissions from road construction.

genic congeners was between 0.032 and 0.110 mg/kg (OSPAR, 1998h). Data from the Dutch coastal zone and from the whole Wadden Sea, showed no significant reduction in PAH levels between 1986 and 1996 (Laane *et al*., 1998). However in the Wadden Sea, an increase was found at 40% of the locations.

Offshore, the most prominent PAHs of those measured were naphthalene, phenanthrene, chrysene and benzo[*a*]pyrene. In Scottish marine sediments, total PAH concentrations were between 0.028 and 0.20 mg/kg.

Concentrations in biota

In the period 1988–95, in the Scheldt estuary, concentrations of fluoranthene and pyrene in blue mussel diminished, while benzo[*a*]anthracene concentrations did not change significantly. In the Ems-Dollard, no change in pyrene, benzo[*a*]pyrene or chrysene concentrations in blue mussel were noticed, but benzo[*a*]anthracene levels decreased.

Reference to EAC and BRC

An intermediate value of the EAC for pyrene in estuarine sediments of 0.1 mg/kg, within the range mentioned in *Table 4.7*, was exceeded at most locations, whereas for the other PAHs, the situation is more favourable, except in the estuaries of the Seine, Humber and Scheldt rivers.

Background values for water and biota are still under discussion. From the data it could be concluded that concentrations of some PAHs (e.g. fluoranthene, pyrene, benzo[*a*]anthracene, chrysene and benzo[*a*]pyrene) in the blue mussel at two sites in the Netherlands are decreasing and below the available EACs.

4.5.4 Other persistent organic compounds

Most persistent organic compounds other than PCBs and PAHs, are not systematically measured. Under CAMP, for example, only two organic substances (α-HCH and γ-HCH (hexachlorocyclohexane)) are to be measured monthly in rainwater on a mandatory basis. As a consequence, relatively few data (in time and space) are available for these compounds, and few EACs and BRCs have been established. Therefore, more information, covering the whole of the Greater North Sea and over a prolonged time is needed.

Estimated atmospheric emissions (1995) of eight organic substances and estimated reductions from 1985–95 are given in *Table 4.17*.

Dioxins and dibenzofurans

Data reflecting the degree of sediment contamination by these compounds are scarce, although one Norwegian study gives some baseline information (NIVA, 1995). Extremely high levels of PCDF/PCDDs in sediments were found in Frierfjorden in 1989, near a magnesium works (0.004 – 0.018 mg/kg TEQ), and in the Kristiansandfjord (0.002 mg/kg TEQ). Even off the south-eastern open sea coast of Norway, some 20 km from the waste outfall, the local background was exceeded 5 – 100 times. In the Dutch coastal zone, highest concentrations of PCDDs and PCDFs have been measured in the sediments of the Scheldt and Rhine estuaries (Evers *et al*., 1997). The absolute concentrations of PCDD and PCDF did not change much between 1985 and 1994 in these areas.

The presence of dioxins in the tissue of several seafood species at some Norwegian locations reflects the high values in sediments. Dioxin levels in crab hepatopancreas were 708 ng TEQ/kg ww (1993) at Frierfjorden, the same location where high dioxin levels in the sediment were found. Also in the nearby Breviksfjord, high levels were observed in the same matrix (481 ng TEQ/kg ww). Mixed soft bottom fauna also showed high levels at the same locations (Frierfjorden: 312 ng TEQ/kg ww; Breviksfjord: 64 ng TEQ/kg ww). Finally, in Frierfjorden, the

Table 4.17 Estimated atmospheric emissions of eight organic substances in 1995 and estimated percent reductions from 1985 to 1995. Source: 4NSC (1995).

Substance	Belgium		Denmark		France		Germany[11]	
	1995	%	1995	%	1995	%	1995	%
Lindane	-	>50[2]	0	100	-	-	-	-
Pentachlorophenol	0[1]	100	0	≈ 100	-	-	0	100
Hexachlorobenzene	-	-	0	100	-	-	<0.09	-
Carbontetrachloride	29.6	90	0	≈ 100	-	-	0	100
Trichloroethylene	1300	61	1200[3]	40-60	-	-	<14 000	<66
Tetrachlorethylene	2390	60	300[3]	50/70	-	-	<11 693	<83
Trichlorobenzene	3[12]	0	0	≈ 100	-	-	-	-
Trichloroethane	0[13]	100	400[3]	25/100	-	-	<26 000	100

1: No use in 1995. 2: Used tonnage. 3: Emissions in 1993/94 and with estimated reductions in 1985/1985-99. 4: No commercial use. The figure given concerns one industrial plant. There is no estimate for 1985, but the discharges of HCB are nearly eliminated and atmospheric emissions of total chloroorganic substances are greatly reduced. It is therefore assumed that the 50% target is achieved. 5: Discontinued 1 January 1996. The figure includes emissions of carbontetrachloride as a by-product associated with the production of chlorinated monomeres. 6: The figures are based on used quantities. The atmospheric emissions within the Swedish part of the catchment area can be anticipated to be about 20% of the figures.

stomach content of cod again yielded a high value of 208 ng TEQ/kg ww (1994 values). There are also restrictions on consumption of seafood in this area due to dioxins.

Toxaphene

The pesticide toxaphene has been found in German estuaries and related plumes. Very high toxaphene concentrations have been found in the blubber of dolphins from the central North Sea (19 mg/kg) (ICES, 1997).

Brominated flame retardants (BFRs)

Brominated compounds (PBBs and PBDEs) and chlorinated paraffins (CPs) are used as flame retardants and as additives in a number of products. Levels for DeBDE in sediments at various locations in the North Sea ranged from less than 1 x 10^{-4} to 1.7 mg/kg. High levels were also found in the river Scheldt (0.2 mg/kg) and off the Humber (0.04 mg/kg). Levels for other BFRs and CPs were less than 0.01 mg/kg (OSPAR, 1997a).

In the tissue of different fish species collected off the Dutch coast, concentrations of these compounds from less than 5 x 10^{-6} mg/kg dw up to 5.5 mg/kg (for BDE-47) were observed. (Boon *et al*., 1997).

Concentrations of CPs in river sediments were far lower than those of the BFRs measured in the DIFFCHEM Survey (OSPAR, 1997a), with the highest levels found in the river Scheldt (0.006 mg/kg). High concentrations of BFRs have been demonstrated in the blubber of marine mammals.

Hexachlorocyclohexanes (HCHs)

HCHs are a group of chlorinated pesticides. Application of γ-HCH (lindane) is restricted. At some locations in the Skagerrak, there was a correlation between the deposition of HCHs and the amount of precipitation. Deposition fluxes for the HCHs were up to 60 ng/m^2day.

Concentrations of HCHs in water are presented in *Figures 4.13* and *4.14*. Typically, values for α-HCH were lower than for γ-HCH. As was already reported in the 1993 QSR, a clear gradient is observed for γ-HCH, with low concentrations in the northern North Sea and higher levels in the southern North Sea.

Data on HCHs in sediments are only available for the Scheldt estuary and for the English coast. Values for α-HCH were low at the various locations (less than 0.001 mg/kg), while the concentration of γ-HCH was relatively high in the Scheldt estuary (up to 0.004 mg/kg in 1992). In the Scheldt estuary, an upstream gradient was noticed for α-HCH (period 1990–5).

Concentrations of α-HCH and γ-HCH in fish liver and mussel tissue decreased between 1990 and 1995, especially in relatively polluted regions of river estuaries, fjords and near coastal zones. In contrast, a significant upward trend was observed over the same period in dab muscle collected from Lista (Norway).

Highest values for γ-HCH exceeded EAC levels in mussels from the East Frisian Wadden Sea and at a site in the Scheldt estuary. No EACs or BRCs are available for α-HCH.

Hexachlorobenzene

Hexachlorobenzene (HCB) was found in the Forth estuary as a result of a known industrial discharge (0.009 µg/l). Comparison with former data showed that a significant reduction of HCB was observed, which corresponded to reduced effluent discharges.

Estuarine sediments are a source of HCB, which in addition to point source discharges may result in elevated concentrations in the water column. Concentrations of HCB in the Elbe estuary were around 0.003 µg/l, while in the German Bight a decrease was noticed (values less than 0.001 µg/l; Goerke and Weber, 1998). Results from

The Netherlands		Norway		Sweden[6]		Switzerland		United Kingdom[10]	
1995	%	1995	%	1995	%	1995	%	1995	%
≈ 0	>99	0	-	0	-	0	100	-	-
38	24	-	-	0	-	0	100	-	>50
0.002	98	0.13[4]	>50	0	-	0	100	-	>50
116	80	<2	>84/100[5]	1	>50[7]	-	>50	-	>50
972	52	320	58	2000	>50[8]	-	>50	-	-
1310	50	350	71	400	>50	-	>50	-	>50
0.43	45	0	-	0	-	-	80-90	-	>50
<2500	>51	200	87/100[5]	1	>50[9]	-	>50	-	>50

7: Banned for professional use 1993. 8: Banned for professional use from 1996. 9: Banned for professional use 1995. 10: Data are insufficient to determine specific reductions. However, where phase out or significant reductions in sales/use are known, anticipated reductions in inputs are indicated. 11: Data from the Federal Republic before its reunification. 12: The emissions are diffuse and too small to be considered as requiring priority attention. 13: Banned from 1 January 1996. Trichloroethane is expected to be replaced in some case by trichloroethylene.

Figure 4.13 **Geographical distribution of α-HCH concentrations in surface water (at a depth of 7m). Source: Theobald *et al*. (1996).**

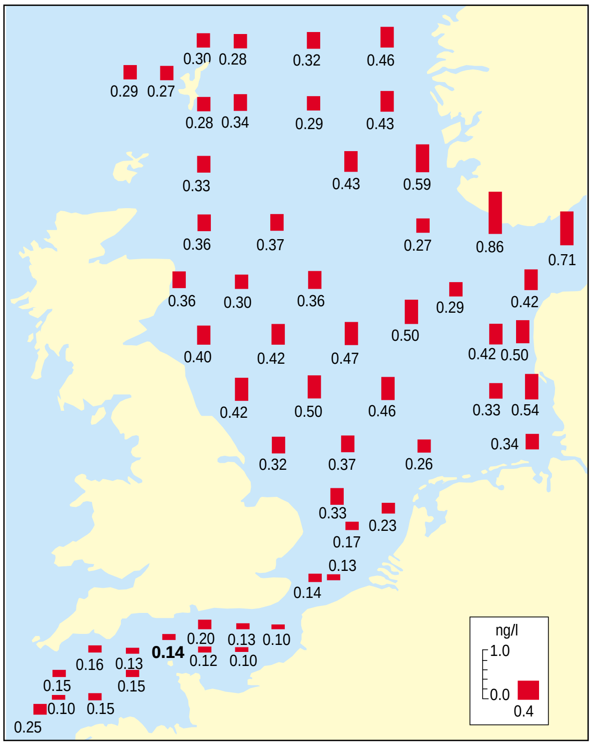
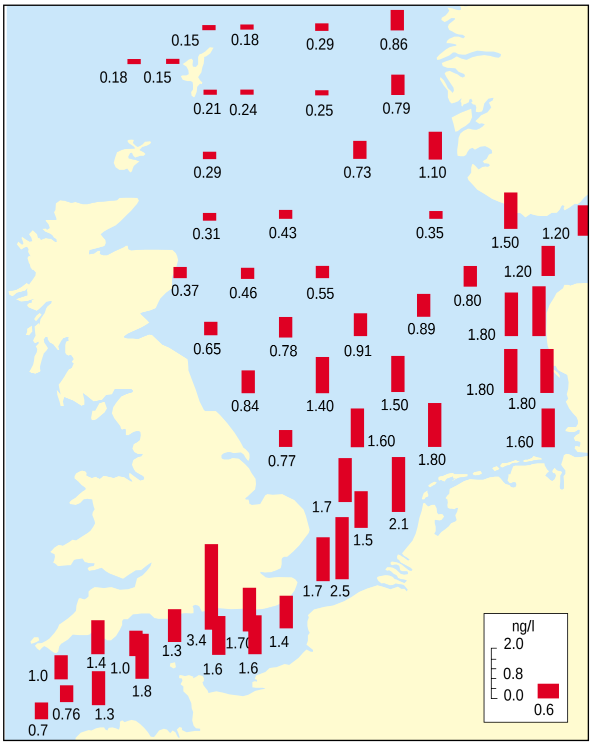


Figure 4.14 **Geographical distribution of γ-HCH concentrations in surface water (at a depth of 7m). Source: Theobald *et al*. (1996).**



other sites (e.g. in the Tay estuary) were below the detection limit.

In sediments, high concentrations were measured in the Forth estuary (3 – 130 µg/kg), with a rapid decrease in the coastal zone (2 to 4 µg/kg) and further offshore (less than 0.2 µg/kg). Concentrations in the Forth were two orders of magnitude higher than elsewhere in Scotland, reflecting well known historic inputs. Concentrations in the Scheldt estuary were comparable (0.3 – 5.3 µg/kg).

Between 1988 and 1995, concentrations of HCB, determined in fish liver and blue mussel, decreased at most locations (Weser, Elbe, Dutch coastal zone, Ems estuary, Oslofjord, Kattegat and Southern Bight). A good correlation was observed between high HCB levels in sediments and levels in biota, for example in the Forth estuary (e.g. in dab and flounder). EACs or BRCs are not available for HCB.

DDT and analogues

Concentrations in sediments greater than 1 µg/kg for total DDT (the sum of DDE, DDD and DDT) were detected off the Tyne (1.2 µg/kg) and in the estuaries of the Forth (3 – 12 µg/kg) and Scheldt (1 – 13 µg/kg).

Levels of DDT in dab liver from the British coastal zone and offshore regions were less than 0.1 mg/kg ww The highest values were observed in 1993 in the Forth estuary (0.07 mg/kg ww) and in the Humber (0.09 mg/kg ww). In these samples, DDT was only a small fraction of the 'total DDT' concentration, indicating that inputs were not of recent origin. Downward trends for DDT in cod and plaice liver were evident in time series from the Southern Bight and in herring muscle from Kattegat (Fladen). At other locations, levels were below the detection limits.

Mean values for p,p'-DDE in the period 1990–5, varied from less than 10 – 630 µg/kg dw in blue mussel and from 32 – 1000 µg/kg dw in fish.

For p,p'-DDT, no EACs have been established, while for p,p'-DDE, EACs were available for mussel and fish. At only one location (Sørfjord), the EAC was exceeded by a factor 1.5. There were no BRCs either for p,p'-DDE or for p,p'-DDT.

Dieldrin, simazine, atrazine

The herbicides simazine and atrazine were detected in the waters of the Wear (0.027 µg/l atrazine), Humber, Tees and Tyne (up to 0.006 µg/l simazine) and in the German Bight. Concentrations were, however, low at most locations and generally decreased offshore (e.g. dieldrin concentrations were all below 0.15 ng/l offshore).

In sediments from the Scheldt estuary (Vyncke *et al*., 1997) and from UK coasts and estuaries, the (banned) pesticide dieldrin was detected at all the sites that were sampled, albeit at generally low concentrations. The highest concentrations were found in the Forth and Scheldt estuaries (0.001 – 0.002 mg/kg). The difference between estuarine and offshore sites was small.

Elevated concentrations of dieldrin in dab liver were found in the Thames estuary (0.049 mg/kg ww), in the outer Moray Firth (0.072 mg/kg ww) and in the Forth estuary (0.023 mg/kg ww). All UK sites showed downward trends in all tissues. A significant downward trend was noticed for dieldrin in blue mussel and in fish tissue (Scheldt estuary, Ems-Dollard estuary).

EACs were only available for dieldrin. Concentrations of dieldrin in different biota were all far below EAC values.

Other compounds

Only few studies on other compounds were available. Such compounds are reported below. When present in marine waters these substances attained concentrations equal to or even higher than those for commonly measured contaminants such as HCHs, PCBs or HCB.

It was calculated that the influx to the Greater North Sea of biogenic halocarbons, like tetrachlorocarbon, chlorinated ethenes and tribromomethane, via the Skagerrak is 40 t/yr (Abrahamsson and Ekdahl, 1996).

Benzothiazoles (Bester *et al*., 1997) were measured in estuarine and marine waters at concentrations, ranging from 0.04 – 1.37 µg/l for methylthiobenzo-thiazole in the North Sea, while 55 µg/l were found in the river Elbe. The values for benzothiazole vary from 0.25 – 2.7 µg/l.

HHCB (trade name galaxolide®) and AHTN (trade name tonalid®), which are musk fragrances from laundry detergents and cosmetics (as replacements for nitroaromatic musks), enter the aquatic environment via sewage treatment plants. These compounds bioaccumulate in marine organisms. Musk fragrances are considered to be endocrine disrupters, although this has not been proved for HHCB and AHTN. In the period 1990–5, HHCB concentrations ranged from 0.09 – 4.8 µg/l in the German Bight and up to 95 µg/l in the Elbe estuary, while for AHTN they varied from 0.08 – 2.6 µg/l, and up to 67 µg/l in the river Elbe. Whilst AHTN concentrations remained relatively constant, HHCB showed an upward trend at some stations.

Concentrations of 2,5-dichloroaniline from upstream industrial activity, were up to 1 µg/l in the Elbe estuary. In the German Bight they ranged from less than 0.01 – 0.65 µg/l (Bester *et al*., 1998).

High levels of octylphenol and nonylphenol ethoxylates (OPE and NPE, cleaning agents with a suspected endocrine disruptive capacity) were found in sediments in the river Scheldt (20 µg/kg and 300 µg/kg respectively) and in the Elbe (5.6 µg/kg and 107 µg/kg respectively). In the UK, the OPE levels varied between less than 0.1 µg/kg and 15 µg/kg and the NPE levels between 23 and 44 µg/kg.

A remarkably high content of polychlorinated terphenyls (PCTs) was found in the livers of Wadden Sea eider ducks (*Somateria mollissima*) (Arochlor A5442 content 3.4 – 4.0 mg/kg fat). In the same area, in different marine wildlife samples, chlordane concentrations of 0.006 – 1.273 mg/kg ww were found (Boon *et al*., 1998).

4.6. **Multiple chemical inputs**

4.6.1 Mariculture

Anti-microbial and anti-parasite compounds are the two main groups of chemicals that may be found in measurable concentrations in sediments around fish farms. The list of anti-microbial compounds (oxytetracycline, oxolinic acid, sulphonamides, penicillin, and other therapeutants) is undergoing continuous change as new compounds are brought forward for licensing. The organo-phosphorous compounds dichlorvos and azamethiphos are authorised for use against parasites, and, recently, hydrogen peroxide treatment has been introduced.

Most antibiotics are administered as feed additives and enter the sea via waste feed and faeces. Despite the uncertainties in some estimates, there is a consistent pattern of reduction in the use of anti-microbial agents in Norway, Ireland and Scotland, even though the production of fish has increased considerably. The total amount of antibacterial agents reported for Norwegian mariculture in 1995 was 3.2 t (26.8 t in 1991) and the same trend is noticed for dichlorvos (395 kg used in Norway in 1995, down from 3 400 kg in 1990). More than 1 500 tonnes of hydrogen peroxide was used in 1995.

Most anti-microbial compounds readily associate with particles. Studies have shown that the concentrations of oxytetracycline in sediments are between 0.1 and 10 mg/kg near the farm cages. The residues are confined to a very small area (about twice the area of the cages) and to the top 10 cm layer of the sediments (Kerry *et al*., 1995, 1996). Reported half-lives of these pharmaceuticals vary from a few days to six months (Samuelsen *et al*., 1992). By contrast, dichlorvos is fairly soluble in seawater and is not found in marine sediments. Hydrogen peroxide degrades rather rapidly in the sea.

4.6.2 Offshore chemicals (other than oil)

Sources of contaminants arising from the offshore oil and gas industry are drilling muds and cuttings, produced water and spills. Other substances apart from oil are involved (*Tables 4.18* and *4.19*).

An estimate of the amounts of chemical substances discharged in the Norwegian sector via produced water from 23 oil producing platforms shows that the level of aromatics (mostly phenols and benzoic acids) is higher than the aliphatics (dispersed oil). Phenol and its lower alkylated homologues (C1 – C3) constitute the bulk of the phenols, while the C4 – C8 phenols, suspected of having endocrine impacts, constitute only about 5% of their total amount. Naphthalenes dominate the PAHs, while the heavier PAHs, potential carcinogenic and mutagenic compounds, are present in very low concentrations (OSPAR, 1998i).

Barium and iron are the most dominant metals, but smaller amounts of other metals are also found, especially nickel, lead and zinc. *Table 4.20* gives a partial view of the amounts of various priority contaminants in produced water discharged in the North Sea. As the amount of produced water has increased substantially (see section 4.7), it might be expected that the amount of chemicals associated with this particular source would follow the same trend. The sector concerned has considered that data on the exact composition of some chemicals are confidential to the relevant national authorities. Contracting Parties have data on the composition and quantities discharged and OSPAR is in the process of harmonising the format under which such data is collected and reported on a regional basis. Under these circumstances, it is not yet possible to present a complete overview of the total amount of offshore chemicals on a region-wide basis.

4.6.3 Industrial and municipal wastes

The dumping of chemical wastes into the North Sea ceased in 1993 (see section 3.9). However, direct discharge of wastes from the production of titanium dioxide still continued in 1994, mostly in French and UK estuarine waters (Seine, Humber and Tees). *Table 4.21* shows the decreasing trend in the amount of waste and related substances discharged during the period 1984–94 by France, the United Kingdom, Germany, The Netherlands and Norway. Sewage sludge dumping was completely phased out by the end of 1998 (see section 3.9.2).

Table 4.18 Discharges of priority substances (kg) via chemicals used in offshore oil and gas production in 1996 – available data are for the Danish sector only.

Substance	Quantity discharged
Cadmium (and compounds)	22
Mercury (and compounds)	25
Zinc	634
Lead	1 604
Chromium	124
Nickel	47
Copper	108

Table 4.19 Discharges (tonnes) of production, utility, and drilling chemicals from oil and gas production facilities in 1996 – available data are for the Danish sector only.

Substance / use	Total quantity discharged
Antifoams	0
Biocides	29
Carrier solvents	8231
Corrosion inhibitors	18
Defoamers	4
Demulsifiers	24
Dispersants	7
Drilling lubricants	89
Emulsifiers	7
Flocculants (water injection)	8
Fluid loss control agents	174
Gas treatment chemicals	0
Gels	1
Inorganic chemicals	2569
Lost circulation materials	35
Oxygen scavengers	1
Pipe release agents	5
Polymeric viscosifiers and filtrate reducers	227
Scale inhibitors	6
Scale inhibitors/encapsulators	123
Surfactants/detergents	0
Thinners	0
Viscosifiers	205
Weighting agents and inorganic gelling products	21 909
Others	2548

Table 4.20 Discharges of priority substances via produced water from offshore installations in 1996. Source of data: OSPAR (1998i,j).

Substance	Quantity discharged (t)	Average concentration* (mg/l)
Cadmium	0.6	0.01
Mercury	0.3	0.0003
Lead	6.9	0.09
Nickel	18.6	0.3
Total aromatics*	2130	28
Phenol/benzoic acids*	1345	14
Benzene	454	n.i.
PAHs (95% naphtalene)	28	0.36

Data show estimated discharges for Denmark, the Netherlands and Norway.

Information from the United Kingdom is not available.

* Data for Norway only.

n.i. no information.

4.6.4 Dumping of dredged materials

On account of the huge quantities of dredged material involved, the amount of associated heavy metals deposited in the marine environment is considerable. However, much of the trace metal content is of geological origin and many operations simply relocate the material rather than constituting a new addition to the environment.

Information provided by The Netherlands for dredging activities in the Dutch sector (OSPAR, 1997f) indicates that the anthropogenic contribution is very low for chromium, copper and nickel (0 – 2%), medium for mercury, arsenic, lead and zinc (30 – 50%) and predominant for cadmium (70%).

Despite the higher dredging activity in 1994 compared to 1990, the contaminant loads have shown a decreasing trend for materials originating from estuaries and navigation channels *(Tables 4.22–4)*. Most probably, this evolution reflects the reduction of heavy metal inputs to rivers and estuaries and the resulting improvement in the quality of dredged material. For the dredging of harbours no changes were observed.

Table 4.21 Discharges of waste and associated contaminants from the production of titanium dioxide. Source of data: OSPAR (1998e).

	1984 (10^6 t)	1990 (10^6 t)	1994 (10^6 t)
Total waste discharged	1.076	1.094	0.468
Iron(II) sulfate	0.345	0.256	0.107
Sulfuric acid	0.598	0.510	0.260
	(t)	(t)	(t)
Chromium	511	491	217
Lead	23	18	4.6
Nickel	31	31	8
Copper	21	29	8
	(kg)	(kg)	(kg)
Cadmium	297	130	54
Mercury	128	72	31

4.7 Oil

4.7.1 Sources and inputs

The input of petroleum hydrocarbons in the North Sea is the result of sea-based activities (mainly shipping and offshore activities), coastal discharges of sewage and industrial effluents, dumping of oil-contaminated dredged materials, riverine inputs, atmospheric deposition and natural seeps. Discharges from sea-based activities and refinery effluents have been closely monitored over the past years, but there has been less focus on the inputs due to other sources. Methodological problems (sampling and analysis) are still to be solved for the evaluation of riverine oil input. Oil slicks are frequently observed originating from both ships and offshore installations, and are the result of both legal and illegal discharges or accidents. The majority of these slicks consist of ships' bilge oil, but crude oil and lubricating oils also occur along with non-mineral oils.

Offshore oil and gas industry

Figure 4.15 shows the quantities of oil discharged from 1984-97 (OSPAR, 1997g; 1999). A total of 11 800 t was discharged in 1995, of which 65% originated from produced water, 33% from cuttings, 2% from accidental spills and less than 0.01% from flaring operations. Between 1984 and 1995, the total amount of oil discharged has been reduced by about 50%, despite the fact that the number of offshore installations (oil and gas) increased from 143 to 458 units over the same period.

The amount of oil discharged via cuttings has been drastically reduced by 83% between 1984 and 1995. This is mainly due to the fact that diesel oil-based drilling muds were phased out in 1986 and oil-based muds (OBM) were partially replaced by water-based muds or muds containing organic-phase drilling fluids (OBF) after 1987. No OBM has been used in the Danish sector since 1991. In the UK

Table 4.22 Dredged material and associated contaminant loads (tonnes) dumped in the Greater North Sea in 1994 compared to 1990. Source: 1990 data from North Sea Task Force (1993), 1994 data from OSPAR (1997f).

	Amount dumped	Contaminant load										
		Cd	Hg	As	Cr	Cu	Ni	Pb	Zn	PCBs	PAHs	Oil
Harbour areas*	99 763 914	29.0	11.7	293	1918	997	785	1946	6398	0.3	15	1988
Estuaries and Navigation channels*	87 813 089	31.3	5.4	216	699	300	378	750	1991	0.1	n.i.	n.i.
TOTAL (1994)*†	187 577 003	60	17	510	2600	1300	1200	2700	8400	0.4	15	2000
TOTAL (1990)†*	*136 037 942*	*71*	*19*	*720*	*2800*	*1300*	*1200*	*2700*	*7900*	*0.6*	*n.i.*	*n.i.*

* Including dumping in internal waters; † Total contaminant loads rounded to two significant digits; n.i. no information.

Table 4.23 Dredged material and associated contaminant loads (tonnes) from harbour areas, dumped in the Greater North Sea in 1994 compared to 1990. Source: 1990 data from North Sea Task Force (1993), 1994 data from OSPAR (1997f).

	Amount dumped	Contaminant load										
		Cd	Hg	As	Cr	Cu	Ni	Pb	Zn	PCBs	PAHs	Oil
Belgium	5 839 198	10.0	1.5	51	175	115	83	268	601	0.06	n.i.	n.i.
Internal waters	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Denmark	3 275 590	0.09	0.04	0.08	10	3.2	2.0	7.6	26	n.i.	n.i.	n.i.
Internal waters	774 030	0.04	0.02	n.i.	5	1.5	0.02	3.8	12	n.i.	n.i.	n.i.
France	7 279 935	2.4	0.84	36	172	74	40	152	1 480	0.13	0.33	n.i.
Internal waters	1 143 805	0.23	0.04	5.2	17	5.5	6.2	24	39	0.01	n.i.	n.i.
Germany	26 169 000	0.73	1.4	17	70	25	27	67	163	0.04	n.i.	n.i.
Internal waters	26 136 000	0.73	1.4	16	69	25	26	66	161	0.04	n.i.	n.i.
Netherlands	33 610 638	10	4.3	171	509	260	199	556	1 716	0.05	15	1 988
Internal waters	9 055 828	2.7	1.1	50	145	72	54	149	488	0.03	4.4	463
Norway	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	207 327	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
Sweden	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	27 350	0.003	0.002	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.a.	n.a.	n.a.
United Kingdom	23 354 876	5.6	3.6	19	982	520	435	896	2 411	n.i.	n.i.	n.i.
Internal waters	13 660 117	2.2	2.3	3.4	704	300	254	573	1 705	n.i.	n.i.	n.i.
TOTAL (1994)*	48 524 780	22.9	6.8	219	978	593	446	1 131	3 992	0.2	11	1 525
TOTAL (1994)**	99 529 237	29.0	11.7	294	1 918	997	786	1 947	6 397	0.3	15	1 988
*TOTAL (1990)***	*64 546 834*	*29.1*	*10.0*	*440*	*1 770*	*927*	*621*	*1 770*	*5 000*	*0.4*	*n..i.*	*n.i.*

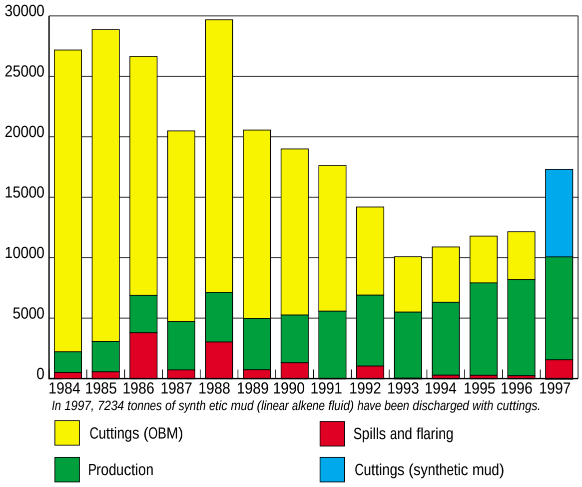
* Excluding dumping in internal waters; ** Including dumping in internal waters; n.i. no or insufficient information; n.a. not applicable.

sector, no OBM cuttings have been discharged since 1996. OBM cuttings from Dutch and Norwegian installations are brought onshore for treatment and disposal. Additionally, OBM has been reinjected in the Norwegian sector (about three-quarters of the generated amount for 1997).

Due to the maturation of the producing fields and the extension of offshore activity, the amount of produced water has increased substantially during the past decade. As a consequence, the quantities of oil discharged via produced water increased from 1 717 t in 1984 to 7 648 t in 1995, of which 2 429 t (32%) originated from the 46 installations not meeting the 40 mg/l oil in water target standard (PARCOM Recommendation, 86/1). Installations in the UK and Norwegian sectors account for 77% and 18%, respectively, of the oil input from this source.

Accidental spills result in relatively minor amounts of oil entering the North Sea. Even if the total number of reported spills seems to follow an increasing trend (198 cases in 1986, 414 in 1992, 621 in 1995), the total

Figure 4.15 **Contribution of different sources of inputs of oil (in tonnes) from offshore installations. Source: OSPAR (1999).**



In 1997, 7234 tonnes of synth etic mud (linear alkene fluid) have been discharged with cuttings.

Table 4.24 Dredged material and associated contaminant loads (tonnes) from estuaries and navigation channels, dumped in the Greater North Sea in 1994 compared to 1990. Source: 1990 data from North Sea Task Force (1993), 1994 data from OSPAR (1997f).

	Amount dumped	Contaminant load										
		Cd	Hg	As	Cr	Cu	Ni	Pb	Zn	PCBs	PAHs	Oil
Belgium	41 764 745	30	4.5	210	550	174	297	545	1 566	0.1	n.i.	n.i.
Internal waters	17 294 584	1.3	0.1	47	99	16	19	55	273	0.003	n.i.	n.i.
Denmark	404 825	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
Internal waters	124 900	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
France	8 298 550	0.001	0.001	n.i.	n.i.	0.2	0.3	0.3	0.9	n.i.	n.i.	n.i.
Internal waters	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Germany	32 962 000	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	32 962 000	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.	n.i.
Netherlands	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Norway	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Sweden	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Internal waters	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
United Kingdom	4 382 969	1.6	0.8	5.7	149	126	81	204	424	n.i.	n.i.	1.2
Internal waters	241 448	0.04	0.06	0.01	4.7	3.0	1.6	5.1	20	n.i.	n.i.	n.i.
TOTAL (1994) *	37 190 157	30.0	5.2	169	595	281	357	690	1 698	0.1	n.i.	n.i.
TOTAL (1994)**	87 813 089	31.3	5.4	216	699	300	378	750	1 991	0.1	n.i.	1.2
*TOTAL (1990)***	*71 491 108*	*41.4*	*8.7*	*279*	*1 040*	*398*	*584*	*971*	*2 900*	*0.2*	*n.i.*	*n.i.*

* Excluding dumping in internal waters; ** Including dumping in internal waters; n.i. no or insufficient information; n.a. not applicable.

quantity of oil discharged in this way has decreased markedly since 1986 (from 3 800 t in 1986 down to 270 t in 1995). In 1995 about 85% of the spills involved less than 1 t of oil.

Produced water overtook cuttings as the main source of oil input from offshore installations in 1993. Since then, the total oil input has been increasing again after the major reduction observed during the period 1984–93.

Shipping

The analysis of oil samples taken from the Danish, German and Dutch coastline between 1990 and 1992 indicates that bunker and lubrication oil residues are the main source of oil pollution in that zone. In the Skagerrak area, crude oil from illegal tank cleaning is also involved. For 1995, the amount of oil discharged into the North Sea area from shipping is estimated at 6 750 t.

Within the scope of the ‘Bonn Agreement’ (1983), regular pollution control flights have been conducted since 1986. Despite a steady increase in the number of airborne missions, the total number of observed slicks has decreased over the period 1989–96, although there was an increase of slicks reported in 1996 and, to a lesser degree, in 1997 for the area under Dutch surveillance. The average volume of identified oil slicks is reducing: out of a total number of 650 slicks observed in 1995 (against 1 104 in 1989), 519 were estimated to be less than 1 m^3, 107 between 1 and 100 m^3 and 2 greater than 100 m^3. An overview of the detected oil slicks (1998) is presented in *Figure 4.16*.

However, airborne missions can only detect a limited number of illegal discharges. For the marine area under Belgian surveillance, an estimate was based on the aerial coverage time, on the mean lifetime of oil slicks and on the influence of weather and sea conditions upon dispersion. It suggests that the reported oil quantities represent only 10% of the actual discharges (Schallier *et al*., 1996).

Refineries

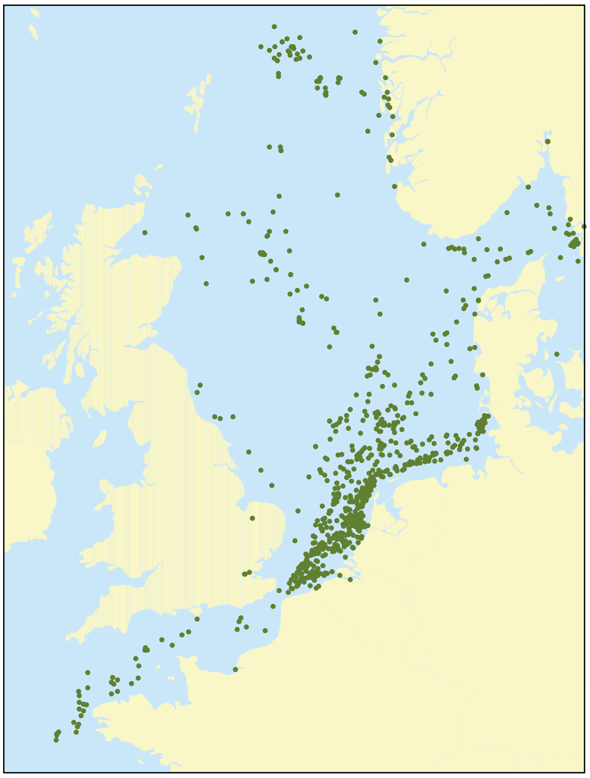
In 1997, 58 refineries were operated by nine OSPAR Contracting Parties, (OSPAR, 1999), discharging their effluents into coastal water (4), estuaries (29) and inland waters (25). About 83% of the total amount of oil was discharged into estuaries, mainly in the UK (58%) and in the Netherlands (15%). The total amount of oil discharged was 670 t in 1997, compared with 1 441 t in 1993 and 9 047 t in 1981. As a result of major treatment system improvements, there has been a strong reduction in the amount of oil discharged per tonne of oil processed (93% reduction between 1981 and 1997).

Other sources

Estimates of oil inputs from other sources have not been subject to regular reporting within OSPAR. However, for the Dutch sector of the North Sea it has been estimated that in 1995 atmospheric deposition and the dumping of harbour dredgings contributed 430 t and 2 000 t, respectively (Evers *et al*., 1997).

The total riverine input of oil was reported as 16 000 – 46 000 t/yr in the 1993 QSR. In the Dutch sector, it has declined from about 5 000 – 6 000 t/yr in 1980 to 580 – 1 235 t/yr in the period 1990–4. In 1995, inputs from the Rhine, Meuse and Scheldt together have been estimated at 1 000 t, which is only 15% of the total inland discharges and trans-boundary influx in the Netherlands for that year (6 640 t). Oil inputs from the Elbe amounted to 750 t/yr between 1989 and 1992. The 1993 QSR indicates that natural seepage of oil is of the order of 1 000 t/yr.

Figure 4.16 **Observed oil slicks in 1998. Source: Bonn Agreement (1999).**



● position of an oil slick observed during 1998

4.7.2 Levels and visible impacts

Water

Values of 1 – 5 µg/l have been reported in water in the vicinity of oil platforms and in areas with intensive shipping. In the German Bight, the distribution of total hydrocarbon is mainly influenced by the Elbe plume, with concentrations ranging from 15 µg/l at the mouth of the estuary to 2 µg/l along the Frisian coast.

Oil slicks

Slicks continue to be predominantly observed along the shipping corridor between the Straits of Dover and the German Bight *(Figure 4.16)*. The rate of oiled seabirds (considered to be an indicator of oil pollution) showed a decreasing trend in nearly all the coastal regions of the North Sea over the period 1984–94 (see Chapter 5).

Sediment

Between 1 and 1.5 x 10^6 t of drill cuttings are currently estimated to lie on the seafloor of the North Sea (USEIA, 1998). Considerable amounts of oil adhering to drill cuttings have contaminated the seabed locally. In a study of 12 well sites on the Dutch continental shelf where OBM cuttings have been discharged during the 1980s and early 1990s, the chemical analysis revealed elevated oil concentrations at 5 of the locations, and traces of oil were visually observed at another 2 locations (Daan and Mulder, 1996). Maximum concentrations, measured over the upper 10 cm layer, were generally in the order of 10 – 50 mg/kg, but at one location a concentration of 200 mg/kg was found. Elevated oil concentrations only occurred within the first 100 – 250 m from the platforms, and it is suggested that the zone where persistent biological effects may be expected on the benthic community generally extends to a radius of about 200 m from OBM discharge sites. In the Norwegian sector, a survey of 14 fields showed that hydrocarbon contaminated sediments could in some cases extend to 2 – 8 km from the fields (Bakke *et al*., 1996).

4.7.3 Reference to EAC and BRC

No EACs or BRCs have currently been agreed by OSPAR.

4.8 **Radionuclides**

4.8.1 Sources and inputs

Data on all relevant sources of radionuclides are reported annually to OSPAR.

Artificial radionuclides

The main sources of artificial radionuclide inputs to the North Sea are discharges from nuclear fuel reprocessing plants (Cap de La Hague and, indirectly, Sellafield), the Baltic Sea outflow and inflow of Atlantic Water. The Baltic Sea outflow continuously supplies more ^{137}Cs (from the Chernobyl accident) than the reprocessing plants (Bailly du Bois *et al*., 1993). In the period covered by this report, the quantities of radionuclides released in liquid effluents from each installation were far below the discharge limits (OSPAR, 1998k).

The discharges of most radionuclides from Sellafield, located in the Irish Sea, and La Hague have reduced significantly in recent years but there have been important exceptions. Discharges of ^{129}I from Cap de La Hague have increased from 0.65 TBq in 1993 to 1.69 TBq in 1996 following the commissioning of a new reprocessing operation (Raisbeck *et al*., 1997). At Sellafield, releases of the actinides and ruthenium have decreased, but there have been consequential increases of the less radiologically significant ^{99}Tc from 6 TBq in 1993 to 150 TBq in 1996, and of ^{129}I from 0.16 TBq in 1993 to 0.41 TBq in 1996 (OSPAR, 1998k,1998l). In addition, remobilization of the actinides and ^{137}Cs from Irish Sea seabed sediments has resulted in the continuing export of these radionuclides via the North Channel to the North Sea (Kershaw *et al*., 1999). For example, it has been estimated that 350 – 570 TBq of ^{137}Cs was remobilised from the seabed in the period 1988–95 (Poole *et al*., 1997).

The total input of alpha-emitting radionuclides into the North Sea has decreased from 3 to 0.32 TBq/yr between 1993 and 1996, mainly caused by reduction of discharges from nuclear reprocessing plants.

The inputs of beta-emitting radionuclides excluding tritium and ^{99}Tc, have decreased from 250 TBq in 1993 to 182 TBq in 1996. Tritium (^{3}H) is the only radionuclide that is still discharged in significant amounts. The releases of tritium, in terms of activity, have increased from 11 000 to 16 800 TBq/yr between 1993 and 1996, mainly due to the discharges from the nuclear reprocessing plants (OSPAR, 1998k, 1998l). Tritium is of minor radiological significance as it is a low-energy beta emitter.

The annual discharges of other radionuclides were much lower than the maxima allowed for each of the nuclear installations (OSPAR, 1998k, 1998l).

Natural radionuclides

Inputs of natural radionuclides originate mainly from anthropogenic sources like mining and ore processing, burning coal, oil or natural gas in thermal power plants and the production of phosphate fertilisers.

The most important non-nuclear industrial process, giving rise to significant radioactive discharges is phosphate fertiliser production. About 1.6×10^{6} t of phospho-gypsum waste were produced in 1993 in the OSPAR countries, of which about 40% was discharged into the marine environment (OSPAR, 1997h). This corresponds to annual discharges of 1.3 TBq ^{226}Ra, 1.12 TBq ^{210}Po and 0.1 TBq ^{238}U. The majority of the uranium contained in the phosphate ore is combined with the phosphoric acid produced, while radium and polonium are contained in the phospho-gypsum. Over the last few years, discharges of phospho-gypsum have tended to decrease. This development is expected to continue mainly due to the reduced production of phosphoric acid by European countries and to the land-based storage of the waste products. The discharge of phospho-gypsum into the sea has already been abandoned in several countries including France, the main producer of phosphate fertilisers in the EU.

4.8.2 Fluxes and transport routes of radioactive substances

The discharges from the Cap de La Hague reprocessing plant are transported by inflowing Atlantic water through the Channel along the continental coast, taking between 12 and 18 months to reach the coastal waters along the eastern North Sea (Salomon *et al*., 1995). The Sellafield discharges are transported from the Irish Sea around Scotland, after which a fraction of the radionuclides cross the North Sea. A transit time of 3 – 4 years is cited for this transport (Dahlgaard, 1995). However, recent observations and modelling of the discharged ^{99}Tc from Sellafield indicate more rapid transport through the North Sea. This is partly caused by the very strong natural flushing of the North Sea in the first half of the 1990s (see ***Figure 2.15a***). Most of the radionuclide input is transported out to the north in the Norwegian Coastal Current.

4.8.3 Levels of radioactive substances: trends and spatial distribution

Water

The levels of ^{99}Tc observed in seawater samples collected in the North Sea were in the range 0.9 – 8.5 Bq/m^{3} in 1996 and in the range 1.7 – 3.4 Bq/m^{3} in 1997 (OSPAR, 1998m) A general increase over large areas of the North Sea was observed compared to the early 1990s, when the values were generally less than 1 Bq/m^{3}. The ^{210}Pb and ^{210}Po concentrations in water samples from Dutch coastal waters varied between 0.4 – 2.9 Bq/m^{3} (the background concentration of ^{210}Pb and ^{210}Po ranges from 0.4 – 1.9 Bq/m^3). These levels can be influenced by discharges from the phosphate-ore processing industries, for instance the highest concentrations, up to 3.2 Bq/m^3 were found at locations in the Scheldt estuary close to past discharges of the phospho-gypsum industry. Measurements undertaken along the French Channel Coast showed a similar range of ^{210}Po concentrations of between 0.2 – 2.7 Bq/m^3 in filtered water. Activities in the North Sea, measured 30 km from the coast, were around 0.7 Bq/m^3 (^{210}Po and ^{210}Pb) and 5.3 Bq/m^3 for ^{226}Ra (OSPAR, 1997h). The natural concentrations of ^{226}Ra range from 0.8 – 8 Bq/m^3.

Sediments

Concentrations of artificial radionuclides in the North Sea sediments were generally low, except near the outlets. The specific activity of ^{134}Cs and ^{137}Cs, a major fraction of the Chernobyl fallout in 1986, decreased between 1990 and 1996. Along the North Sea coast of the UK, values after 1990 (0.3 – 0.6 Bq/kg for ^{134}Cs and 2 – 24 Bq/kg for ^{137}Cs) were much lower than the high specific activities determined in 1987 (values of 6.0 Bq/kg for ^{134}Cs and 65 Bq/kg for ^{137}Cs), confirming a decline in the effect of the Chernobyl accident (Camplin, 1992).

The background activity of natural radionuclides predominantly present in sediments is ^{40}K (2 – 1 000 Bq/kg), ^{232}Th (12 – 50 Bq/kg) and ^{238}U (2.5 – 186 Bq/kg) with its decay product ^{210}Po (100 – 280 Bq/kg) (Laane, 1992).

Measurements of ^{210}Po and ^{210}Pb concentrations in sediments along the French Channel coast in 1992 and 1993 showed values from 10 – 120 Bq/kg. For the same period, the measured activities of sediments along the Dutch coast were 20 Bq/kg (^{226}Ra, ^{210}Pb, ^{210}Po) and 1 Bq/kg (^{231}Pa) (OSPAR, 1997h). Recent measurements of ^{226}Ra in Rotterdam harbour revealed values of about 150 Bq/kg.

Biota

Typical levels of total alpha activity, mainly due to the natural radionuclide ^{210}Po are between 2 – 40 Bq/kg ww in fish and shellfish (Camplin, 1992). The measured ^{210}Po values in marine products show seasonal variations up to a factor of two, with the highest concentrations being observed in summer. Considerable variations are observed between species. The mean concentration in winkles near discharges is 260 Bq/kg, while it is 4 times lower in lobsters. Mussels affected by phospho-gypsum discharges have ^{210}Po concentrations in the order of 20 – 50 Bq/kg, 2 to 3 times higher than in unaffected regions. These values decrease with distance from the point of discharge, by a factor of 10 over 30 km (OSPAR, 1997h).

Concentrations of total beta-activities, mostly due to the natural radionuclide ^{40}K in fish and shellfish were 20 – 130 Bq/kg ww In mussels, total beta-activities were 20 – 60 Bq/kg ww The concentrations of beta/gamma-emitting nuclides in fish and mussels from the entire area of the North Sea were mostly undetectable, with the exception of ^{134}Cs and ^{137}Cs, for which values of 0.02 – 0.04 Bq/kg and 0.4 – 1.4 Bq/kg (ww) respectively, were determined (Camplin, 1992).

4.8.4 Radiation exposure from marine radioactivity and reference to recommended values

Radiation exposures from unenhanced sources of natural radioactivity are in most cases higher than those from artificial radioactivity. An estimate of the maximum likely individual dose to man from natural radionuclides amounts to about 2 mSv/yr. Most of the dose that humans obtain by consuming marine food is due to the alpha-emitter ^{210}Po which was found to be more strongly incorporated into several marine organisms than the other radionuclides (Köster *et al*., 1990). For example, consumption of 1 kg of shrimps from the Dutch coastal zone per year causes a radiation exposure of 25 µSv/yr. Discharges from the phospho-gypsum industry lead to radiation doses to seafood consumers from 100 – 380 µSv/yr.

As far as the artificial radionuclides are concerned, the ICRP (International Commission on Radiological Protection, 1991) recommended a principal dose limit received by an individual of 1 mSv/yr. Considering the public radiation exposure due to only artificial radionuclides, only ^{137}Cs is of any possible significance. However, environmental concentrations are very low. Artificial radionuclides contribute to the public exposure in the order of 1% (fish) and less than 1% (shellfish) of the ^{210}Po exposure.

For artificial radionuclides, the dose to man is at least two orders of magnitude less than the dose from natural radionuclides indicated above. Hence their concentrations in the environment are normally of no concern for human health. External exposure of the critical group based on concentrations of radionuclides in sediments is small, amounting to 10 µSv/yr or 1 % of the ICRP-recommended dose limit.

4.9 Nutrients and oxygen

4.9.1 Introduction

Nutrients (dissolved and particulate forms of nitrogen, phosphorus and silicon) play an important role in aquatic ecosystems, as they form the basis for primary productivity. Man-influenced aquatic systems are often characterised by high nutrient concentrations which may entail mass production of algae which in turn, when decomposing, can result in oxygen depletion. Marine areas such as the Kattegat, the eastern Skagerrak, and various Norwegian fjords, all suffer from periodic low oxygen levels (Preston, 1997). Decreased oxygen levels in late summer have been reported even in the central North Sea. Human impacts on nutrient concentration are mainly confined to the near coastal zone.

4.9.2 Sources and inputs of nutrients, input trends

There are various sources of nutrients, and most are linked to anthropogenic activities. Nitrogen in rivers originates mainly from the leaching of agricultural soils and from urban wastewater discharge. Nitrogen in the atmosphere (nitrous oxide and ammonia) originates from domestic and industrial combustion processes, traffic, as well as from agricultural sources such as animal housing and the spreading of manure. Phosphorus is mainly linked to urban wastewater and agriculture.

Figures 4.17 – 4.19 provide an outline of the annual direct, river, and atmospheric inputs of nutrients to the Greater North Sea between 1990 and 1996. Direct inputs decreased for nitrogen and phosphorus, while river inputs increased for nitrogen and phosphorus until 1995, before decreasing in 1996. For silicon, not being a constituent part of RID, there is insufficient information to draw a sensible conclusion. The pattern for riverine input follows the river water flow which may vary considerably from year to year. In particular, riverine inputs of nutrients were elevated by the large flows to the south-eastern part of the North Sea in the early months of both 1994 and 1995. The Fourth North Sea Conference Progress Report (4NSC, 1995) noted that since 1985 there have been substantial reductions in the input of phosphorus, of the order of 50% in ten years. Atmospheric deposition of nitrogen is variable, and no significant trend can be seen.

A major part of the nutrient inputs from land-based sources enter the North Sea via rivers. They account for 65 – 80 % of the total nitrogen inputs and for 80 – 85 % of the total phosphorus inputs. However, care should be taken in interpreting these values in terms of net inputs, since estuarine processes may seriously affect nutrient concentrations. In anoxic conditions, nitrogen compounds can be denitrified to gaseous forms and escape to the atmosphere and phosphorus can be temporarily stored in sediments.

Riverine inputs of phosphorus to the Wadden Sea have been reduced, causing decreased phosphate concentrations in most parts of this area (Bakker *et al.*, 1999). However input trends may vary on a local scale. Danish data for 1989–96 reveal that while direct inputs via waste water to marine areas were reduced by 66% for nitrogen and 81% for phosphorus, less reduction was seen in riverine inputs, especially nitrogen which showed no trend.

Marine inputs of nutrients i.e. those entering the Greater North Sea from adjacent seas, are difficult to assess. Model calculations (Norwecon model) reveal for the period 1976–95 mean nutrient fluxes of 4 000 ± 1 000 kt N/yr, 700 ± 200 kt P/yr and 3 800 ± 1 000 kt Si/yr (Laane *et al*., 1996a). There is a high variability in the annual fluxes which results mainly from drastic changes in the transport of water into the Greater North Sea. Although these fluxes are substantially higher than anthropogenic inputs, it is important to note that only a proportion of these nutrients fluxes is available for primary production.

4.9.3 Fluxes

Internal nutrient fluxes within the North Sea ecosystem include various physical, chemical and biological processes. Processes affecting nitrogen fluxes are internal recycling of organic and inorganic compounds as dissolved inorganic nitrogen (DIN), uptake by phytoplankton, particulate and dissolved organic nitrogen (PON and DON) mineralisation, sedimentation, DON exudation and nitrification. There are also source and sink

Figure 4.17 **(a) Direct discharges and (b) riverine inputs of total nitrogen to the Greater North Sea from 1990 to 1996 in kt/yr. Source: OSPAR (1998a).**

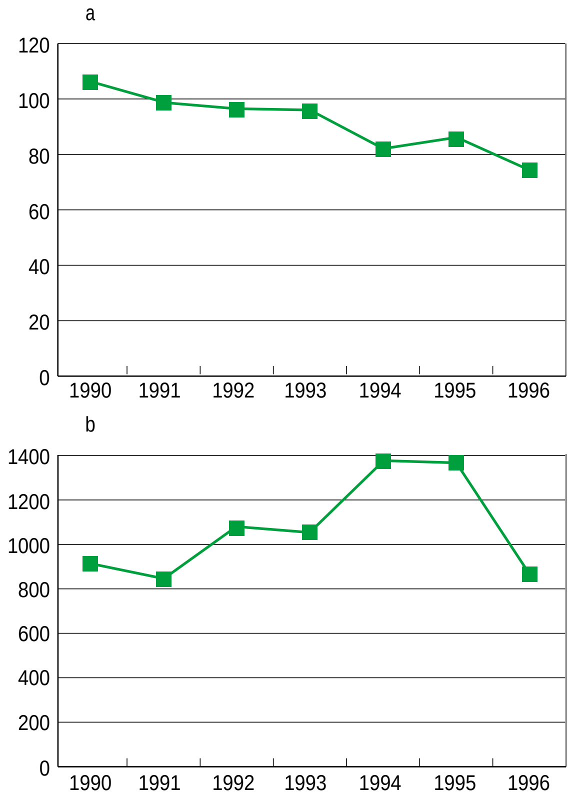
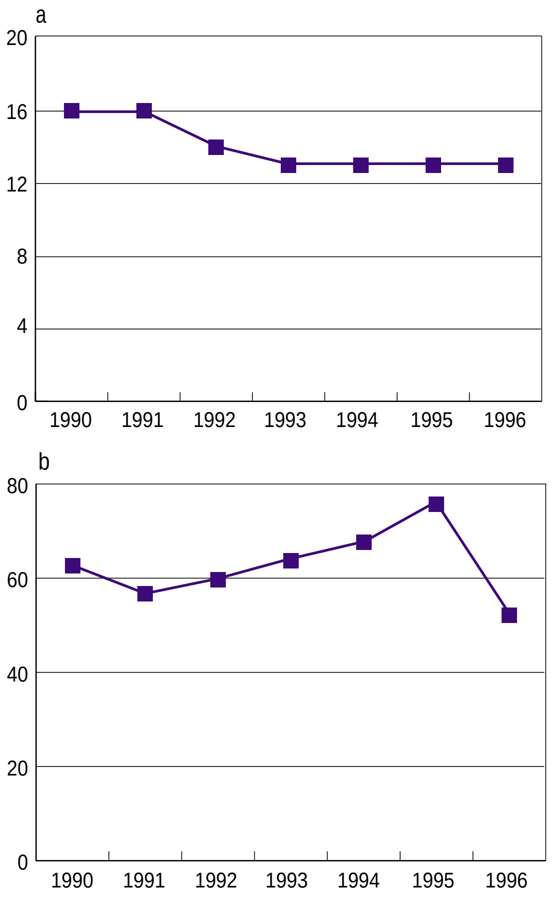


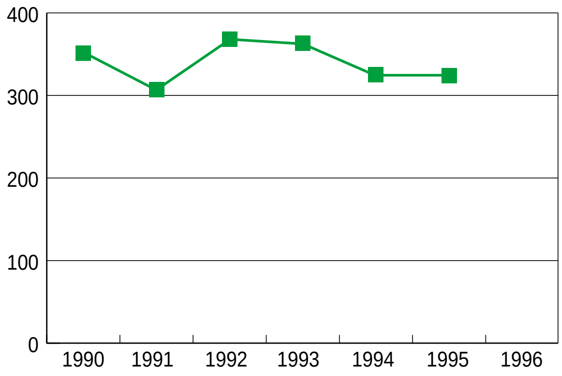
Figure 4.18 **(a) Direct discharges and (b) riverine inputs of total phosphorus to the Greater North Sea from 1990 to 1996 in kt/yr. Source: OSPAR (1998a).**



processes like biological nitrogen-fixation, sediment storage and denitrification. Phosphorus is subject to internal recycling processes such as phosphate uptake by phytoplankton, mineralisation of organic phosphorus, adsorption-desorption to particulate matter and to sink processes like burial. In the same way, silicon is taken up by phytoplankton and submitted to mineralisation and sedimentation processes.

Considering all the internal recycling processes of nitrogen, phosphorus and silicon, recent average data for the North Sea are rather scarce. In particular, there is little recent information about the importance of biological nitrogen-fixation in the North Sea area. It is generally believed to be a minor source of nitrogen in marine systems, although recent estimations made for the North Atlantic seem to show that this source is far from insignificant (Lipshultz and Owens, 1996). Early estimations by Capone and Carpenter (1982) suggest that nitrogen-fixation might account for as much as 2700 kt/yr in the North Sea area, although this value may be overestimated according to more recent observations.

Figure 4.19 **Atmospheric inputs of total nitrogen to the Greater North Sea from 1990 to 1996 in kt/yr. Source of data: OSPAR (1998c).**



Note that no data were available for the year 1996

Denitrification in the North Sea is assessed to be between 0.2 and 0.4 mmol N/m^2/day (Lohse *et al*., 1993). The North Sea sediments could account for a loss of 500 – 1000 kt N/yr.

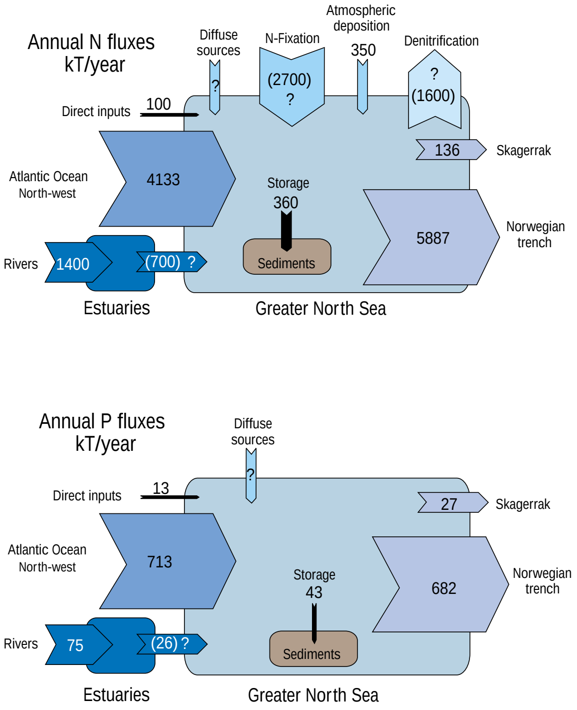
Figure 4.20 represents the summary of the nitrogen and phosphorus balance in the North Sea for the year 1995 including inputs and internal cycling fluxes, based on the approach used by Laane *et al*. (1993, 1996b). The values are in agreement with estimates provided by the European Regional Seas Ecosystem Model calculations, ERSEM (Radach and Lenhart, 1995). Still, as the collected data are from different origins, conclusions have to be drawn with great care. The most important conclusion is certainly the considerable impact of the exchange with the Atlantic Ocean on the nutrient cycling in the North Sea (see Chapter 2 and e.g. *Figure 2.15*).

4.9.4 Concentrations

Nutrient levels and spatial distribution in the Greater North Sea are presented for the years 1993–6 in *Figure 4.21*. Average winter concentrations for these years were not significantly different. Nutrient levels vary from high winter values to low summer values as a result of biological cycling.

Although no clear trend is occurring for the North Sea considered as a whole, this is not the case in local areas that are directly influenced by anthropogenic inputs. For example, a significant decreasing trend (especially for phosphorus) was detected in Danish waters (1989–97) and the German Bight. The decrease of the phosphorus concentrations in nearly all Danish areas is due to a significant decrease in the load from effluents. The same trend was also pointed out for phosphorous in the Wadden Sea (see section 4.9.2).

Figure 4.20 **Annual nitrogen and phosphorus fluxes within the Greater North Sea area. Source of data: sewage and riverine input data: OSPAR (1998b); atmospheric input data: OSPAR (1998c); estuarine inputs calculated according to Nixon *et al*. (1996); Atlantic ocean inputs and sediment storage: Radach and Lenhart (1995); N-fixation: Capone and Carpenter (1982); denitrification value: Seitzinger and Giblin (1996); output calculations to Norwegian Trench and Baltic Sea: budgeting to satisfy steady state conditions (inputs = outputs).**



In the Kattegat and Belt Sea, the winter concentrations of nitrate most often exceed the concentrations determined from pure mixing of Baltic water with Skagerrak water. These excess concentrations are positively correlated to the direct input from land (Denmark, Sweden and Germany), from the atmosphere and also to the freshwater runoff. This means that in this area the local nitrogen load influences the local concentrations significantly. The excess concentrations for phosphate are not strongly correlated to land-based load as sediment release is of great importance to phosphorus concentrations in the Kattegat and the Belt Sea.

4.9.5 Reference to background values

For the reasons given in section 4.3, BRCs were not established for the nutrients nitrogen and phosphorus. The present nutrient distribution shows a significant departure from values mentioned in Table 4.6 especially in areas that are directly under the influence of anthropogenic inputs.

4.9.6 Oxygen

The introduction of large quantities of nutrients can lead to increases in primary production and algal biomass. Degradation of this biomass requires large quantities of oxygen. This can be a major problem in areas with restricted water exchange capacity (such as some Swedish coastal waters) or stratified bodies of water. In these cases major algal blooms have led to serious damage to aquaculture through oxygen depletion and toxin formation (Preston, 1997).

Oxygen depletion can occur in summer periods after water column stratification with limited exchange of dissolved oxygen between the water layers. Generally, since the North Sea is only occasionally stratified, oxygen deficit problems are minor although from time to time they have occurred. However, for stratified waters like the Kattegat, oxygen depletion problems are important (Christensen *et al*., 1998). Statistical analyses of data from Danish fjords have shown that nitrogen loading has the most significant anthropogenic influence in relation to an occurrence of oxygen depletion. Model calculations from the Danish EPA showed that a 50% reduction of the actual nitrogen load would result in an almost equal reduction of the duration of anoxia in these environments (from 493 to 268 days). Since the beginning of the 1990s, no severe oxygen depletion has occurred in the German Bight, but in certain parts low oxygen concentrations still occur occasionally in late summer (e.g., 1994).

4.10 **Observed trends in relation to measures**

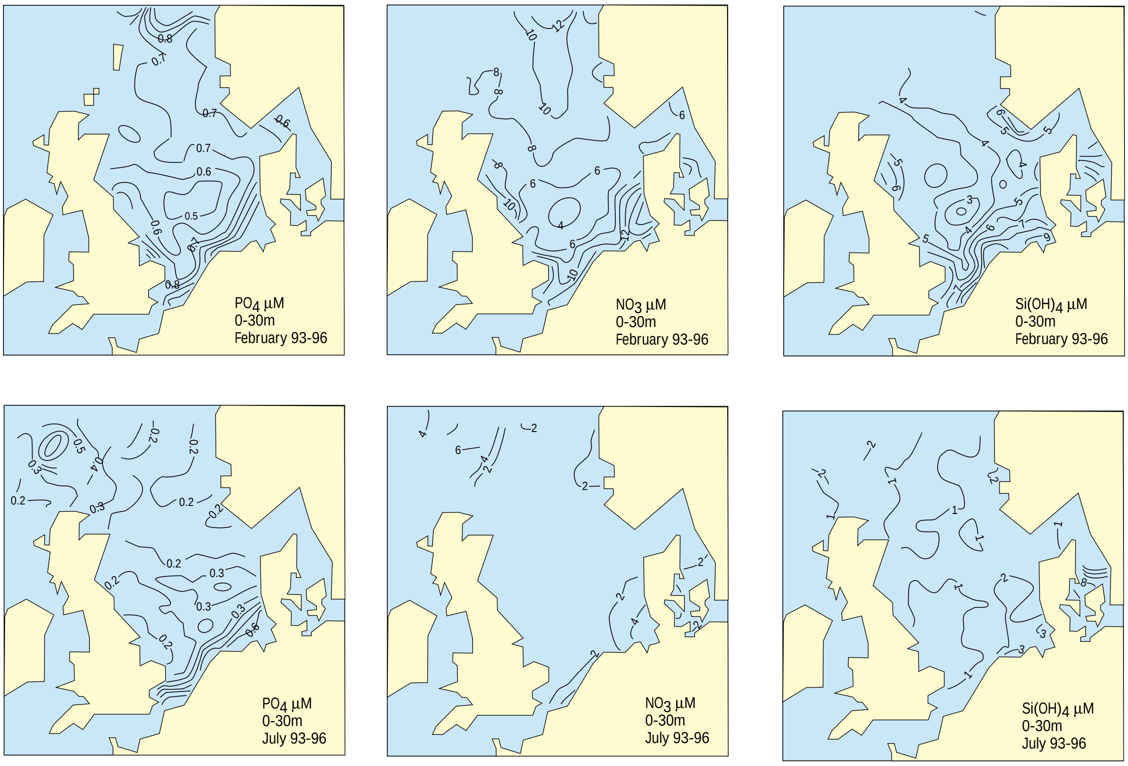
In this section, only clear trends determined with rigorous statistical tests are discussed. Compounds not mentioned here, either do not show any clear trend or insufficient data are available to draw any conclusions. When possible, a link with specific policy measures has been made.

4.10.1 Trace metals

During the last decade, very significant reductions in the discharges and emissions of heavy metals have been achieved by the nine countries involved in the catchment area. Most countries have reached (or are close to) the 50% reduction target set for the period 1985–95.

During this period, cadmium inputs to the receiving waters have been reduced by about 80%. This is mainly due to changes in industrial practice, the adoption and implementation of BAT (Best Available Technology) and

Figure 4.21 **Average distribution (for 1993–6) of phosphate (PO_4^{3-}), nitrate (NO_3^-) and silicate ($Si(OH)_4$) concentrations in surface waters of the Greater North Sea for a winter (February) and a summer (July) situation. Source: Radach and Genkeler (1997).**



to better control of the effluents in the fertiliser, non-ferrous metal, iron and steel and surface treatment industries. Regulations concerning the use of cadmium in batteries and the substitution of cadmium in end products are also involved. The release of cadmium to the atmosphere has been reduced by 55 – 90% during the same period, mainly because of measures taken for flue gas treatment at coal fired power plants and due to the decrease of cadmium emission from waste incineration units.

For mercury, the closing down of important mercury emitters in the former GDR, emission reductions following OSPAR measures (Decision 90/3: a reduction of mercury emissions to less than 2 g/t before 1997, and a recommendation to phase out mercury cells in chlor-alkali plants by 2010) taken at chlor-alkali plants, the installation of gas treatment at power and incineration plants, the substitution of mercury in user products and closer control of diffuse sources (dentistry) have resulted in decreases in discharges of between 60 and 100% to the aquatic environment and of between 25 and 80% in emissions to the atmosphere. The first objective of Decision 90/3 has, on average, been met.

Between 1985 and 1995, waterborne discharges of lead also diminished by between 45 and almost 100% as a result of measures taken in the non-ferrous metals industry, better control of diffuse sources and the recollection of lead batteries. The decrease of between 60 and almost 100% in lead emissions to the atmosphere is the clear consequence of the increasing use of unleaded petrol. (Leaded petrol was banned in Germany in 1988 and in Denmark in 1995. In other countries, reductions in the use of leaded petrol are following the pattern of automobile renewal). Flue gas treatment has also contributed to this trend. The reduction of copper emissions to the air of between 25 and 95%, and to receiving waters of between 25 and 85%, is the result of BAT implementation in the iron, non ferrous and surface treatment industries, and gas treatment at power and incineration plants.

Because of the short residence time of heavy metals in the atmosphere, the various measures taken to control airborne emissions have resulted in a clear decrease in heavy metal deposition to the North Sea (about 30% for cadmium and lead, and 20% for mercury between 1990 and 1995). This pattern has followed the trend already observed in the late 1980s, so that the atmospheric input of cadmium and mercury to the North Sea has been halved since 1987 and reduced by about 60% for lead. This is in contrast with waterborne inputs where only mercury has shown a general decreasing trend over the period 1990–6, while cadmium, lead and copper inputs have not been reduced over the same period (except for 1996). This is mainly attributed to the variation in the hydrological conditions over the period, in particular the large riverine flows to the south-eastern part of the North Sea in 1994 and 1995, but the existence of terrestrial reservoirs (such as contaminated land and sediments) or uncontrolled diffuse sources are also suspected.

As a consequence, only a limited number of decreasing trends have been reported for the concentration of heavy metals in various marine organisms. For seawater and sediments, water bodies such as estuaries, fjords or enclosed seas are more exposed to anthropogenic contamination, but the response to the measures taken is generally more clearly noticeable than in the open sea. This is also the case for areas subjected to dumping. In spite of higher dredging activities, the related contaminant loads have shown a decreasing trend, probably reflecting the reduction of contaminant inputs in rivers and estuaries.

4.10.2 Persistent organic compounds

For persistent organic compounds, data were only available for PCBs and PAHs, but no clear trends can be observed. The use of PCBs was banned in the eighties (more than 90% of the total release of PCBs occurred before 1980; 4NSC, 1995), but despite this measure, only a slow downward trend could be observed due to the fact that polluted sediments act as a source of these contaminants. In the German Bight, α-HCH and γ-HCH have shown a strong decreasing trend over the past 20 years.

In the few places where dioxin levels in biota have been monitored, concentrations have been decreasing since 1990 after reduction measures were taken at industrial sources. However, local concentrations have remained at such a high level that it has led to negative advice on consumption. These high concentrations are probably caused by contaminated sediments and food chain transport.

4.10.3 Oil

In the offshore oil and gas industry, the discharge of oil via cuttings has been strongly reduced (by more than 80%) over a period of ten years, mainly because oil-based drilling mud has been replaced by water-based mud. However, with the amount of oil discharged via production water increasing considerably over the past decade, total oil inputs are increasing slightly after a period of strong reduction.

Illegal discharges from ships continue to be a matter of concern. Oil slicks are still detected along the main shipping corridor in the North Sea, although their frequency and volume seem to have generally decreased.

Application of BAT in the petrochemical industry has been very efficient in reducing the amount of oil discharged. This sector has become a marginal contributor to oil pollution in the North Sea.

4.10.4 Radionuclides

The amounts of artificial radionuclides excluding the less radiologically significant isotopes ^{3}H and ^{99}Tc, discharged annually to the North Sea, have generally decreased over the period 1993–6 and they were much below the permitted limits. Inputs of natural radionuclides, apart from activities related to nuclear facilities, also show a decreasing tendency, mainly due to reduced production of phospho-gypsum by European countries. This development is expected to continue in the future. Radiation exposure from artificial radioactivity is generally much lower than exposure from unenhanced sources of natural radiation and as such it poses no concern for human health.

4.10.5 Nutrients

The general trends for nutrient inputs to the North Sea and nutrient concentration distribution have already been briefly discussed in section 4.9.

The situation concerning nutrients and associated eutrophication effects is rather complex. In those areas almost exclusively influenced by waters from the Atlantic no significant trends in concentrations could be seen over the last decade. A number of areas under the influence of inputs from land-based sources continue to suffer from eutrophication. Trends of decreasing oxygen concentrations, which may be due to eutrophication, enhanced sedimentation and organic matter decomposition, have been documented for deep water in the Kattegat and the basin water in Swedish and Norwegian fjords. However, some improvements with respect to nuisance algal blooms, oxygen depletion and benthos/fish kills are seen in many areas of the North Sea. Significant decreases in concentrations of phosphorus were observed in Danish waters, the German Bight and the Wadden Sea. From 1990–6 direct nitrogen and phosphorus inputs to the North Sea decreased by about 30% and 20% respectively, mainly due to improved waste water treatment. Riverine input increases were mainly due to higher precipitation and, as a result, higher leaching of nitrogen and phosphorus from agricultural soils during 1994–5. For atmospheric inputs, no distinctive changes could be detected.