



Trends in waterborne inputs

Assessment of riverine inputs and direct discharges of nutrients and selected hazardous substances to OSPAR maritime area in 1990 - 2006



OSPAR Convention

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the "OSPAR Convention") was opened for signature at the Ministerial Meeting of the former Oslo and Paris Commissions in Paris on 22 September 1992. The Convention entered into force on 25 March 1998. It has been ratified by Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, Netherlands, Norway, Portugal, Sweden, Switzerland and the United Kingdom and approved by the European Community and Spain.

Convention OSPAR

La Convention pour la protection du milieu marin de l'Atlantique du Nord-Est, dite Convention OSPAR, a été ouverte à la signature à la réunion ministérielle des anciennes Commissions d'Oslo et de Paris, à Paris le 22 septembre 1992. La Convention est entrée en vigueur le 25 mars 1998. La Convention a été ratifiée par l'Allemagne, la Belgique, le Danemark, la Finlande, la France, l'Irlande, l'Islande, le Luxembourg, la Norvège, les Pays-Bas, le Portugal, le Royaume-Uni de Grande Bretagne et d'Irlande du Nord, la Suède et la Suisse et approuvée par la Communauté européenne et l'Espagne.

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Supplementary material (electronically accessible):

Technical supplement 1:	Statistical trend analysis for OSPAR Regions (1990 – 2006)
Technical supplement 2:	Statistical trend analysis for catchments (1990 – 2006)
Technical supplement 3:	Analysis of catchment data in relation to limits of detection
Technical supplement 4:	Complementary graphic presentation of catchment data

Executive Summary

Riverine inputs of heavy metals show in general a significant decrease over the period 1990 – 2006 in the OSPAR Regions Arctic Waters (Region I), Greater North Sea (Region II) and the Celtic Seas (Region III), except for mercury in Region I and lead in Region III. Riverine inputs of nitrogen and phosphorus in the OSPAR Regions I and II show significant decreases over the same period. Direct discharges represent in most cases minor contributions to the total waterborne inputs. Improvements in analytical laboratory techniques over time create difficulties in detecting trends and quantifying reductions accurately, and so contribute to the uncertainties that result from the varying completeness of reporting and monitoring coverage.

Inputs of heavy metals to the OSPAR maritime area are decreasing

Riverine inputs of cadmium, lead and mercury show in many cases statistically significant decreases. Cadmium inputs decreased in Region I (-40%), Region II (-20%) and Region III (-60%) in the period 1990 – 2006. Riverine inputs of lead have fallen in Region I (-85%) and Region II (-50%) and riverine inputs of mercury have significantly reduced in Region II (-75%) and Region III (-85%). For Region II, statistically significant reductions in the main catchments for example cadmium in the Elbe (-40%), mercury in the Rhine and Meuse (-70%) and lead in the Seine (-90%) confirm the overall regional trend. Wide variation in monitoring regimes for rivers and incomplete data on discharges prevent a trend analysis in the Bay of Biscay/Iberian Coast (Region IV).

Direct discharge loads of the three metals from sewage and industrial effluents are much smaller than riverine inputs in most Regions. Reductions achieved in direct discharges of the three metals in the period 1990 – 2006 in Regions I – III range between -70% for cadmium in Region I and mercury in Region II and -95% for cadmium and mercury in Region III.

Progress in reducing waterborne inputs of the metals to the marine environment since 1998 has been less marked than in the early 1990s.

Riverine inputs of nitrogen and phosphorus have decreased in 1990 – 2006 in the Regions I and II, but no changes could be detected in the Regions III and IV

Riverine inputs of nitrogen to the sea have decreased to varying degrees in the period 1990 - 2006 with statistically significant downward trends in Region I (-50%) and Region II (-25%). There is no trend in riverine inputs in Regions III and IV. Phosphorus inputs show similar regional patterns with reductions in riverine inputs of nitrogen in Region I (-55%) and Region II (-50%) and no trends in Regions III and IV. Large decreases in the nitrogen loads carried by the rivers Elbe and Rhine and the phosphorus loads carried by the rivers Seine, Elbe, Rhine and Meuse underlie the clear fall in river inputs to Region II.

Direct discharges significantly decreased in Region II (-35%) and Region III (-30%) for nitrogen and in Region III (-50%) for phosphorus in the period 1990 – 2006. In Region I, the increase of direct discharges of nitrogen and phosphorus reported since 2000 is a result of the growing mariculture production and offsets the decrease in riverine inputs. Direct discharges account now for the greater fraction of waterborne inputs in Region I. In all other Regions, direct discharges are the minor part of the waterborne nutrient inputs to the sea.

Récapitulatif

Les apports fluviaux de métaux lourds accusent, dans l'ensemble, une baisse significative entre 1990 et 2006 dans les Régions OSPAR, eaux arctiques (Région I), mer du Nord au sens large (Région II) et mers celtiques (Région III), à l'exception du mercure dans la Région I et du plomb dans la Région III. Les apports fluviaux d'azote et de phosphore dans les Régions OSPAR I et II accusent des baisses significatives durant la même période. Les rejets directs représentent, dans la plupart des cas, des contributions minimes au total des apports aquatiques. L'amélioration de techniques analytiques de laboratoire dans le temps entraîne des problèmes lorsqu'il s'agit de déterminer des tendances et de quantifier avec précision les réductions. Elle contribue donc aux incertitudes qui découlent des divers états de complétude de la notification et de la couverture de la surveillance.

Les apports de métaux lourds dans la zone maritime OSPAR sont en baisse

Les apports fluviaux de cadmium, de plomb et de mercure accusent dans de nombreux cas des baisses statistiquement significatives. Les apports de cadmium ont diminué dans la Région I (-40%), la Région II (-20%) et la Région III (-60%) entre 1990 et 2006. Les apports fluviaux de plomb ont baissé dans la Région I (-85%) et la Région II (-50%) et les apports fluviaux de mercure ont baissé de manière significative dans la Région II (-75%) et la Région III (-85%). Des réductions statistiquement significatives dans les principaux bassins hydrographiques de la Région II confirment la tendance régionale d'ensemble. Il s'agit par exemple du cadmium dans l'Elbe (-40%), du mercure dans le Rhin et la Meuse (-70%) et du plomb dans la Seine (-90%). Des variations importantes dans les régimes de surveillance des fleuves ainsi que des données incomplètes sur les rejets ne permettent pas de réaliser une analyse des tendances dans le Golfe de Gascogne/la Côte ibérique (Région IV).

Les charges des rejets directs des trois métaux provenant des eaux usées et des effluents industriels sont beaucoup moins grandes que celles des apports fluviaux dans la plupart des régions. Les réductions obtenues pour les rejets directs des trois métaux entre 1990 et 2006 dans les Régions I à III se situent entre -70% pour le cadmium dans la Région I et le mercure dans la Région II et -95% pour le cadmium et le mercure dans la Région III.

Les progrès réalisés en ce qui concerne les apports aquatiques de métaux dans le milieu marin depuis 1998 sont moins marqués qu'au début des années 1990.

Les apports fluviaux d'azote et de phosphore ont diminué entre 1990 et 2006 dans les Régions I et II, mais on ne relève aucun changement dans les Régions III et IV

Les apports fluviaux d'azote à la mer ont diminué dans diverses mesures entre 1990 et 2006, des tendances à la baisse statistiquement significatives étant relevées dans la Région I (-50%) et la Région II (-25%). On ne relève aucune tendance pour les apports fluviaux dans les Régions III et IV. On relève des tendances régionales similaires pour les apports de phosphore, des réductions des apports fluviaux d'azote étant relevées dans la Région I (-55%) et la Région II (-50%) et aucune tendance dans les Régions III et IV. Des diminutions importantes des charges d'azote transportées par l'Elbe et le Rhin et des charges de phosphore transportées par la Seine, l'Elbe, le Rhin et la Meuse soulignent la baisse nette des apports fluviaux dans la Région II

Les rejets directs d'azote ont diminué de manière significative dans la Région II (-35%) et la Région III (-30%) et ceux de phosphore dans la Région III (-50%) entre 1990 et 2006. L'augmentation des rejets directs d'azote et de phosphore dans la Région I notifiés depuis 2000 découle de l'exploitation maricole croissante et est compensée par la diminution des apports fluviaux. Les rejets directs représentent maintenant la majeure partie des apports aquatiques dans la Région I. Dans toutes les autres Régions, les rejets directs représentent une partie minime des apports aquatiques de nutriments à la mer.

Extended Executive Summary

What is the problem?

Pollution of the North-East Atlantic with contaminants and nutrients and associated adverse effects on the marine ecosystem is a well documented problem. Eutrophication caused by anthropogenic nutrient enrichment is a problem in many areas of the Greater North Sea, Celtic Seas and, to a lesser extent, the Bay of Biscay. Contamination of sediments and biota with cadmium, lead and mercury are still at unacceptable levels in many coastal areas of Regions II, III and IV. Except for the Wider Atlantic region where the only human populations are associated with the Azores archipelago, an important pathway of contaminants and nutrients to the sea is via rivers, which collect pollutants from land-based point sources (*e.g.* sewage plants and industry) and diffuse losses (*e.g.* agriculture, run-off from land and atmospheric deposition in river basins). There are also large reservoirs of contaminants in river sediments due to the legacy of the past. Contaminants and nutrients are also directly discharged into the sea for example through waste water outlets or activities in coastal waters such as marine aquaculture.

What has been done?

OSPAR's objective is to achieve a healthy and clean sea by continuously reducing releases of hazardous substances and nutrients to the environment. Cadmium, lead and mercury have been identified for priority action and every effort should be made to move towards ceasing emissions, discharges and losses by 2020. Discharges and losses of nitrogen and phosphorus should be reduced by 50% compared to input levels in 1985 in areas where their input can contribute to eutrophication problems. OSPAR and EC measures target emissions, discharges and losses of heavy metals and nutrients from point and diffuse sources. This includes for example the Integrated Pollution Prevention and Control (IPPC) Directive (2008/1/EC), the Urban Waste Water Treatment Directive (91/271/EEC), Nitrates Directive (91/676/EEC) and EC water legislation including the Water Framework Directive (2000/60/EC).

What does monitoring tell us?

Long-term monitoring of inputs of pollutants to coastal waters provides an indication of whether measures implemented are working. The purpose of the OSPAR Comprehensive Study on Riverine Inputs and Direct Discharges (RID Study) is to assess, as accurately as possible, all riverine and direct inputs of selected pollutants to the OSPAR maritime area on an annual basis and to review data periodically to determine their temporal trends. Under the RID Study Contracting Parties monitor and report annually since 1990 inputs of selected heavy metals and nutrients via rivers and via direct discharges to the sea from sewage, industrial effluents, and more recently marine aquaculture. The RID Principles set out the monitoring regime to be employed for generating and reporting input data and to this end describe for example the relevant substances and river systems covered, sampling approach, locations and frequency, detection limits, calculation methodologies and quality assurance issues (OSPAR, 1998). Quality assurance issues that need to be considered include:

- data quality (e.g. statistical representativeness and accuracy);
- harmonisation and transparency in procedures in compliance with the RID Principles;
- reliability (methods in measurements, analyses, uncertainty), and
- comparability of results, procedures and tools.

There is no doubt that management decisions linked to the environment have become more complex over the most recent years. This is due to more integrated and cross-sector approaches such as required in the implementation of the EC Water Framework Directive. Consequently, this requires water quality monitoring programmes to provide data for multiple purposes, including trend analyses to detect improvement or deterioration in water quality with time (Box 1).

There is a considerable geographical span in the areas monitored under the RID Study ranging from Iceland, Norway and Sweden in the northern part of Europe, through Denmark,

Box 1

Methods improved in 1990 – 2006

Improvements in analytical laboratory techniques between 1990 and 2006 enable concentrations of many parameters to be detected at much lower levels and with higher accuracy and precision than in earlier years. This makes trend detection and an accurate quantification of the reductions more difficult. Varying completeness of reporting, monitoring coverage and differences in analytical performances contribute to uncertainties.

Germany, the Netherlands, Belgium and the UK in more central parts of Europe, to France, Spain and Portugal in southern Europe. The variations in climate, geographical location, implementation of measures, population density and land-use pattern are all 'external' elements that influence concentrations of pollutants discharged with rivers, effluents from industrial plants and municipal waste water treatment plants, and the magnitude of losses from agricultural activities.

The analyses of the reporting of riverine inputs and direct discharges for the selected pollutants to the OSPAR maritime area and its Regions in the period 1990 – 2006 have identified inconsistencies and challenges with regard to sound and reliable analyses (Box 2).

In terms of geographic coverage, the OSPAR Region II (Greater North Sea), especially the main body of the North Sea, is the maritime area that is most completely covered in terms of complete time series and monitored rivers, although gaps exist. This region also includes some of the most important catchments contributing heavy metals and nutrients to the OSPAR maritime area, such as those of the rivers Rhine and Elbe. Data coverage for heavy

Box 2

Challenges in estimating direct discharges

There are large variations in the way that direct discharges are accounted for amongst Contracting Parties. Some report on direct discharges from sewage treatment plants, industries and fish farming, others do not report any direct discharges at all. Given that large cities are located along the shores of the marine coastline for basically all relevant OSPAR countries, the reported discharges can have been seriously underestimated. However, direct discharges appear to be relatively less important than riverine inputs.

metals in Region IV (Bay of Biscay and Iberian Coast) has been increasing throughout the period, including reporting on an increasing number of rivers. Region IV is the region which is most affected by uncertainties due to changes in monitoring methods.

There has been no trend in river flow in the period 1990 – 2006

The volume of water flow in rivers influences the inputs to the sea of the different pollutants. One challenge is to separate the human-induced (anthropogenic) variability and trends from the natural variability (hydro-meteorology). Even if this theoretically can be included in formal statistical trend analysis methods, finding the causal and underlying mechanisms for water quality changes still remains a challenge. Some of the assessed datasets have been adjusted for water flow in the trend analysis where the river loads observed were flow-dependent and adjustment was advised. There is no detected trend in the annual water volume discharged by rivers to the OSPAR maritime area during the period 1990 – 2006 (Figure 0.1).



Figure 0.1: Trends in river flow in 1990 - 2006 in Regions I, II and III of the OSPAR area

Inputs of heavy metals to the OSPAR maritime area are decreasing but most reductions occurred before 1998

In general, improved treatment of effluents from industrial plants and municipal waste water treatment has reduced discharges of heavy metals from point sources to surface waters. This is due to, *inter alia*, the implementation of best available techniques in large industrial installations and associated discharge limit values under OSPAR and subsequent EC measures under the IPPC Directive (2008/1/EC). EC measures that will contribute to further improvements include the Water Framework Directive (2000/60/EC), the Dangerous Substances Directive (2006/11/EC) and the Drinking Water Directive 98/83/EC, as well as national measures to combat pollution from heavy metals.

In the OSPAR maritime area, input levels of heavy metals have decreased in most regions. However, not in all cases statistically significant trends have been detected (see Table 0.1 for trends in riverine inputs).

Table 0.1: Statistically significant trends in riverine inputs of heavy metals to the OSPAR Regions in 1990 – 2006

Riverine inputs	Region I	Region II	Region III	Region IV
Cadmium	-40% ↓	-20% ↓	-60% ↓	No trend
Copper	-45% ↓	-40% ↓	-30% ↓	No trend
Lead	-85% ↓	-50% ↓	No trend	No trend
Mercury	No trend	-75% ↓	-85% ↓	No trend

Cadmium

There is a statistically significant downward trend in riverine inputs of cadmium to Region I (Arctic Waters) of about 40%, Region II (Greater North Sea) of about 20% and Region III (Celtic Seas) of about 60% during the period 1990 – 2006. *In the Arctic Waters* there has been a steady decrease also since 1998, with one slight increase in 2004. Compared to riverine inputs, the direct discharges of cadmium were low throughout the period. There is a statistically significant downward trend of direct discharges of about 70% in this region during the period 1990 – 2006. *In the Greater North Sea* region the



Figure 0.2: Total riverine inputs and direct discharges of cadmium to Regions I – III in 1990 – 2006

regional trend is supported by a 40% downward trend in the river Elbe. There has been a steady decrease since 1998, with one slight increase in 2002. The direct discharges represent less than 10% of the total inputs and show a 95% downward trend. *In the Celtic Sea Waters*, the riverine inputs in 2006 were at the same level as in 1998, with a peak in the year 2000. The direct discharges represented a minor contribution to the total inputs to this region. They showed a downward trend of about 95%. Finally, *in Bay of Biscay and Iberian Coast* no trend analysis could be undertaken due to varying monitoring regimes and numbers of rivers monitored. An increase in the input over recent years could be attributed to the increased number of monitoring locations.

Copper

There is a statistically significant downward trend in riverine inputs of copper to Region I (Arctic Waters) of about 45%, to Region II (Greater North Sea) of about 40% and Region III (Celtic Seas) of about 30% during the period 1990 – 2006. *In the Arctic Waters*, there has been a steady decrease also since 1998, with one slight increase in 2004. Direct discharges contributed 5 – 20% to the total inputs of copper in 1990 – 2004. Since 2005, direct discharges are more important than riverine inputs due to losses of copper from antifouling treatment of net cages used in marine aquaculture. *In the Greater North Sea*,



Figure 0.3: Total riverine inputs and direct discharges of copper to Regions I - IV in 1990 – 2006

the regional trend is supported by similar trends in the rivers Seine, Meuse and Rhine. There has also been a steady decrease since 1999, with one slight increase in 2001. The direct discharges represented on average less than 15% of the total inputs with a downward trend of about 70%. *In the Celtic Seas*, the riverine inputs in 2006 were higher than in 1998, with lower values in the years in between. Since 2002/2003 the direct discharges of copper represent a minor portion of the total inputs of copper to the region. Over the whole period they show a downward trend of about 90%. *In the Bay of Biscay and Iberian Coast*, due to changed monitoring strategies and numbers of rivers monitored (thus higher loads detected), no trend analysis could be undertaken.

Lead

There is a statistically significant downward trend in riverine inputs of lead to Region I (Arctic Waters of about 85% and Region II (Greater North Sea) of about 50% during the period 1990 - 2006. In the Arctic Waters, there has been a steady decrease also since 1998. Direct discharges contributed 20 -30% to the total inputs of lead. In the Greater North Sea, the regional trend is supported by a 90% decline in riverine inputs from the river Seine. There has also been a steady decrease since 1999, with one slight increase in 2000. Direct discharges represented on average less than 5% of the



Figure 0.4: Total riverine inputs and direct discharges of lead to Regions I – IV in 1990 – 2006

total inputs and showed a downward trend of about 80%. *In the Celtic Seas*, the total inputs of lead in 2006 were at about the same level as in the beginning of the 1990s. The riverine inputs in 1998 were slightly higher than in 2006 with much lower levels in the years 2003 – 2005. Since 2003 the direct discharges of lead represent a minor portion of the total inputs of lead to the Region. *In the Bay of Biscay and Iberian Coast*, no trend analysis could be undertaken due to varying monitoring regimes and numbers of rivers monitored. An increase in the input over recent years could be attributed to the increased number of monitoring locations.

Mercury

There is a statistically significant downward trend in riverine inputs of mercury to Region II (Greater North Sea) of about 75% and Region III (Celtic Seas) of about 85% during the period 1990-2006. *In the Arctic Waters,* there is no clear statistical trend in inputs of mercury during the period 1990 – 2006. Since 1998 there have been strong interannual variations in riverine inputs with a peak in 2003 (six times higher than in 1998). Direct discharges contributed to less than 15% to the total inputs of mercury most of the period. *In the Greater North Sea,* the regional trend is supported by a 70% decline



Figure 0.5: Total riverine inputs and direct discharges of mercury to Regions I – III in 1990 – 2006

in riverine inputs from the rivers Rhine and Meuse. There has been a steady decrease also since 1998, with a slight increase in 2002. The direct discharges were of marginal importance, but showed a 75% downward trend over the period. *In the Celtic Seas*, there has been a strong decrease in inputs since 1998. In the period 1990 – 2003, direct discharges of mercury represented a substantial portion of the total inputs of mercury. *In the Bay of Biscay and Iberian Coast*, no trend analysis could be undertaken due to varying monitoring regimes and numbers of rivers monitored.

Riverine inputs of nitrogen and phosphorus have decreased in 1990 – 2006 in the Regions Arctic Waters and Greater North Sea. No real changes in riverine inputs could be detected in the Regions Celtic Seas and Bay of Biscay/Iberian coast.

In general, improved waste water treatment e.g. due to the Urban Wastewater Treatment Directive (91/271/EEC) and the IPPC Directive (2008/1/EC) have reduced nitrogen and especially phosphorus discharges to surface waters. The implementation of PARCOM Recommendation 88/2 on the Reduction in Inputs of Nutrients to the Paris Convention Area, which aims at a reduction of 50% of inputs to areas affected by eutrophication compared to input levels in 1985, is still ongoing. It has set in motion a number of initiatives to reduce discharges/losses of nitrogen and phosphorus from in particular waste water treatment plants and agricultural activities. Furthermore, in the Nordic Countries reductions of discharges from point sources have already taken place well before 1990 as a result of improved waste water treatment. Measures as a consequence of the Nitrates Directive (91/676/EEC) and Drinking Water Directive (98/83/EC), and other measures implemented, have also reduced nitrogen losses, especially from agriculture. The ban on phosphorus in detergents has further contributed to reduced phosphorus discharges/losses to surface waters. These achievements are reflected in falling overall input levels of nitrogen and phosphorus in the regions of Arctic Waters and the Greater North Sea in 1990 – 2006 (Table 0.2).

Table 0.2: Statistically significant trends of riverine inputs of nitrogen and phosphorus to the OSPAR Regions in 1990 – 2006

Riverine inputs	Region I	Region II	Region III	Region IV
Nitrogen	-50% ↓	-25% ↓	No trend	No trend
Phosphorus	-55% ↓	-50% ↓	No trend	No trend

Nitrogen

In general, direct discharges only constitute a minor part of total inputs of nitrogen to the sea, except for the Arctic Waters where discharges reported since 2000 are higher than the riverine inputs. This is due to increased marine aquaculture production.

There is a statistically significant downward trend in riverine inputs of nitrogen to Region I (Arctic Waters) of about 50% and Region II (Greater North Sea) of about 25%. *In the Arctic Waters*, there is a marked decrease since year 2000 in riverine inputs. When the direct discharges and riverine inputs are assessed together there is no clear trend.



nitrogen to Regions I - IV in 1990 – 2006

The reason is a strong increase in marine aquaculture production resulting in increased direct discharges to the sea and the fact that discharges from marine aquaculture have not been reported prior to 2000. *In the Greater North Sea*, the downward trend is supported by monitoring results in several major rivers such as the Elbe, Meuse and Rhine, showing statistically significant downward trends ranging between 25 and 45%. *In the Celtic Sea* and the *Bay of Biscay and Iberian Coast*, there is no clear trend in the riverine nitrogen inputs for the period 1990 – 2006. However, there is a statistically significant downward trend of about 30% for direct nitrogen discharges to Region III (Celtic Seas).

The main part of the reduction in riverine inputs of nitrogen to Region II (Greater North Sea) and Region III (Celtic Seas) since 1990 occurred in the period 1998 – 2006, taking into account the influence of variations in runoff.

Phosphorus

Direct phosphorus discharges to Arctic Waters are higher than the riverine inputs. In the Celtic Seas, direct discharges and riverine inputs of phosphorus are more or less of the same magnitude. In the Greater North Sea direct discharges are markedly lower than the riverine inputs.

There is a statistically significant upward trend in total inputs (direct discharges and riverine inputs) of phosphorus to Region I (Arctic Waters) of about 55%, and a downward trend in Region II (Greater North Sea) of about 50%. *In the Arctic Waters*, the upward trend can be explained by a statistically significant upward trend in direct



Figure 0.7: Total riverine inputs and direct discharges of phosphorus to Regions I - IV in 1990 – 2006

phosphorus inputs of about 100%. This mainly reflects increased marine aquaculture production and the fact that direct discharges of phosphorus from fish farming have not been reported prior to 2000. However, riverine inputs in Region I show a statistical significant downward trend of about 55%. *In the Greater North Sea* the downward trend is supported by monitoring results in several major rivers such as the Seine, Elbe and Meuse, showing statistically significant downward trends of about 40 - 80%. *In the Celtic Seas* and the *Bay of Biscay and Iberian Coast*, there is no clear trend in riverine inputs of phosphorus in the period 1990 – 2006. However, there is a statistically significant downward trend of about 50% in direct discharges of phosphorus and of about 40% in total phosphorus inputs (riverine and direct) to the Celtic Seas.

Taking into account the influence of variations in runoff, the main part of the reduction in riverine phosphorus inputs to Region III occurred after 1998. The phosphorus reduction to Region II during 1998 to 2006 is within the same range as the reductions during 1990 to 1998. For Region I, riverine phosphorus inputs seem to increase, but this could be due to missing data of agricultural phosphorus losses before 2000 in this Region.

Electronic navigator to complementary QSR assessments and documentation

QSR assessments

- Trends in atmospheric concentrations and deposition (OSPAR, 2009a)
- Trends and concentrations in marine sediments and biota (OSPAR, 2009b)
- Status and trends of marine chemical pollution (OSPAR, 2009c)
- → Towards the cessation target for priority chemicals (OSPAR, 2008a)
- Eutrophication status of the OSPAR area (OSPAR, 2008b)

Complementary documentation

- → RID Principles (agreement 1998-5)
- Annual RID data reports. Latest report taken into account OSPAR (2008c)
- → 2005 RID data assessment (OSPAR, 2005)

1. Introduction

1.1. Scope and context

This report is the second comprehensive scientific assessment of trends in data collected under the OSPAR Comprehensive Study on Riverine Inputs and Directive Discharges ("RID Study") (agreement 1998-5). A first assessment of input trends in the OSPAR Regions and sub-regions of the Greater North Sea was undertaken in 2005 for the period 1990 – 2002 (OSPAR, 2005). This report describes the results of the trend analysis of data collected under the RID Study in the period 1990 – 2006. The assessment is a contribution to the Quality Status Report 2010 and provides supporting evidence for conclusions on progress towards the specific targets of the OSPAR Hazardous Substances Strategy and Eutrophication Strategy to reduce discharges and associated riverine inputs of pollutants to the North-East Atlantic (for complementary QSR assessments and RID documentation see electronic navigator box p. 12).

The RID Study forms one element within the wider Joint Assessment and Monitoring Programme of OSPAR. The purpose of the RID Study is to assess, as accurately as possible, all riverine and direct inputs of selected pollutants to the OSPAR maritime area on an annual basis. The RID Principles define the monitoring regime to be employed for generating and reporting input data. They describe, for example, the relevant substances and river systems covered, the sampling approach, locations and frequency, detection limits, calculation methodologies and quality assurance to be applied.

Under the RID Principles, Contracting Parties should aim to monitor at least 90% of the inputs of each selected pollutant on a regular basis. A number of determinants are to be monitored on a mandatory basis and this assessment considers six of these determinants:

- total cadmium (Cd);
- total copper (Cu);
- total mercury (Hg);
- total lead (Pb);
- total nitrogen (N, or tot-N), and
- total phosphorus (P, or tot-P).

Water discharge (Q) was included in the assessment.

Sources for monitoring and reporting of direct discharges under the RID Principles include sewage and industrial effluents downstream river monitoring points and aquaculture. As far as practicable, estimate inputs from unmonitored areas (including diffuse sources and minor direct sources and rivers) should complement the percentage monitored to 100%. As a rule, this assessment has not taken account of diffuse losses from unmonitored areas as reporting on these sources is considered highly insufficient.

1.2 Objectives

The objectives of the work were:

- a. to identify and assess the trends in riverine inputs and direct discharges of the six determinants referred to above to:
 - i. OSPAR Regions I IV (Figure 1.1; Regional approach)
 - ii. ten catchments within the OSPAR Regions I IV (Catchment approach)
- b. to assess the synergies between these Regional and Catchment approaches.



Figure 1.1: Map showing the OSPAR maritime area, with Region I (Arctic Waters), Region II (Greater North Sea), Region III (Celtic Seas), Region IV (Bay of Biscay and Iberian Coast) and Region V (Wider Atlantic)

1.3 Work carried out

- The statistical analyses carried out on OSPAR Regions I IV were performed by Steffen Uhlig, Kirstin Kunath, Michaela Adamcova (quo data GmbH, Germany) and Mark Moens (MUMM, Belgium). This work was financed by the German Federal Ministry for the Environment, Nature Protection and Nuclear Safety;
- The statistical analyses carried out on the eight catchments were carried out by Steffen Uhlig, Kirstin Kunath, Michaela Adamcova (quo data GmbH, Germany). This work was financed by the German Federal Ministry for the Environment, Nature Protection and Nuclear Safety;
- c. The assessment of the analyses and the drafting of the reports on the four Regions and the Catchments were done by Stig A. Borgvang, Per Stålnacke, Ståle Haaland, Annelene Pengerud and Paul Andreas Aakerøy (Bioforsk, Norwegian Institute for Agricultural and Environmental Research, Norway). This work was financed by the Norwegian Pollution Control Authority.
- d. The assessment of the water discharge analyses was carried out by Jens Fölster (Swedish University of Agriculture (SLU), Sweden).

The data used were validated by Contracting Parties prior to the analyses.

The national reports submitted by each Contracting Party since 1990 were studied one by one and the comments made in these reports were summarised on a country by country basis.

2. Methodology and input data

2.1. General information on OSPAR Regions

2.1.1 Region I: Arctic Waters

The only contribution taken into account for this region is from Norway through the Barents and the Norwegian Seas. The drainage area of the Arctic Sea Waters covers approximately 187 000 km². Due to too short time series reported by Iceland, these data were excluded.

2.1.2 Region II: Greater North Sea

This Region includes the following sea areas and countries: North Sea (Belgium, the Netherlands, Germany, Denmark, Norway and the UK), Skagerrak (Denmark, Sweden and Norway), Kattegat (Sweden and Denmark), and the Channel (UK and France). The drainage area of the Greater North Sea Region is about 910 000 km².

2.1.3 Region III: Celtic Seas

Region III includes the Irish Sea and the whole of Ireland and the western part of the UK. It extends between 60° N and 48° N and between 5° W and the west coast of Great Britain to the 200 m depth contour to the west of 6° W. The drainage area of Region III covers 102 000 km².

2.1.4 Region IV: Bay of Biscay and Iberian Coast

The Bay of Biscay and Iberian Coast region extends from 48°N to 36° N and from 11° W to the coastlines of France, Spain and Portugal. The drainage area for Region IV covers 655 000 km².

2.2 Monitoring data

According to the RID Principles (agreement 1998-5), the aim of the RID Study is "to monitor on a regular basis at least 90% of the inputs of each selected pollutant". Both riverine inputs and direct discharges are being monitored. A main river is defined as "a river to be monitored at least once a month (12 datasets) every year in accordance with the objectives of the comprehensive study as set out in paragraph 1.4. Main rivers should be major load bearing rivers". A tributary river is defined as "a river downstream of a river monitoring point. A tributary river should be a minor load bearing river and can be sampled at a frequency determined by each Contracting Party". Direct discharges are anthropogenic discharges from industry, sewage treatment plants and aquaculture.

In addition to the monitoring of rivers and direct discharges it is desirable to "as far as is practicable estimate inputs from unmonitored areas (including diffuse sources, and minor direct sources and rivers) complementing the percentage monitored".

2.3 Statistical methodology

2.3.1 Data used

The data had been validated by Contracting Parties. Although through the initial screening it became apparent that there were possible errors in some of the data, it was decided to use all data without deleting or correcting any potential outliers. Since the statistical methods used are not sensitive to outliers, single erroneous annual data will have negligible impact on the results. The data for Regions were aggregated from sub-region data reported by each Contracting Party.

2.3.2 Preparation of datasets

The regional assessment was carried out only on consistent time series, *i.e.* it should be guaranteed that there are no missing data in the underlying OSPAR sub-regions for the entire time period considered. For each of the sub-regions and parameters with missing data it had to be decided whether the gaps can be filled with estimated data or whether the whole series could not be taken into account. The following rule was established in order to guarantee reproducible results:

If in the input series of a parameter and a sub-region 3 or more subsequent years were missing or if there were less than 10 years of data are available for the period 1990 – 2006, the whole time series of this sub-region and this parameter were to be excluded from the analyses. Exception: If there were only 3 missing data not at the margins of the time series and if the variation within the time series is low, data can be accepted. In case of acceptance, missing data of riverine inputs and discharges were estimated with an appropriate estimation method (LOESS or Theil slope, adjusted or unadjusted load).

This procedure avoided spurious trends caused by missing data. However, it was not possible to examine whether the input figures for the sub-regions themselves took into account all rivers of the respective area.

2.3.3 Treatment of minimum/maximum data (less than LOD) for regional data

At the OSPAR RTrend workshop (16-17 October 2007, Linköping, Sweden) it was agreed to introduce an additional statistical evaluation in RTrend focussing on the maximum impact that measurement results below the Limit of Detection (LOD) may have on computed linear trends. For this reason a new module was implemented in RTrend that would calculate a worst case trend. The worst case trend is a trend with minimum steepness derived from the lower and upper estimates each year. If the worst case trend is still significantly different from zero, it may indicate that this trend is not caused by measurement results below the LOD. It needs to be mentioned that sometimes it was not clear whether LODs were reported or LOQs (Limits of Quantification). LODs have been used to address LOD and LOQ collectively.

As the maximum impact of measurements below the LOD could then easily be assessed by comparing the worst case trend with the trend based on the annual averages (lower + upper estimates)/2, it was decided to use the annual averages for the calculation of the trends. Note that in the 2005 RID data assessment for nutrients the maximum figures were used.

2.3.4 Treatment of data below LOD in catchment analyses

Data reported below the LOD were replaced by LOD/2. Thus the maximum bias due to data below the LOD could be minimised. It should be noted however that spurious trends can be caused by temporal trends in the LOD. As this is relevant for several rivers and determinants, estimated LOESS levels are also given when data below the LOD were replaced by both the LOD itself and 0.

2.3.5 p-Value and significance level

The statistical significance of a (test) result is an estimated measure of the degree to which it is "true" (in the sense of "representative of the population"). The smaller the p-value, the more strongly the test rejects the null hypothesis (*i.e.* the hypothesis being tested). A p-value of 0.05 – or less – rejects the null hypothesis "at the 5% level". This means that the statistical assumptions used imply that in only in 5% of the time the supposed statistical process would produce a finding this extreme if the null hypothesis were true. Values of 5% and 1% are common significance levels to which p-values are compared. In the RID trend assessment it means that the lower the p-value, the higher the significance of the trend.

When significance is used in the assessment, the statistical significance is meant. The following values were used in this assessment:

- Insignificant: p >0.05
- Significant: 0.01< p < 0.05
- Highly significant: p < 0.01

2.3.6 Flow-adjustment and trend assessment

The flow-adjustment and trend assessment procedures are given in Annex I.

Trend analyses were performed with non-parametric methods that are not sensitive to outliers. When concentrations or transports (loads) were significantly correlated to river discharge, trend tests were additionally made on flow adjusted data. The flow adjustment procedures assume linear or log-linear relationships between discharge and transport or flow that are constant over time. For further details, see Annex I and JAMP Guidance on Input Trend Assessment and the Adjustment of Loads (OSPAR agreement 2003-9).

This report contains the assessment and a summary of the data and the statistical analyses. The full statistics and complementary data are available electronically as <u>Technical Supplement 1</u> and <u>Technical Supplement 2</u>.

2.3.7 Software

The analysis was carried out with an upgraded version of RTrend (RTrend RID A 3.3.0.0). The upgrade concerned:

- the consideration of minimum and maximum annual inputs;
- the calculation of the worst-case trend due to measurements below the LOD;
- a new consistency check of the data;
- a simplified import, and
- a simplified user interface.

3. **PART I – Synthesis of the Region and Catchment reports**

3.1. Introduction

The OSPAR Comprehensive Study on Riverine Inputs and Direct Discharges (RID) collects and compiles information on waterborne loads of nutrients and trace metals reaching the OSPAR maritime area. The detailed assessment of their trends over time and space covers the data collected for the years 1990 to 2006 and are set out in chapter 4 (Part II: Regional approach) and chapter 5 (Part III: Catchment approach). The objectives of the work were to identify and assess any trends in riverine inputs and direct discharges of the six determinants referred to above to the OSPAR Regions I – IV, and in 10 important catchments within the OSPAR maritime area (see Figure 1.1 and 3.1). Region V has not been included in these assessments as there are very few inputs by rivers and direct discharges in the Wider Atlantic Region. Also the monitoring activities are rather limited. Direct discharges were not included in the catchment assessment.



Figure 3.1: The ten river catchments that are part of the Catchment assessment

Both the riverine inputs and the direct discharges to the maritime area, to waters downstream of the riverine monitoring points, have been assessed. Annual riverine input and direct discharges data and the input-data at sub-regional national level for the regional assessment were screened for incompleteness, inconsistencies and anomalies. In this process, a number of data-series were discarded based upon the 'exclusion rules' defined above.

The focus in this chapter is to outline the main results both at regional and single catchment level. When studying the results one should keep in mind that the OSPAR maritime area covers a considerable geographical area. Variations in climate, geographical location, population density, land-use pattern and implementation of measures are all 'external' elements that will influence concentrations in rivers and effluents.

With reference to the above mentioned, it is no surprise to see that there are considerable differences in concentrations in rivers in Europe, as exemplified by the wide span in average total nitrogen and phosphorus concentrations shown in table 3.1 (average over the monitoring period reported for the various rivers).

River	tot-N (mg/l)	tot-P (mg/l)
Ölfusá (IS)	0.07	0.012
Alta (NO)	0.19	0.013
Glomma (NO)	0.60	0.021
Göta Älv (SE)	0.85	0.020
Elbe (DE)	4.97	0.241
Rhine (at Maassluis, NL)	3.12	0.251
Rhine/Meuse (at Haringvlietsluis, NL)	3.13	0.169
Rhine (at IJsselmeer, NL)	1.88	0.145
Seine (FR)	7.33	0.559
Loire (FR)	4.50	0.228
Garonne (FR)	-	0.139

Table 3.1: Average over the total monitoring period of total nitrogen and phosphorus concentrations in the rivers used in the Catchment approach

In addition to the elements mentioned above, it is necessary to include factors linked to the monitoring exercises. A number of quality assurance issues needed to be considered, such as:

- data quality as such (*e.g.*, statistical representativeness and accuracy);
- harmonisation and transparency in procedures, principles applied;
- methods used for sampling and analyses, uncertainty, and
- comparability of results, procedures and tools.

3.2 Overall results

In the following tables and figures whenever both region and catchment results are presented together, only riverine inputs have been taken into account, as in most cases where catchment data were submitted, they did not include direct discharges data. In general, for the direct discharges determinants (especially nutrients) were less well reported than those in riverine inputs. There were significant gaps in the reporting datasets of trace metals (especially for mercury) for both riverine inputs and direct discharges. There appeared large variations in the way that direct discharges were accounted for in the Contracting Parties' annual RID data reports. Some Contracting Parties included direct discharges at all. Given that a number of the larger cities are located along the shores of the marine coastline of basically all relevant OSPAR countries, the reported discharges may have been seriously underestimated. In general the assessment shows that direct discharges appear of relatively less importance than riverine inputs. This may be reasonable for areas where large cities are located upstream of major rivers, but the issue of whether discharges from coastal urban areas are sufficiently accounted for should be raised.

Inputs to the different regions were compared and the levels of change show the statistically estimated change in percent from the beginning to the end of each of the time periods considered. In most cases the assessed period is 1990 - 2006, but because of lack of data some *datasets* cover shorter time

periods. The levels of change were estimated according to the LOESS or Theil-slope (see glossary and JAMP Guidance (OSPAR agreement 2003-9).

3.2.1 Representativeness of catchment data for the regional analyses

The rivers included in the catchment analyses contribute in varying degrees to the total inputs to their specific region. For some rivers the catchment represents a substantial parts of the total region drainage area, as *e.g.* in case of the river Rhine (see also table 3.2).

Table 3.2: Overview of drainage area for each	OSPAR region and the rivers included in the catchment
approach	

Region	Total drainage area (km²)	Rivers included in the catchment approach	Percentage river catchment to the total drainage area	Comments
Region I	187 000	Ölfusá, Alta	Minor	
Region II	910 000	Rhine, Meuse, Elbe, Glomma, Göta Älv, Seine	50%	Rhine, Meuse and Elbe account for 40% of the drainage area.
Region III	102 000	None	NA	No information on single catchments submitted
Region IV	655 000	Loire, Garonne	About 30%	

In terms of geographic coverage the Greater North Sea (Region II), especially the main body of the North Sea, is the maritime area that is most completely covered in terms of complete time-series and monitored rivers, although even there gaps do exist. This Region also includes some of the most important contributors of heavy metals and nutrient inputs to the OSPAR maritime area, such as the rivers Rhine and Elbe. For these reasons the comparisons made in the following sections use Region II data as example. It is important to realise that when total regional inputs of one particular parameter are shown together with inputs from one or several rivers, the inter-annual variation patterns may or may not be similar. Hence no firm conclusions can be drawn from such comparisons for, *inter alia*, the following reasons:

- There are considerably different practices amongst Contracting Parties in sampling strategies (including sampling frequency, distance from river mouth and location in the river), in methods to define pollutant losses (by direct monitoring, estimation and/or modelling), whether and how direct discharges are accounted for and the yes/no inclusion of land areas/sources. Over time, sampling locations and coverage of monitored areas as well as sampling procedures and collection of hydrological data may have changed;
- Differences laboratories, even within a country, may have been involved in the analyses, employing different analytical methods and calculation practices for the estimations of discharges and inputs. Sweden, for example, has confirmed that there was a change in laboratory methods in mid 1990s for many parameters (see comments in Tables 3.3 and onwards, linked to the Göta Älv).
- Limits of detection (LOD) may vary considerably between laboratories and/or countries. Over time improvements in the analytical processes will usually have resulted in lower scatter of results and in lower LODs.

- Amongst Contracting Parties there may have been varying completeness of reporting, *e.g.* more or fewer rivers reported from one year to another.
- 57. The results of the comparisons must be seen against this background.

3.2.2 Water Discharge

Figure 3.2 shows the average daily water discharges to OSPAR Region II and the rivers Rhine, Meuse and Elbe. No visual upward or downward trend in the total water discharge can be noted. However, a step-trend (1990 – 1995; 1996 – 2002 and 2003 – 2006) can be noted in both the total water discharges to Region II and the total from the three largest rivers.



Figure 3.2: Average daily water discharge to Region II, and the contribution by the rivers Rhine, Meuse and Elbe

3.3 Riverine inputs

3.3.1 Riverine inputs of cadmium

Region I:	The significant downward trend of 43% is highly affected by decreased LOD over time and many observations below LOD. This implies that no firm conclusions can be drawn as the trend might be spurious and may not necessarily show 'real' changes in inputs.
Region II:	The significant downward trend of 20% is highly affected by changed LOD over time, many observations are below LOD and concentrations peak in the beginning of the time series; the 40% decline in riverine inputs in the Elbe River is not affected by LOD issues.
Region III:	A significant downward trend of approximately 60% was detected.
Region IV:	The significant increase of 570% is largely due to an increase in sampling points, incomplete datasets (in the earlier years), and LOD issues (<i>e.g.</i> Loire).



Figure 3.3: Total (and total flow adjusted) annual riverine inputs of cadmium to Regions I – IV

Figure 3.3 shows the total (and total flow adjusted) annual riverine inputs of cadmium to the Regions I - IV for the period 1990 - 2006. Table 3.3 summarises the trend analyses results for cadmium. Given the uncertainties, introduced by changed LODs over time, many data below LOD and some unexpected variability and drops in concentrations in the beginning of the series for some rivers, it is clear that the detected trends should be treated with great caution. In fact, the overall statistically

significant downward trends observed for Regions I and II are all affected by substantial uncertainty at the single catchment level.

The significant increases for Region IV are directly attributed to change in the monitoring strategies and an increase in monitoring activities in this Region. In the first decade of the monitoring period, sampling locations were rather scarce. Since riverine and direct inputs are expressed as total loads of pollutants (rather than loads/flow), an increase in sampling points by 2003 directly led to an increase in reported loads. Similar increases in loads – for the same reason – have been observed for the metals Cu and Pb. This exercise also illustrates the great value of undertaking analyses at catchment level.

Table 3.3: Riverine cadmium trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) are marked in grey

River	Trend sign	Level of change (%)	p-value	Comment
Total Region I (NO)	\downarrow	-43	0.001	
Selected rivers:				
Ölfusá (IS)	\downarrow	-62	0.37	1997–2006
Alta (NO)	\downarrow	-136	0.00001	Trend highly affected by LOD; decrease in LOD over time, many observations below LOD
FR, NL, NO, SE, UK)	\downarrow	-20	0.02	
Selected rivers:				
Seine (FR)	Ļ	-106	0.30	Trend highly affected by LOD; variable LOD in time (0.1–10 μ g/l), many observations below LOD; sudden drop in concentrations after 1993
Elbe (DE)	Ļ	-42	0.03	1992–2006. High concentrations in 1992–1993; no data below the LOD
Rhine (at Maassluis, NL)	ſ	+128	0.44	1990–2005. Trend affected by high LODs since 1995; many observations below LOD
Rhine (at IJsselmeer, NL)	↓	-55	0.008	1990–2005. Trend affected by high peaks and variability in beginning of time series, high LODs since 1995; many observations below LOD
Rhine/Meuse (at Haringvlietsluis, NL)	\downarrow	-3	0.96	1990–2005
Glomma (NO)	Ļ	-87	0.02	Trend highly affected by LOD; decrease in LOD over time and many observations below LOD
Göta Älv (SE)	\downarrow	-6	0.63	Change in laboratory method mid-1990ies
Total Region III (IE, UK)	\downarrow	-63	0.0002	
Γ				
Total Region IV (ES, PT)	ſ	+570	0.08	
Selected rivers:				
Garonne (FR)	-	-	-	Low sampling frequency (2-4 samples/yr) combined with missing data certain years.
Loire (FR)	1	+444	0.01	Trend highly affected by LOD; since 2000 all measurements were below LOD, mainly be- cause of large fluctuations in LOD (0.1–10 μg/l)

3.3.2 Riverine input of Copper

Region I:	A downward significant trend of approximately 45%, which was also supported by a reduction in inputs from the Alta River.
Region II:	A downward significant trend of 40%, which is supported by corresponding significant downward trends in the rivers Seine, Meuse and Rhine. High inputs, variability and concentration peaks in the beginning of the 1990s explain to a large degree the detected trends.
Region III:	A downward significant trend of approximately 30% was detected.
Region IV:	No firm conclusions can be drawn due to incomplete datasets.



Figure 3.4: Total annual riverine inputs of copper to OSPAR Regions I – IV

Figure 3.4 shows the total (and total flow adjusted) annual riverine inputs of copper to the Regions I - IV for the period 1990 - 2006. Table 3.4 summarises the trend analyses results for copper.

Table 3.4: Riverine copper trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) are marked in grey

River	Trend sign	Level of change (%)	p-value	Comment
Total Region I (NO)	\downarrow	-44	0.006	
Selected rivers:				
Ölfusá (IS)	↓	-24	0.24	1997–2006
Alta (NO)	Ļ	-87	0.001	

Total Region II (BE, DE, DK, FR, NL, NO, SE, UK)	Ļ	-40	0.00005	
Selected rivers:				
Seine (FR)	¢	+50	0.003	Trend is explained by low inputs in 1990 and 1991; gaps (less frequent) in data for 1993–1997
Elbe (DE)	↓	-13	0.22	1992–2006; high inputs in 1992, first year
Rhine (at Maassluis, NL)	Ļ	-27	0.01	1990–2005
Rhine (at IJsselmeer, NL)	Ļ	-38	0.01	1990–2005; high peaks and variability 1990– 1995
Rhine/Meuse (at Haringvlietsluis, NL)	Ļ	-30	0.10	1990–2005
Glomma (NO)	↓	-10	0.53	
Göta Älv (SE)	¢	+53	0.001	Trend explained by change in laboratory in mid 1990s

Total Region III (IE, UK) ↓

Total Region IV (ES, PT)	↑	+346	0.11	
Selected rivers:				
Garonne (FR)	-	-	-	Low sampling frequency (2-4 samples/yr) combined with missing data in certain years
Loire (FR)	-	-	-	Low sampling frequency (2-4 samples/yr) combined with missing data in certain years.

0.04

-30

3.3.3 Riverine input of lead

Region I: A downward significant trend of 85%, which is highly affected by decreased LOD over time and many observations below LOD. This implies that no firm conclusions can be drawn.

- Region II: Downward significant trend of approximately 50% is highly affected by changed LOD over time, many observations below LOD and concentration peaks in the beginning of the time series; the 90% decline in riverine inputs in the Seine River is, however, not affected by the LOD.
- Region III: No significant trend.

Region IV: No firm conclusions can be drawn due to incomplete *datasets*.



Figure 3.5: Total (and total flow adjusted) annual riverine inputs of lead to Regions I – IV

Figure 3.5 below shows the total (and total flow adjusted) annual riverine inputs of lead to the Regions I - IV for the period 1990 – 2006. Table 3.5 summarises the trend analyses results for lead.

Table 3.5: Riverine lead trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) marked in grey

River	Trend sign	Level of change (%)	p- value	Comment
Total Region I (NO)	\downarrow	-85	0.0006	
Selected rivers:				
Ölfusá (IS)	\downarrow	-4	1	1997–2006; 25% of data below LOD
Alta (NO)	↓	-109	0.02	Trend highly affected by LOD; decrease in LOD over time and many observations below LOD

Total Region II (BE, DE, DK, FR, NL, NO, SE, UK)	Ļ	-52	0.0005	
Selected rivers:				
Seine (FR)	\downarrow	-91	0.006	No important impact of data below LOD.
Elbe (DE)	↓	-15	0.19	1992–2006; sudden drop in concentrations after 1994, but no data below the LOD
Rhine (at Maassluis, NL)	Ļ	-77	0.007	1990–2005; trend affected by variable LOD over time (0.1–0.9 μ g/l) and many values below LOD
Rhine (at IJsselmeer, NL)	Ļ	-59	0.01	1990–2005; some minor LOD-problems
Rhine/Meuse (at Haringvlietsluis, NL)	↓	-54	0.14	1990–2005
Glomma (NO)	↓	-44	0.08	Trend highly affected by LOD; decrease in LOD over time and many observations below LOD
Göta Älv (SE)	↑	+181	0.001	Trend explained by changed laboratory mid 1995
Total Region III (IE, UK)	\downarrow	-22	0.20	
Total Region IV (ES, PT)	↑	+318	0.48	
Selected rivers:				
Garonne (FR)	-	-	-	Low sampling frequency (2-4 samples/yr) combined with missing data in certain years
Loire (FR)	-	-	-	Low sampling frequency (2-4 samples/yr) combined with missing data in certain years.

3.3.4 Riverine input of mercury

Region I:	The downward significant upward trend of 160% is highly affected by changed LOD over time, change in chemical laboratory in 1999–2003 with substantially higher concentrations and many observations below LOD. This implies that no firm conclusions can be drawn as the trends are spurious and do not show 'real' changes in inputs.
Region II:	The highly significant downward trend of 75% is partly affected by changed LOD over time, many observations below LOD and concentration peaks in the beginning of the time series; the about 70% decline in riverine inputs in Rhine and Rhine/Meuse is, however, not affected by LOD issues
Region III:	A downward significant trend of approximately 85% was detected
Region IV:	Incomplete datasets



Figure 3.6: Total (and total flow adjusted) annual riverine inputs of mercury to Regions I-III

Figure 3.6 below shows the total (and total flow adjusted) annual riverine inputs of mercury to the Regions I – III for the period 1990 – 2006. Hg data for Region IV were incomplete and have not been included. Table 3.6 summarises the trend analyses results for mercury.

Table 3.6: Riverine mercury trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) are marked in grey

River	Trend sign	Level of change (%)	p-value	Comment
Total Region I (NO)	Î	+160	0.04	
Selected rivers:				
Ölfusá (IS)	\downarrow	-10	0.15	1997–2006; 74 out of 79 values below LOD
Alta (NO)	Ţ	+116	0.30	Trend highly affected by data below LOD, changed LOD over time and changed laboratory 1999–2003; about 50% of data below LOD

Total Region II (BE, DE, DK, FR, NL, NO, SE, UK)	Ļ	-75	0.0005	
Selected rivers:				
Seine (FR)	1	-149	0.20	High LODs in the first years, variable LOD over time (0.1-10 μg/l); since 2000 all measurements below the LOD; large gaps in series and differences in sampling frequency; sudden drop in concentrations after 1994
Elbe (DE)	Ļ	-69	0.00001	1992–2006; there was no data below the LOD, but high peaks and variability in concentrations in 1990–1995 explain to a large degree the downward trend
Rhine (at Maassluis, NL)	Ļ	-87	0.00001	1990–2005; many observations (220 out of 335) below LOD and variable LOD over time (0.01- 0.001 μg/l)
Rhine	\downarrow	-68	0.003	1990–2005; peak concentrations in 1990–1994
(at IJsselmeer, NL)				
Rhine/Meuse (at Haringvlietsluis, NL)	Ļ	-71	0.005	1990–2005
Glomma (NO)	Ţ	+293	0.27	Trend highly affected by data below LOD, changed LOD over time and changed laboratory 1999–2003; about 50% of data below LOD
Göta Älv (SE)	\downarrow	-58	0.00001	1995–2006; peak values in 1995–1998
Total Region III (IE, UK)	Ļ	-84	0.00001	

Total Region III (IE, UK)

0.00001

Total Region IV (ES, PT)	-	-	-	Incomplete data
Selected rivers:				
Garonne (FR)	-	-	-	No data available
Loire (FR)	Ļ	-71	0.048	A considerable part of the data below LOD, near- ly constant LOD for the whole period $(0.1 \ \mu g/l)$

3.3.5 Riverine inputs of nitrogen

Region I:	The downward statistically significant trend of 50% (Norwegian data only) is explained by a highly uncertain with regard to the decline in tributary rivers due to low sampling frequency (1-4 samples per annum) and can thus be spurious.
Region II:	The downward statistically significant trend of 20% is supported by highly significant trends in the rivers Elbe (-45%), Meuse (-27%), Rhine/Meuse (-39%) and Rhine (-27%).
Region III:	No statistically significant trend detected

Region IV: No statistically significant trend detected. NB! Data for 1997-2006 only

Figure 3.7 below shows the total (and total flow adjusted) annual riverine inputs of nitrogen to the Regions I - IV for the period 1990 - 2006. Table 3.7 summarises the trend analyses results for nitrogen.



Figure 3.7: Total (and total flow adjusted) annual riverine inputs of nitrogen (tot-N) to the Regions I – IV

Table 3.7: Riverine total nitrogen trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) are marked in grey

River	Trend sign	Level of change (%)	p-value	Comment
Total Region I (NO)	Ļ	-51	0.005	
Selected rivers:				
Ölfusá (IS)	Ť	+93	0.02	1997–2006
Alta (NO)	\downarrow	-22	0.09	

Total Region II (BE, DE, DK, FR, NL, NO, SE, UK)	Ļ	-21	0.0004	
Selected rivers:				
Seine (FR)	\downarrow	-10	0.31	_
Elbe (DE)	\downarrow	-45	0.00001	1992–2006
Rhine (at Maassluis, NL)	Ļ	-27	0.007	1990–2005
Rhine (at IJsselmeer, NL)	Ļ	-39	0.02	1990–2005; 'lake-like' seasonal pattern with zero concentrations during summer
Rhine/Meuse (at Haringvlietsluis, NL)	Ļ	-27	0.003	1990–2005
Glomma (NO)	1	+21	0.08	
Göta Älv (SE)	\downarrow	-4	0.41	
Total Region III (IE, UK)	\downarrow	-12	0.28	
Total Region IV (ES, PT)	\downarrow	-14	0.86	1997–2006
Selected rivers:				
Garonne (FR)	-	-	-	
Loire (FR)	\downarrow	-25	0.11	Trend might be affected by data below the LOD



Figure 3.8: Annual riverine inputs of nitrogen (tot-N) to Region II and the total inputs from the rivers Rhine, Meuse and Elbe

3.3.6 Riverine input of phosphorus

- Region I: The downward significant trend of approximately 55% (Norwegian data only) is explained by a high uncertainty with regard to the decline in tributary rivers due to a low sampling frequency (1-4 samples/yr), and can thus be spurious.
- Region II: The significant downward trend of approximately 50% is supported by highly significant trends in the rivers Seine (-83%), Elbe (-43%), Meuse (-46%), and the tendencies although not statistically significant in Rhine/Meuse and Rhine.
- Region III: No significant trend detected.
- Region IV: No significant trend detected.



Figure 3.9: Total (and total flow adjusted) annual riverine inputs of phosphorus (tot-P) to the Regions I - IV

Figure 3.9 shows the total (and total flow adjusted) annual riverine inputs of phosphorus (tot-P) to the Regions I - IV for the period 1990 - 2006. Table 3.8 summarises the trend analyses results for phosphorus.

Table 3.8: Riverine total phosphorus (tot-P) trend analyses summary results for catchments and regions. Statistically significant trends (p < 0.05) are marked in grey

River	Trend sign	Level of change (%)	p-value	Comment
Total Region I (NO)	\downarrow	-56	0.01	
Selected rivers:				
Ölfusá (IS)	Ť	+8	0.72	1997–2006
Alta (NO)	\downarrow	-41	0.58	

Total Region II (BE, DE, DK,	↓	-53	0.0002	
FR, NL, NO, SE, UK)				
Selected rivers:				
Seine (FR)	\downarrow	-83	0.00001	
Elbe (DE)	\downarrow	-43	0.00001	1992–2006
Rhine	↓	-46	0.02	1990–2005
(at Maassluis, NL)				
Rhine (at IJsselmeer, NL)	\downarrow	-23	0.23	1990–2005
Rhine/Meuse (at Haringvlietsluis, NL)	\downarrow	-35	0.14	1990–2005
Glomma (NO)	Ļ	-20	0.49	
Göta Älv (SE)	¢	+25	0.08	
Total Region III (IE, UK)	\downarrow	-17	0.25	
Total Region IV (ES, PT)	\downarrow	-27	0.30	1997–2006
Selected rivers:				
Garonne (FR)	↓	-59	0.21	
Loire (FR)	Ļ	-74	0.01	



Figure 3.10: Annual riverine inputs of total phosphorus to Region II and the sum of the rivers Rhine, Meuse and Elbe

3.4 Direct Discharges

Figures 3.11 to 3.16 show the annual direct discharges of the six parameters to the Regions I – III. For Region IV the data on the direct discharges was insufficient and thus was not taken into account.

Region I:

- Due to lack of data from Iceland for the period 1990 1996 only Norwegian data was considered;
- Compared to riverine inputs, the direct discharges of *cadmium* were low throughout the whole period (Figure 3.11) with a decreasing trend over the period 1990 2006. A significant reduction in the range of 70% (p <0.035) was detected;
- Direct discharges contribute to about 5 20% of total inputs of *copper* to Arctic Waters in years 1990 2004. Fish farming is the most important direct source of copper (Borgvang *et al.*, 2007), as a result of losses of Cu used in anti-fouling treatment of net cages. Such losses were only reported for 2005 and 2006;
- There is no significant long-term trend in direct discharges of *lead* to Region I (p <0.187);
- The sudden shift in level of direct discharges of total *nitrogen* and *phosphorus* in the year 2000 and the detected statistically significant upward trend is due to the fact that Norway in this year started to report on the N and P losses from aquaculture.

Region II:

Direct discharges for all substances, except for phosphorus, account for only a minor fraction
of the total inputs. Significant downward trends were noted for all the trace metal discharges
and nutrients (75% for Cd; 70% for Cu, 80% for Pb and 70% for Hg), nitrogen (35%) and
phosphorus (30%). It shall be remarked that reported direct discharge data was scarce as
compared to riverine input data. Thus the trend estimates should be treated with great caution.

It should be noted that the trace metal discharges from Denmark and France were not included in the analyses due to incomplete reporting. In addition, many countries have in their annual reports reported uncertainties in especially the first years of the 1990s.

 For example, in the Netherlands there was a different monitoring system in the years 1990 – 1992 as compared to the following years. The nature of these changes was not reported, however. Similarly there was no indication on how the changes may have affected the magnitude in discharges and the subsequent trend results.

Region III:

- There has been a strong significant decrease in direct discharges of all the heavy metals (except lead) for the time period concerned, in particular since 2002–2003, notably -95% for Cd and Hg, and -90% for Cu;
- Lead direct inputs have decreased by 45% over the period, but the decrease was not statistically significant;
- Highly significant decreasing trends of 30% for nitrogen and 40% for phosphorus were detected for the direct discharges.


Figure 3.11: Annual direct discharges of cadmium to the Regions I – III



Figure 3.12: Annual direct discharges of copper to the Regions I – III



Figure 3.13: Annual direct discharges of lead to the Regions I – III



Figure 3.14: Annual direct discharges of mercury to the Regions I – III



Figure 3.15: Annual direct discharges of total nitrogen to the Regions I – III



Figure 3.16. Annual direct discharges of total phosphorus to the Regions I - III

3.5 Limitations in OSPAR-RID trend analyses assessments

There is no doubt that management decisions have become more complex over the most recent years. This is due to more integrated and cross-sectorial approaches such as required by the implementation of the EC Water Framework Directive. This consequently requires water quality monitoring programmes that provide data for multiple purposes, including trend analyses to detect improvement or deterioration in water quality with time. One particular challenge in trend analyses is to separate the human-induced (anthropogenic) variability and trends from the natural variability (hydrometeorology). Even if this theoretically can be included in formal statistical trend analysis methods, the causal and underlying mechanisms for water quality changes still remain as a particular challenge.

In order to have a sound and decent chance to both separate the anthropogenic and natural variability, and interpret the causes for trends it requires that the input data is 'optimal' both from a statistical point of view (random sampling, independence between single observations, few outliers, and consistent variability over the years) and 'narrative' interpretation point of view.

One particular and obvious problem encountered in this trend assessment is that the data between the years in a long-term trend perspective (*i.e.* 1990 - 2006) in some cases are not fully comparable. One specific challenge is the technological development between 1990 and 2006 with *e.g.* improved chemical laboratory analytical techniques, which results in the fact that in recent years concentrations at low levels can be detected with a considerably higher accuracy and precision than in the early 1990s. This has usually also resulted in a decrease of the limit of detection. An illustration of changing LODs is given in Table 3.9 for the Norwegian LODs for lead for the period 1990 – 2007.

Year	LOD Pb
1990	0.5
1991	0.1
1992-1998	0.02
1999	0.01 (0.1) ¹
2000	0.01
2001	0.01-0.02 (0.1) ¹
2002-2003	0.02-0.05 (0.2) ¹
2004-2007	0.005

Table 3.9: Changes in detection limits (LOD) for lead (µg/l) during the period 1990-2007

¹ The values in parenthesis are probably due to errors, as the detection limits (LOD) may have been given in wrong units

To illustrate the effect of decreasing LODs on the assessment a theoretical example could be as follows. Say that we in the 1990 had a LOD of 100 μ g/l, whereas we in 2006 are able to determine concentrations down to an LOD of 10 μ g/l. Suppose further that we in the early 1990 have 100% of the results (annual concentrations) below LOD. In this situation any concentration between 0 and 100 μ g/l is in fact possible. So, in 2006 with the lower detection limits, if we may observe 'true' analytical results between 11 and 99 μ g/l, it is evident that both upward and downwards trends are likely to be detected statistically. This is shown in several examples for single rivers and especially for the heavy metals Cd, Pb and Hg, as is shown for the different countries in Table 3.10.

In support of the role of the LOD 'problem' in the trend analyses, Annex II provides the LODs reported by the Contracting Parties (sometimes distinguishing several periods).

Another 'trend-dilemma' is that incomplete time-series (*e.g.*, data with entirely missing observations in single years etc.) seriously limit the statistical definition of trends, if any.

In fact, it was surprising to note in the 'catchment' analyses that many of the included river cases (10 rivers and 11 monitoring sites) had incomplete *datasets*, especially for the trace metals. In that respect one should realise that all catchments included are 'large' and account for *e.g.* Region II for a substantial proportions of the total riverine inputs.

Cd (µg/l)					
	Recommended	Minimum	Maximum	Difference	Region
BE	0.01	0.06	5	4.94	Ш
DE	0.01	0.01	0.05	0.04	Ш
DK	0.01	0.004	0.005	0.001	=
ES	0.01	0.001	20	19.999	IV
FR ¹)	0.01	0.5	2	1.5	=
FR	0.01	0.005	10	9.995	IV
IE	0.01	0.05	0.1	0.05	Ш
IS	0.01	NI	NI	NI	I
NL	0.01	0.01	0.2	0.19	Ш
NO	0.01	0.001	0.1	0.099	I, II
PT	0.01	0.01	0.03	0.02	IV
SE	0.01	0.003	0.003	0	II
UK	0.01	0.02	0.02	0	II, III

Table 3.10: Limits of detection (LODs) achieved for cadmium (in µg/l)

NI = No information

¹) For the year 2006

3.6 Recommendations

It is considered important that Contracting Parties have access to detailed information about detection limits and analytical laboratory methods. Changes in laboratories and detection limits over time should be duly registered – preferably in a common database, since there were examples in this assessment that this could influence the trends significantly.

The RID Principles state that it is necessary to choose an analytical method, which gives at least 70 % of positive findings (*i.e.* no more than 30% of the samples below the detection limit). This is an issue for countries with relatively 'clean rivers' despite LOD values that correspond to the OSPAR standards; *e.g.*, in Region I, and should be considered when reviewing the RID Principles.

In this assessment, it was noted that detection limits have changed over time, vary considerable between countries and laboratories and are, in some cases, far above the OSPAR recommended values. The detection limit should be at least as low as the limits adopted by OSPAR in 2005. The use of different detection limits is clearly a considerable problem both within countries and for comparison of result between countries and should be considered in future reporting.

Optimally the same accredited laboratory should be used for all rivers within a country. If this is not possible, intercomparison of results should take place. In cases of changes of laboratories, over time an intercomparison should be carried out during a certain 'transfer of responsibility period'. This is most likely a problem in some countries both in terms of using several laboratories and in cases where laboratories have changed in the period 1990 to date.

Historical QA, *i.e.* monitoring results checked against historical data should be undertaken by qualified researchers with experience in assessing water quality data. This should be done as soon as possible after analysis so whenever anomalies are found, the samples can be re-analysed.

Given differences in and quality of reporting between countries and over years (*e.g.*, due to reason mentioned above) trustworthy trend analyses may be difficult to carry out for aggregated data such as for the four Regions in this assessment. Attempts of further harmonisation of data between countries and historical reconstruction should be pursued.

The implementation of the various trend assessments in this study and also in the 2005 trend assessment (for period 1990-2002), highlights the importance of ensuring that *datasets* for inputs and flows are as complete and consistent as possible before applying the statistical tools. Ideally, all significant anomalies should be addressed and Contracting Parties should be urged to complete their *datasets* prior to the assessment

There is, for some *datasets*, an indication of under-reporting in the earlier years of the RID programme, as well as a higher degree of uncertainty in the reported values in the beginning of the time series. This is a factor which could influence the outcome of any assessments by *e.g.*, reducing any downward trends that are identified or increasing any upward trends. This is a matter that should be considered more fully before any further trend analyses are carried out. One approach would be to reduce the period of assessment to include only those years for which data are more consistent.

Recognising that inputs and direct discharge *datasets* are not so well developed for Region IV as they are for other Regions, one should consider how best to progressively develop and present the data that is available for this Region, and seek to undertake assessments when sufficient time series of consistent data become available, either at local or regional scale.

Catchment specific input data improve the reliability of Region assessments to a large degree. For future assessments it is recommended that countries also report to the extent possible input data catchment by catchment.

4. Part II – Regional Assessment

Part II describes the results of assessment of Riverine Inputs and Direct Discharges to the OSPAR Regions I – IV for the period 1990 – 2006, based on the Comprehensive Study on Riverine Inputs and Direct Discharges. Two types of assessment have been carried out, namely on the riverine inputs and the direct discharges to these four Regions of the maritime area from below the monitoring points. The determinants assessed were cadmium (Cd), copper (Cu), lead (Pb), mercury (Hg), nitrogen (tot-N) and phosphorus (tot-P). Water discharge (Q) was also included in the assessment though it was not statistically tested for trends. The detailed statistical trend analysis is available electronically as **Technical Supplement 1.**

Riverine Inputs and Direct Discharges to OSPAR Region I (Arctic Waters)

Figure 4.1: Map showing the OSPAR maritime area with the Region I (Arctic Waters) highlighted in dark blue

4.1.1 **Regional coverage**

4.1.

The assessment covers Region I of the OSPAR maritime area. This is a region with dense population, few industries and little agricultural activity. Only the contributions of Norway to this region through the Barents and the Norwegian Seas were taken into account as there are no available Icelandic data for the period 1990 to 1996.

Table 4.1 shows the geographic coverage of the reported inputs to Region I, whereas Figure 4.2 indicates the sampling sites of the rivers included in the Norwegian river monitoring programme.



Table 4.1: Geographic coverage (km²) of reported inputs to Region I

Figure 4.2: River sampling sites in the Norwegian RID programme. Red dots represent the 10 main rivers (sampled monthly). Yellow dots represent the 36 tributary rivers (sampled quarterly). Numbers next to the dots refer to the national river register (REGINE; www.nve.no). Source: Skarbøvik et al. (2008)

4.1.2 Data check prior to assessment

The annual inputs data were firstly screened for incompleteness, inconsistencies and anomalies. In this process, it was noted that the Norwegian direct discharges of Cd, Hg and Pb to the Barents Sea were reported as zero in the years 2004-2006, but also the other years the direct discharges have a marginal impact on the total inputs given that direct discharges account for a relative low fraction in relation to the riverine inputs of these substances.

4.1.3 Results and discussion

River water discharge (Q)

The total river water discharge (main and tributary rivers in Norway) to Region I showed a rather low inter-annual variation and no obvious trend during the period 1990-2006, see Figure 4.3.



Figure 4.3: Total reported average daily river water discharge to Region I for the period 1990 – 2006

Cadmium (Cd)

Between 1990 and 2006, the total riverine input of cadmium (Figure 4.4.) shows a highly significant monotonic downward trend (p <0.002). The reduction is about 85% without LOD-correction and around 45% with LOD-reduction, with a decrease from over 6 t/yr to around 3 t/yr in the beginning of the 1990s, and further down to less than 1 t/yr from the year 1998 onwards. Exception was the year 2004 with an input of 1.3 t/yr.

The relatively high inputs in 2004 are most likely due to the fact that there was a change in the Norwegian river monitoring strategy in that year (see next section for details). The reason for the general downward trend is most likely due to changes in LOD over the years, rather than real changes in inputs. For example, in 1990 the LOD was $0.1 \mu g/l$, whereas in the period 1992-1998 it was $0.01 \mu g/l$; with variable LODs in the range of $0.001 - 0.02 \mu g/l$ in the period 1999-2003. Known improvement in sewage treatment and reduced discharges from industry in Norway could certainly also influence the decline in cadmium inputs. However, it is difficult to compare the influence of reduced discharges to the reduced inputs with the importance of changes in LOD over the time period 1990 – 2006. Annex II shows the most recent reported LOD for cadmium for this Region.

Compared to riverine inputs, the direct discharges of Cd were low throughout the whole period (Figure 4.4), with a decreasing trend over the years 1990 – 2006. A significant (p < 0.035) reduction in the range of 70% was also detected. The reason for the downward trend is most likely due to reduced discharges, in combination with improved effluent treatment from industrial plants in Norway. It is important to note, however, that there is no reporting on cadmium discharges from municipal treatment plants in the period covered by this assessment.

The total input of Cd is strongly dominated by riverine inputs.



Figure 4.4: Annual riverine inputs and direct discharges of cadmium (Cd) to OSPAR Region I

Copper (Cu)

Annual riverine inputs of Cu to Region I in the period 1990 - 2006 varied between 66 and 233 t/yr (Figure 4.5), with a highly significant monotonic downward trend (p <0.006). The decline is in the range of 40% (LOESS analysis), with a decrease of around 100 t/yr in the early/mid 1990s; a further reduction to about 60 – 100 t/yr was observed from 1998 onwards. There was a peculiar pattern after 1998 with large inter-annual variability. The period 2000 – 2003 was characterised by low inputs in these four consecutive years, but increased inputs from 2004 onwards were reported (Figure 4.5). This is most likely due to a change in the Norwegian river monitoring strategy in 2004. More specifically, in the years 1990 – 2003, 155 tributary rivers were monitored only once per year while from 2004 onwards the strategy was changed to the quarterly sampling of 36 rivers (Borgvang *et al.*, 2006). This change led to higher estimated riverine inputs since it increased the probability of capturing concentration peaks during high water discharges and flood events. It should in this connection be noted that the tributary rivers in Norway account for a substantial fraction of the total riverine inputs (see example in Table 4.2 below). As a consequence, the decrease detected should be treated with great caution. Annex II shows the most recent reported LOD for copper for Region I.

Table 4.2: Riverine inputs of Cu from Norway to Region I in 2006 (in t/yr). Source: Skarbøvik et a	Ι.
(2007)	

	Barents Sea	Norwegian Sea	Total Region I
Main rivers (3)	1.6	16.3	17.9
Tributary rivers (13)	35.8	42.9	78.7
Total	37.4	59.2	96.6



Figure 4.5: Annual riverine inputs and direct discharges of copper (Cu) to Region I

Direct discharges contributed about 5 - 20% of total inputs of Cu to Region I in years 1990 - 2004. Fish farming was the most important direct source for copper which is attributed to losses of Cu used in anti-fouling treatment of net cages (Borgvang *et al.*, 2007). Such losses were reported only in 2005 and 2006, which explains the strong increase observed for these years. Since 2005, the direct discharges of Cu are about 50% higher than the riverine inputs.

Lead (Pb)

The LOD for lead (Pb) in the Norwegian RID-programme has lowered by a factor of 100 during the monitoring period (1990 – 2006), see Annex II. At the same time many reported concentrations were at LOD level or slightly above. As a result no reliable trend assessment of the annual riverine inputs of lead could be made. Nonetheless, bearing these uncertainties in mind, annual riverine inputs of Pb ranged from about 50 to 5 t/yr in the period 1990 – 2006, with a highly significant monotonic downward trend (p <0.001). The reduction has been in the range of 85% (estimated from Theil slope) from about 40 t/yr in the years 1990 – 1991, to less than 10 t/yr from 1998 onwards.

Annual direct discharges of Pb varied between 0.6 - 1.7 t throughout the time period, with an average of 1.5 t /yr. This equals about 20 - 30% of the riverine inputs the more recent years. For the years 2000 and 2001 considerably higher direct discharges were reported: 5.3 and 3.7 t/yr respectively (Figure 4.6). There is no significant long-term trend of direct Pb discharges to Region I (p <0.187).

Total inputs of Pb in the reporting period are dominated by riverine inputs and ranged from 6 51 to 6 t/yr, with an average input of 19 t/yr.



Figure 4.6: Annual riverine inputs and direct discharges of lead (Pb) to Region I

Mercury (Hg)

The median annual riverine input of mercury to Region I in the period 1990 – 2006 is 0.22 t, with a range from 0.1 to almost 1 t/yr. The highest inputs were reported for the period 1999 – 2003 (Figure 4.7) and are attributed to a change in laboratory. Hence, trends should be interpreted with caution. Overall, the trend analysis detected a very high significant upward trend ($p < 2.6 \times 10^{-6}$). Annex II shows the most recent reported LOD for mercury for Region I.

The median annual input of mercury by direct discharges was 0.02 t (range 0 to 0.1 t/yr). No direct discharges were reported for the period 2004 - 2006. The trend analyses revealed a highly significant downward monotonic trend (p <0.001). In 1990 and 1991, the direct discharges of mercury contributed to about 40% of the total inputs. Since 1992, direct discharges represent only a small share of the total inputs of mercury to Region I, with a contribution less than 15%.



Figure 4.7: Annual riverine inputs and direct discharges of mercury (Hg) to Region I

Total nitrogen (tot-N)

The annual median value of riverine inputs for the period 1990 – 2006 was 26 kt, ranging from 13 to 32 kt/yr. For the long-term development, a significant monotonic non-linear decreasing trend of about 50% was detected (p <0.004). There is a considerable downward development in the riverine inputs since 2002 (Figure 4.8). Considering the individual inputs from three main rivers (Orkla, Vefsna and Alta Älv, see Figure 4.9), no decreasing trend can be seen in two out of three. Only in the Vefsna river a decreasing trend could be observed, indicating that the inputs from tributary rivers explain the overall downward input trend. The reasons for this are not know, but low sampling frequency and changes monitoring regime since 2004 may have influenced the results.

For the direct discharges the annual inputs range from about 5 to 25 t, with markedly higher inputs from 2000 onwards, leading to a significant monotonic non-linear increasing trend (p < 0.001). The increase is explained by the fact that nitrogen losses from aquaculture were not reported before the year 2000. These inputs account for a substantial portion of the total inputs Table 4.3).

Table 4.3: Riverine inputs and direct discharges of nitrogen (tot-N, in t/yr) in 2006 from Norway to Region I. Source: Skarbøvik et al., 2007

	Norwegian Sea	Barents Sea	Total
Main 3 rivers	1 178	436	1 614
Tributary rivers	9 489	3 801	13 290
Direct inputs			
Sewage effluents	3 202	222	3 424
Industrial effluent	186		186
Fish farming	21 865	1 547	23 412



Figure 4.8: Annual riverine inputs and direct discharges of total nitrogen (tot-N) to OSPAR Region I



Figure 4.9: Annual inputs of total nitrogen (tot-N), flow-normalised inputs and trend-smoother for three main rivers in Norway. Source: Skarbøvik et al. (2008)

Total phosphorus (tot-P)

Figure 4.10 shows the annual riverine inputs and direct discharges of total phosphorus (tot-P) to Region I. For riverine inputs a significant downward trend was detected over the study period 1990 - 2006 (p <0.012), notably the result in the marked lower inputs since 2003. The three main rivers Orkla, Vefsna and Altaelven show a similar pattern. However, these rivers only contribute to about 10% of the total riverine inputs (Skarbøvik *et al.*, 2007), meaning that the trend pattern is to a large extent explained by the inputs from the tributary rivers.

Nn. For direct discharges of tot-P between 1990 and 1999 the inputs varied between 0.5 and 1.4 kt/yr. There has been a strong increase in inputs in the range of 2.7 to 5.4 kt/yr since the year 2000 (Figure 4.10). As for total nitrogen, this could be attributed to tot-P losses from aquaculture that were reported from 2000 onwards. This also implies that the highly significant upward trend (p < 0.001) is just an artefact and should not be interpreted as a real increase in the discharges.

Aquaculture plays a major role for the total phosphorus inputs to the Region I waters. Skarbøvik *et al.* (2007) reported that in 2006 this source accounted for about 80% of the tot-P inputs.



Figure 4.10: Annual riverine inputs and direct discharges of total phosphorus (tot-P) to Region I



4.2 Riverine Inputs and Direct Discharges to OSPAR Region II (Greater North Sea)

Figure 4.11: Map showing the OSPAR maritime area. OSPAR Region II (Greater North Sea) is highlighted in dark blue

4.2.1 Regional coverage

Region II of the OSPAR maritime area, the Greater North Sea, includes the following sea areas and countries: North Sea (Belgium, The Netherlands, Germany, Denmark, Norway and the UK), Skagerrak (Denmark, Sweden and Norway), Kattegat (Denmark and Sweden) and the Channel (France and UK). The drainage area of the Greater North Sea is about 910 000 km². Two river catchments – of the rivers Rhine and Elbe – account for 35% of the drainage area (Figure 4.12). The UK does not report on identified rivers, but has 39 sampling regions around the UK (in the OSPAR Region II: 24 and in Region III: 15), which incorporate well over 200 rivers. The UK reports the information aggregated as inputs for the 39 sampling regions.





4.2.2 Data check prior to assessment

The annual inputs and direct discharges data were screened for incompleteness, inconsistencies and anomalies. In this process, based upon the 'exclusion rules' listed in Section 2, the following data were excluded:

- Denmark: all the heavy metal input data. The Danish monitoring programme has until recently focused on monitoring of nitrogen and phosphorus compounds, as well as on organic matter. From the late 1990s some heavy metals and hazardous substances have been monitored in few selected rivers and at point sources. For rivers most concentrations have been under the detection limit and no total inputs to coastal waters have been estimated.
- France: all Channel data due to incomplete data series, except those of the river Seine.

The quality of the input data, drainage basin coverage and consistency in reporting was regarded as satisfactory for proper and sound statistical analyses. This is due to rather consistent reporting for the large rivers in the Region (*e.g.*, Elbe, Weser and Rhine) draining catchments with a high population density and heavy industrial and agricultural activities.

The potential uncertainties for proper statistical analyses linked to the different levels of LOD between laboratories and countries, and with changes over time were not possible to quantify, despite attempts to assess these via the annual national reports. There are also gaps in the reporting of single river data between years; they were, in some cases, estimated by statistical relationships (Section 2), but this might have influenced the reliability of the statistical analyses and the interpretation. All this implies that the identification of statistically significant trends should be interpreted and approached with caution since the changes may be due to other reasons than 'real' input changes to the Greater North Sea over time. This is particularly true for the interpretation of and comparisons between single consecutive years. The following should be noted in particular:

- In Germany, data on heavy metal inputs were missing for the river Elbe for the period 1990 1991;
- In the Netherlands, the monitoring system was different for the periods 1990 1992 and 1993 2006. (*i.e.* number of rivers monitored and reported). In addition, in the Wadden Sea basin new sampling sites where introduced in 1997. There was no reporting on discharges from sewage and industrial effluents for the years 1993 and 1994, but the reporting was resumed for the year 1995;
- The direct discharges from the Netherlands showed scarce and incomplete data for the period 1990 1993. In 1994, no direct discharges from the Rhine area were reported;
- For many countries there has been a substantial change in LODs over the monitoring period 1990 2006 for Cd, Hg and, to some extent, for Pb;
- In Sweden, riverine inputs of Hg in 1990 1994 were based on area-specific inputs from other monitored rivers; in addition, there chemical laboratory methods for Cd, Cu and Pb changed in 1995.

In order to provide a better platform for a Regional assessment, the interpretation of trends within Region II to some extent focused on the large rivers (Elbe, Rhine, Seine) and UK regions, accounting for almost 60% of the entire drainage area. Furthermore, the direct discharges were given less attention (except for tot-P) given their relatively low contribution to the total inputs.

4.2.3 Results and discussion

River water discharge (Q)

The river water discharges (Q) for the Region II and for the rivers Rhine and Elbe are presented for the period 1990 – 2006 in Figure 4.13. Total river discharge (red bars) to Region II increased between 1990 and 1995, followed by around 40% drop in 1996 and a subsequent increase until 2003, when a new reduction occurred that was relatively stable in the following years (2004 – 2006). The first question raised from this analysis was whether this pattern reflects the 'true' trend-pattern, is purely a reflection of a change in the number of rivers reported or represents other inconsistencies in the reported data. A more closer look, on a subset of single large rivers showed that this pattern was mainly due to the discharge pattern for the river Rhine that account for some 25% of the total annual river water discharge to Region II (Figure 4.13; blue bars in upper panel). Although less prominent as for the Rhine, the river Elbe also showed a similar pattern (Figure 4.13; lower panel). These observations should be kept in mind in the analyses of the riverine inputs given in the following sections.



Figure 4.13: Annual total river water discharge 1990 – 2006 to Region II (red bars in both charts), river Rhine (blue bars in the upper chart) and Elbe (blue bars in the lower chart). The relative shares (% of total) of Rhine and Elbe to the total water discharge in Region II are indicated by a solid line.

Cadmium (Cd)

The annual riverine inputs of cadmium varied from 51 to 17 t, with a median value of 32 t (Figure 4.14). A downward step-trend was noted with three different distinct periods (1990 - 1995; 1996 - 2002 and 2003 - 2006) with relatively similar inputs within each period followed by a sudden downward jump in the next period. At first sight, the reason for this pattern seems to be related to the same step-trend periods of the river water discharge pattern described in the previous section. However, for cadmium there is decrease in the discharged amount between the three periods while for the water discharge there is no such change in levels between the periods.



Figure 4.14: Riverine inputs, flow-adjusted riverine inputs and direct discharges of cadmium (Cd) to Region II

A further study of the annual national data reports did not reveal any further insights into this issue. However, when analysing data from the five main rivers, it was evident that the trend in the first period (1990 – 1995) to a large degree was explained by a considerable drop in cadmium concentrations between 1990 – 1994 and 1995 – 2006 in the river Seine (Figure 4.15). In the period 1990 – 1994, the river Seine was responsible for between 7% (in 1990) and 40% (in 1993) of the total riverine inputs of cadmium to Region II. The annual riverine cadmium input from the river Seine shows a considerable drop from the year 1995 onwards. Such a substantial change within just three years could be regarded as remarkable. The high annual input in 1993 from the river Seine is primarily due to one single concentration observation only.

For the river Rhine no step-trend in the Cd concentrations could be detected. However, a different pattern in the concentration trend exists between three different branches of the river (Figure 4.16); the Haringvliet consists of a mix of rivers Rhine and Meuse waters, though.



Figure 4.15: Annual average riverine concentrations of cadmium, lead, copper and mercury in the river Seine, 1990 – 2006

It was found that the riverine inputs of cadmium were flow-dependent, but this could not explain the trend even though the step-trends become less prominent when inputs where flow-adjusted (Figure 4.14). The statistical test based on flow-adjusted inputs still revealed a highly significant non-monotonic decreasing trend after LOD-correction (p < 0.02). Noteworthy was also the substantial difference in the change of the level between 1990 and 2006, estimated at around 60% without LOD-correction and 20% with LOD-correction. This might indicate that (changes in) LOD may be one explanation to the step-trends.

For the LOD-correction method changes in LOD-values over the years has not been taken into account. Reason is that there has been no procedure or requirement within OSPAR for reporting on changes in LOD. The assumption that such changes may have a considerable influence on input estimates, for cadmium but also for *e.g.* for mercury, cannot be substantiated. There are, however, examples from Norway where there are considerable changes in LOD-values during 1990 – 2006 (Table 3.9 for Pb), a situation that most likely has also taken place in other countries due to general improvements of chemical laboratory techniques over time. Annex II shows the most recent reported LODs for cadmium.

The direct discharges of cadmium were on average at least a factor ten lower in comparison with the riverine inputs (median = 2.6 t/yr; range 0.5 - 7.3 t/yr) (Figure 4.14). The statistical test detected a highly significant non-monotonic decreasing trend (p <10⁻⁶), which was estimated as a 95% decrease. It should, however, be noted that the cadmium discharges from Denmark and France were not included in the analyses due to incomplete reporting. In addition, many countries have in their annual reports reported on existing uncertainties, especially for the early 1990s. For example, in the

Netherlands there was a different monitoring system in 1990 – 1992 compared subsequent years. It was, however, not reported what these changes where and how they may have affected the magnitude in discharges and the subsequent trend results.





Copper (Cu)

A step-trend was observed for the copper riverine inputs in the same three time periods as detected for cadmium: for 1990 – 1995, 1996 – 2002 and 2003 – 2006 (Figure 4.17), albeit less prominent. The particular hydrological conditions in the period 2003 to 2006 may have influenced the inputs, however, and the confirmation of any decrease should be assessed over a longer time period. The same pattern in input data for both cadmium and copper might indicate two things. Firstly, that the same pollution source is responsible for the trend and, secondly, incompleteness in the reported data *e.g.* in the number of rivers included. Changes in LOD is a less likely explanation as reported Cu concentrations were usually well above the LOD, but information about the number of LODs used for Cu were not available for this assessment. In addition, changes in laboratory methods may have been part of the explanation. For example in Sweden, there was a change in method in 1995.

The riverine Cu inputs were found to be flow-dependent, but the water discharge did not explain the decrease in inputs over time, even though the flow-adjusted inputs levelled out some of the variability between adjacent years. The riverine inputs for copper varied from around 1500 to 840 t/yr (flow adjusted values), with a median value of 1050 t/yr (Figure 4.17). A highly significant non-monotonic decreasing trend in the flow-adjusted riverine inputs was detected (p <0.00005), with a decrease in input of about 40% between 1990 and 2006.

The direct discharges were on average almost a factor 7 lower in comparison with the riverine inputs (median = 151 t/yr, range 312 - 79 t/yr) (Figure 4.17). A highly significant non-monotonic decreasing trend (p <0.00002) was detected, with a reduction in direct discharges of around 70% between 1990 and 2006. Copper discharges from Denmark and France were not included in the analyses, however, due to incomplete reporting. In addition, many countries have in their annual reports reported uncertainties, notably for the early 1990s.



Figure 4.17: Annual riverine inputs and direct discharges of copper (Cu) to Region II

Lead (Pb)

Similarly to cadmium and copper, a step-trend could be detected in the same three time periods Figure 4.18. Changes in LOD are a less likely explanation to the trend given that Pb concentrations, similar to Cu, are normally well above LOD. The riverine inputs were found to be flow-dependent, but did not explain the reduction in inputs over time. Even after flow-adjustment there remained a relatively large inter-annual variability. The annual riverine inputs of lead varied with a factor three between the most extreme years (440 t and 1280 t; flow-adjusted) with an overall median value of 832 t. A highly significant non-monotonic decreasing trend in the flow-adjusted riverine inputs was detected (p <0.0005), an estimated decrease in input of about 50% between 1990 and 2006.

The direct discharges of lead were on average more than a factor 17 lower than the riverine inputs (median = 50 t/yr; range 21 – 130 t/yr) (Figure 4.18). A highly significant non-monotonic decreasing trend was detected ($p < 3 \times 10^{-6}$), or about 80% between the years 1990 and 2006. Lead discharges from Denmark and France were not included, though. Many countries reported uncertainties in especially the earlier 1990s. This might indicate that the reported percentage reduction has been overestimated.



Figure 4.18: Annual riverine inputs and direct discharges of lead (Pb) to OSPAR Region II

Mercury (Hg)

The riverine inputs of mercury to Region II showed a step-wise trend in three distinct periods (1990 – 1995, 1996 – 2002 and 2003 – 2006, Figure 4.19), similar to Cd, Cu and Pb. Whether this reflects a true decrease or is the result of changed reporting and/or changed LODs due to improved analytical methods was not possible to determine given the very scarce information made available. The trend analysis showed a highly significant monotonic downward trend (p <0.0005) with a decrease of about 70% over the period 1990 – 2006.



Figure 4.19: Annual riverine inputs and direct discharges of mercury (Hg) to OSPAR Region II

The annual inputs of Hg by direct discharges are presented in Figure 4.19. The relatively high discharges in the years 1990 and 1991 are remarkable. The trend in the reduction of direct discharges was highly significant ($p < 5 \times 10^{-5}$), and amounted to some 75% between 1990 and 2006. In the explanatory notes to their annual RID reports many countries reported uncertainties, in particular with

regard to the early 1990s. As compared to the riverine inputs, the direct discharges represented only a marginal part of the total inputs.

Total nitrogen (tot-N)

Compared to phosphorus, the annual riverine inputs of total nitrogen to Region II showed relatively low inter-annual variability during the time period 1990 – 2006 (Figure 4.20). Flow-adjustment reduced the inter-annual variability resulting in a rather smooth downward pattern and low variability between adjacent years. The years 2005 and 1994 were the two years with the lowest and highest riverine flow-adjusted inputs (805 kt/yr and 1186 kt/yr, respectively). The high riverine inputs of tot-N in 1994, 1995 and 2002 are explained by high water discharge (see Figure 4.13). A significant non-monotonic decreasing trend in the flow-adjusted riverine inputs was determined (p <0.0001), which in percentage was estimated to represent a 20% decrease over the time period considered. This is primarily based on large reductions in the concentration in the rivers Rhine and Elbe (Figure 4.21). Furthermore, a reduction of about 45% since 1989 in the nitrogen inputs to all Danish coastal waters can be calculated if the inputs are adjusted for discharge variation. This reduction in nitrogen inputs can be assigned to a reduction in discharges from point sources (approximately 75 % since the mid-1980s), but also as an effect of reduced losses from agricultural activities. For the other countries we have few concrete explanations for the reasons for the decline.



Figure 4.20: Annual riverine inputs and direct discharges of nitrogen (tot-N) to OSPAR Region II

The input by direct discharges of total nitrogen showed a monotonic non-linear decreasing pattern (Figure 4.22). The statistical test showed a highly significant decrease ($p < 3 \times 10^{-7}$), a decrease in input over the period 1990 – 2006 from 107 to 71 kt/y. The direct discharges are more than 10 times lower than the riverine inputs of tot-N.



Figure 4.21: Annual mean concentrations of total nitrogen (tot-N) in the rivers Seine, Elbe and Rhine (2 branches) for the period 1990-2006

Total phosphorus (tot-P)

The inter-annual riverine inputs of total phosphorus varied with a factor of about two between 1990 and 2006; the years with the lowest and highest riverine inputs were 2005 and 1995 (33 kt/yr and 75 kt/yr, respectively), see Figure 4.22. According to the statistical test the riverine inputs were found to be flow-dependent, which is shown by the somewhat more smoothened curve and less variability between adjacent years in adjusted inputs compared to the unadjusted inputs. Noteworthy is the high adjusted inputs in 2004, breaking the downward trend. This was due to a very high input figure in 2004 reported by the UK for the North Sea South (and probable a typing error by a factor 10; R. Moxon, pers. comm.). Despite this 'outlier', a significant non-monotonic decreasing trend in the flow-adjusted riverine inputs was detected (p <0.0002), and an estimated about 50% decrease between 1990 and 2006.

A closer look at the data for four of the larger rivers (Göta Älv, Elbe, Rhine and Seine) also showed that the riverine inputs of total phosphorus to Region II have been reduced. More specifically, the Seine, Elbe and Rhine (three different branches), all show a decrease in tot-P concentrations. For the Göta Älv no change is observed, but the concentrations are more than an order of magnitude lower than for the other rivers (Figure 4.23). In addition, the overall reduction in riverine inputs of phosphorus to Danish marine waters since 1989 was approx. 80% (after flow-adjustment). This reduction can only be assigned to a large reduction in the discharges from point sources (more than 90% since the mid-1980s). In France, the implementation of the EC Urban Wastewater Directive (91/271/EEC) has improved the wastewater treatment, and hence reduced the discharges of phosphorus *e.g.* by introducing tertiary treatment to a larger number of plants. As a result, between 2001 and 2004, the

connection rate to tertiary treatment plants increased from 26.5 to 46.5%. For the other countries we have few concrete explanations of the reasons for the decline.

The nominal median value of the direct discharges between 1990 and 2006 was 13.1 kt/yr, with a range of 17.3 to 9.2 kt/yr (Figure 4.22). The reason for the high discharges in 2006 is unknown. For the direct discharges a highly significant (p < 0.004) decrease of 30% over the period 1990 – 2006 was calculated.

The total inputs (direct discharges and riverine inputs) showed a high inter-annual variability



Figure 4.22: Annual riverine inputs, direct discharges and total inputs (lower panel) of total phosphorus (tot-P) to Region II



Figure 4.23: Annual mean concentrations of total phosphorus (tot-P) in the rivers Seine, Elbe, Rhine (3 branches) and Göta Älv



4.3 Riverine Inputs and Direct Discharges to OSPAR Region III (Celtic Seas)

Figure 4.24: Map showing the OSPAR maritime area. OSPAR Region III (Celtic Seas) is highlighted in dark blue

4.3.1 Regional coverage

The UK reported on 15 sampling regions within the OSPAR Region III, which includes well over 200 rivers (Figure 4.25). As mentioned earlier, the data concern aggregated inputs for the sampling regions and not individual rivers. The UK regions discharging to the Region III cover 67 000 km². Ireland reported on 14 rivers discharging into Region III, which account for a total catchment area of $35\ 000\ \text{km}^2$.



Figure 4.25: Riverine sampling regions in the United Kingdom

4.3.2 Data check prior to assessment

First, the annual inputs data were screened for incompleteness, inconsistencies and anomalies. In this process, on the basis of the agreed 'exclusion rules' the input data for mercury (Hg) reported by Ireland were excluded. For Hg only the inputs from the UK to Region III could be taken into account. Furthermore, for some of the Irish data interpolations were necessary to allow a consistent trend analysis.

The runoff pattern for direct discharges showed a peculiar pattern with a sudden drop of a factor ten in 1995 to 347 000 m^3/d , and remained at that level for the subsequent years.

4.3.3 Results and discussion

River water discharge (Q)

The total reported river water discharge to Region III showed rather low inter-annual variation and no obvious trend (Figure 4.26).



Figure 4.26: Total reported river water discharge to OSPAR Region III during the period 1990-2006

Cadmium (Cd)

Both measured and flow adjusted riverine inputs of cadmium are shown in Figure 4.27. In the early 1990s the annual riverine inputs were between 10 and 20 t. They dropped to below 10 t/yr from 1995 onwards. The reason for the general downward trend is most likely due to changes in LOD over the years. The level of decrease of riverine inputs of cadmium between 1990 and 2006 is about 60%; the detected downward trend is highly significant (p < 0.0002).

The range of direct discharges of cadmium to Region III over the considered period decreased from 27 t/yr in 1990 to about 0.2 t/yr in 2004 (Figure 4.27), or about 95%. The downward trend is highly significant ($p < 3 \times 10^{-6}$). From 2004 onwards the direct discharges represent a minor contribution to the total inputs of cadmium to Region III as they became strongly dominated by riverine inputs. The general decline in cadmium inputs from rivers could be a result of changed LOD over the time period (but quantitative data on this was lacking).



Figure 4.27: Riverine inputs and direct discharges of cadmium (Cd) to OSPAR Region III

Copper (Cu)

Riverine inputs of Cu show considerably annual variations; they range between 170 t/yr in 1993 and 400 t/yr in 2003 (Figure 4.28). Both measured and flow adjusted riverine inputs are shown in. The level of decrease of flow-adjusted riverine inputs of copper between 1990 and 2006 is about 30%; the downward trend is highly significant (p < 0.044). Notable is the relatively high riverine input in 2006, even after flow-adjustment. The reason is unknown.

The direct discharges of copper to Region II has for the reporting period always been lower than the riverine inputs. They become a minor portion of the total inputs since 2002 (Figure 4.28). The range of direct copper discharges over the considered period varied from about 100 t/yr in the early 1990s to 20 t/yr in the period 2002 – 2006. From the trend analysis, the level of decrease of the direct discharges of copper between 1990 and 2006 is about 90%; the downward trend is highly significant ($p < 2.2 \times 10^{-6}$).



Figure 4.28: Annual riverine inputs and direct discharges of copper (Cu) to OSPAR Region III

Lead (Pb)

The inputs of lead in Region III show considerably annual variations and range between 370 t/yr in 1990 and 92 t/yr in 2003, with an average of 227 t/yr. Both measured and flow adjusted riverine inputs are shown in (Figure 4.29). The level of decrease of riverine inputs of lead between 1990 and 2006 is about 20 %, but the declining trend is not statistically significant (p < 0.2).

Direct discharges of lead are presented in (Figure 4.29). The range of direct lead discharges over the considered period varied from about 94 t/yr in the 1993 to 17 t/yr in 2005. The average annual direct discharges of lead were 48 t/yr. After an initial increase in the early 1990s, the direct Pb discharges decrease from 1998 onwards. From the trend analysis, the level of decrease of the direct discharges of lead between 1990 and 2006 is about 43%, but this was not statistically significant (p<0.11).

The total inputs of lead to OSPAR Region III have since 2002/2003 been strongly dominated by riverine inputs due to the decline in direct discharges. Total inputs of lead in 2006 were at about the same level as in the beginning of the 1990s.



Figure 4.29: Riverine inputs and direct discharges of lead (Pb) to OSPAR Region III

Mercury (Hg)

The assessment of the inputs of Hg to Region II is based on UK data only.

The riverine inputs of mercury ranged from 2-3 t/yr in 1990 – 1995 to about 0.5 t/yr in 2003 – 2006, with an average of about 1.4 t/yr (Figure 4.30). The level of decrease of riverine inputs of mercury between 1990 and 2006 was about 85 %, and the decreasing trend is highly significant ($p<5\times10^{-6}$).

The direct discharges of mercury over the considered period varied from about 3 t/yr in 1993 to less than 0.1 t/yr in 2004-2006, with an average annual direct Hg discharge of 0.7 t (Figure 4.30). Trend analysis showed that the level of decrease of the direct discharges of mercury between 1990 and 2006 was about 95%; the downward trend is highly significant ($p < 2.2 \times 10^{-6}$).

Total inputs of mercury to Region III from Ireland have been reduced considerably since the 1990s, but the 'real' reduction in inputs may be smaller than measured due to changes in LOD during the considered period. Direct discharges of mercury, which were at the same level as riverine inputs early in the 1990s, now represent only a small portion of the total inputs.



Figure 4.30: Annual riverine inputs and direct discharges of mercury (Hg) to Region III

Total nitrogen (tot-N)

There have been relatively small annual variations in flow-adjusted riverine inputs of total nitrogen between 1990 and 2006, at an average of about 250 kt/yr. Both measured and flow adjusted riverine inputs are shown in Figure 4.31. The low inputs in 2003 could to a large degree be explained by water flow, as can be seen from the flow adjusted values for that year. The level of decrease amounts to about 12 - 13% for over the full period; the slightly decreasing trend is not significant (p <0.28).

The annual range of the direct discharges of total nitrogen has been between 53 kt in 1992 and 30 kt in 2006, with an average of 39 kt (Figure 4.31). From the trend analysis the level of decrease of the direct discharges of nitrogen between 1990 and 2006 is calculated at about 30%; the downward trend is highly significant (p < 0.002).

The level of total inputs of nitrogen to Region III in 2006 was about the same as in the beginning of the 1990s. No statistical trend could be detected for tot-N. The total input of nitrogen is strongly dominated by riverine input as the direct discharges represent about 15% of the total inputs.



Figure 4.31: Annual riverine inputs and direct discharges of nitrogen (tot-N) to Region III

Total phosphorus (tot-P)

Both measured and flow adjusted riverine inputs are shown in (Figure 4.32). There have been considerable annual variations of riverine inputs of total phosphorus to Region III in the monitoring period, with a range from 7 kt in 2003 to 20 kt in 1998, with an average annual riverine input of about 12 kt. A trend assessment of the period 1990 to 2006 indicates a decrease in the order of 20%, despite that the input levels in 2005 - 2006 were similar to the early 1990s. The downward trends are not significant for both unadjusted and flow adjusted inputs (p <0.52 and p <0.25, respectively).

Direct discharges of tot-P varied for the monitoring period between about 6 kt/yr and 13 kt/yr, with an average of 8.4 kt/yr (Figure 4.32). They constitute a considerable portion of the total input of phosphorus in Region III, in the order of 40% of the total inputs. The level of decrease over the considered period is about 55%, and the downward trend is highly significant (p < 0.0001).



Figure 4.32: Annual riverine inputs and direct discharges of phosphorus (tot-P) to Region III



4.4 Riverine Inputs and Direct Discharges to OSPAR Region IV (Bay of Biscay and Iberian Coast)

Figure 4.33: Map showing the OSPAR maritime area. OSPAR Region IV (Bay of Biscay and Iberian Coast) is highlighted in dark blue

4.4.1 Regional coverage

The assessment covers OSPAR Region IV with catchments from France, Spain and Portugal. The OSPAR drainage area is 655 000 km² (Table 4.4), and more than 75% of the total drainage is covered by six rivers. The basins of many of the international Iberian rivers are significantly regulated, with installed storage capacities ranging from 25% to 200% of the average annual run-off. It is possible to store 95% of the run-off in the Tagus river, while the storage capacity of the Guadiana river is currently larger than the annual run-off.

River	Drainage area (km²)
Loire	118,420
Douro	97,682
Tagus	80,629
Garonne/Dordogne	79,180
Guadiana	66,960
Guadalquivir	54,970
Other river basins	156,896
Total	654,737

Table 4.4: Drainage areas for the six largest rivers in Region IV
4.4.2 Data check and analysis undertaken

The gaps and incomplete time-series made it impossible to perform a trend analysis on a sound scientific platform. Attempts to perform such analyses and assessments were, however, made based on the most complete data. These included:

- Cd, Cu and Pb: 1990-2006, based on the datasets from Spain and Portugal;
- tot-N: 1997-2006, based on the datasets from Spain and Portugal;
- tot-P: 1997-2006, based on the complete datasets from France, Spain and Portugal.

4.4.3 Results and discussion

The reported inputs based on these datasets are given in the Figures 4.34 to 4.38, but they are not further commented due to the mentioned high uncertainties. It is also, from the figures, obvious that even these datasets suffer from uncertainty as given by *e.g.* the large inter annual variability especially for the metal inputs. The nutrient series (10 years) may also be regarded as too short for adequate statistical trend analyses.



Figure 4.34: Annual riverine inputs of cadmium (Cd) to Region IV (Spain and Portugal)



Figure 4.35: Annual riverine inputs of copper (Cu) to Region IV (Spain and Portugal)



Figure 4.36: Annual riverine inputs of lead (Pb) to Region IV (Spain and Portugal)





Figure 4.37: Annual riverine inputs of total nitrogen (tot-N) to Region IV (Spain and Portugal)

Figure 4.38: Annual riverine inputs (1997-2006) of total phosphorus (tot-P) to Region IV (France, Spain and Portugal)

4.5 Limitations and uncertainties

All riverine inputs reflect a mix of natural and anthropogenic sources, with natural sources in many cases being negligible compared to anthropogenic sources. No attempt has been made in this assessment to differentiate the two.

4.5.1 Identified data gaps, anomalies and/or inconsistencies

Gaps, anomalies and inconsistencies are linked to issues such as:

- Improved laboratory analyses and subsequent change of detection limits over time (related in particular to the long-term RID assessment in order to be able to produce reliable trend analyses);
- Direct discharges and diffuse losses related primarily to differences in methodology;

- Changes in the location and/or number of sampling sites and coverage of monitored areas (related to the distance from the sampling site to the sea, with information on what *was* done to estimate the areas downstream of the sampling sites);
- Changes in sampling procedures (related to the representativeness of the samples, as well as the question of common sampling strategies);
- Hydrological data and calculation practices (related to the type of hydrological monitoring performed by the Contracting Parties);
- The need for harmonisation of monitoring practices between contracting parties.

4.5.2 Completeness of monitoring

(related to the requirement of monitoring 90% of the total land area)

According to the RID Principles, 90% of the land area draining into the OSPAR maritime area shall be monitored. Of the countries that have made information available for the RID Centre exercise, only the Netherlands, Germany and Belgium report that more than 90% of the drainage area has been monitored.

4.5.3 Completeness of time-series

A number of time-series has been discarded, *i.e.* inputs and assessments have not been taken into account because of their inconsistencies, gaps, etc (see also Section 2.3.2 on preparation of datasets). Examples of such situations are (see also Section 3):

For Region I:

- The trend assessment *was* based on data from Norwegian rivers only due to too short time series for the Icelandic rivers;
- The Norwegian direct inputs of Cd, Hg and Pb to the Barents Sea were not taken account of as data from 2004 2006 were missing.

For Region II:

- The heavy metal discharges to the Kattegat, Skagerrak and the North Sea of Denmark were not taken account;
- All Channel data submitted by France, except those for the Seine, were not taken account.

For Region III:

• Input data of mercury from Ireland were not taken into account.

For Region IV:

- Data for nutrients from Spain were only available from 1997 and onwards, hence no trend analysis covering the entire time period 1990 2006 could be undertaken;
- The availability of data from France for riverine inputs of total nitrogen and heavy metals was too low to perform a satisfactory trend assessment.

Contracting Parties also have very different monitoring strategies for their rivers. Whereas some countries report on a few, large rivers (such as Germany), others include a large set of smaller rivers draining into the coastal areas (*e.g.* UK, Ireland, Norway, Denmark and Sweden). This may affect the number of samples annually collected from each river. However, most countries sample a minimum of 12 samples per year from their main rivers, but there are exceptions (*e.g.* Ireland and Iceland). Most Contracting Parties reported that the sampling was evenly distributed as once per month (*e.g.* Norway, UK), or more often (*e.g.* Belgium, the Netherlands). Germany reported that their sampling was based

on cross-section sampling (*i.e.* samples from several defined positions on the river cross-section) in the rivers Elbe (fortnightly), Weser and Ems (monthly), and representative random sampling (*i.e.* several samples taken across the river but not at defined positions) in the Eider (monthly). None of the country appeared to conduct event-triggered sampling (*e.g.* sampling during flood events). For those countries reporting on 'tributaries', the sampling frequency varied. Germany sampled their tributaries monthly (representative random sampling), whereas in France the sampling frequency for tributaries varied between 4 and 12 times per year. In Spain and Belgium the tributaries were usually sampled 12 times a year. In Norway, 'tributary rivers' entering the sea were sampled 4 times a year. An effort *was* made, however, to sample during different climatic conditions (such as snow melt season, summer low flow, autumn rains).

4.5.4 Sources not accounted for

Unmonitored areas

Inputs from unmonitored areas downstream sampling points were not taken account of in this assessment. Within the RID-Centre exercise, information has been gathered as to the importance of such unmonitored areas (OSPAR, 2008c). France, Norway and Sweden have provided information on the unmonitored areas downstream of the sampling locations. For these countries, the unmonitored area ranged from about 3000 to about 30 000 km². For some countries, a relative significant area in catchments reported as 'monitored' was in fact not monitored. Four of the Contracting Parties have reported that they do not estimate the inputs from these areas, whereas six Contracting Parties have reported different methodologies for estimates, as follows (based on OSPAR, 2008c):

- Belgium: Assumed that there are no sources downstream of sampling points;
- France: The area downstream of the river sampling points *was* regarded as 'OSPAR Coast' for the main rivers. All other areas downstream of the sampling points were included as direct discharges. For the areas that have no 'correct' sampling point, the inputs were estimated with a reference monitored tributary;
- Germany: For the Eider catchment area calculation of inputs was made by extrapolation of the monitored areas;
- Iceland: Pollution sources downstream of the two monitored rivers were not included in the reporting:
- Ireland: Calculations of inputs were made by extrapolation of the monitored areas;
- Netherlands: Losses from unmonitored areas were generally not reported as most sampling points were at sluices and river mouths. The exception *was* the location Maassluis (Rhine), where some direct discharges downstream of the sampling point were reported;
- Norway: Modelling was used to determine the inputs from the unmonitored areas; the model was based on estimated inputs from diffuse sources (natural background values; agricultural land; sewage from scattered dwellings) and point sources (industry; waste water treatment plants; fish farming);
- Spain: Inputs from unmonitored areas were not calculated;
- Sweden: Calculations of inputs were performed by using the area specific load from similar and adjacent monitored areas.

From the above listed information, it becomes clear that Contracting Parties have different ways of dealing with the reporting of input areas downstream of river sampling points. Since coastal areas in Europe often are important sites for industry, cities, tourist resorts, mariculture, harbours and other

activities, it is possible that the areas downstream the sampling locations may contribute significantly to the total inputs.

Point sources

A number of Contracting Parties has reported data on point sources such as industries, sewage treatment plants and aquaculture. Other countries do not seem to include such data in their reporting, *e.g.* Iceland. In Sweden, annual reporting of direct discharges was restricted to municipal treatment plants designed for more than 2000 'population equivalents' and the most important industrial point sources. In Spain, the industrial effluents reported were based on industries' discharge declarations, regional discharge registries, direct control measurements, discharge permits, concentration values from previous years when effluents where considered similar and data were not available, and fixed values when measurements were below detection limits. Belgium reported that no point sources exist outside the monitored rivers catchments. For all point source estimations made by Contracting Parties it is believed that the number of samples varies among the different discharge sites, but no specific information has been made available. In general it seems that the reporting of point sources is inconsistent and in many cases highly insufficient to provide a complete overview of direct discharges to the various Regions of the OSPAR maritime area. Other possible direct sources that were not taken into account because data were lacking may include storm water overflows from sewage treatment plants.

4.5.5 Uncertainties and limitations in data; data quality

Event based sampling

Riverine inputs are usually highest during floods and in some types of rivers a majority of the inputs may be transported during only a few days of high discharge conditions. Hence, sampling during flood events is regarded as important in order to achieve realistic estimates of the loads. However, most Contracting Parties do not include in their monitoring strategy to allow for additional sampling during floods. This may be due to the extra costs associated with additional sampling which not only comprises the cost of analyses, but also the logistics of organising sampling at short notice during flood events.

Laboratories

For the chemical analysis of samples, several countries use different laboratories for the riverine input samples and the direct discharges samples. However, Ireland, Norway and Sweden (except for Hg) used the same laboratory thereby obtaining, most likely, a better platform for a holistic assessment.

4.5.6 Aggregation to regional level versus catchment related data, level of uncertainty

A number of questions may also be raised that relate to issues of data uncertainty (see also OSPAR, 2008c), *viz*.:

- Why is an awareness of uncertainty essential in evaluating the state of knowledge about environmental variables/systems?
- How can information on uncertainty be obtained in the first place and what are the problems to be solved?
- How can the uncertainty related information be organised and used in a way that is useful for answering practical questions about the sufficiency and accuracy of results?
- What is the uncertainty at all levels of data gathering and 'data manipulation'?
- What is 'acceptable' uncertainty for water managers, for Contracting Parties, for OSPAR?

Contracting Parties should endeavour to assess the uncertainty of the results that they 'accept', or – phrased in a different way – costs and benefits of more accurate data should be assessed and become a common understanding to all parties involved.

4.5.7 Differences in use of lower/upper estimates, LOD vs. LOQ

There are discrepancies in the ways Contracting Parties report upper and lower values. The concept of upper and lower values is derived from the detection limits of each parameter. The reported data should be seen in light of the Limits of Detection (LODs) and Limits of Quantification (LOQs) reported. The general idea is that in those cases where the analytical results reported are below the limit of detection, two load estimates should be supplied, one assuming that the true concentration is zero and the other assuming that the true concentration is the limit of detection. This will provide minimum and maximum concentrations within which the true estimate will fall. As was explained in section 2.3 these data will then provide lower and upper boundaries for the estimate. Another source of discrepancies for the upper estimates could have been the use of the LOQ instead of the LOD. Countries using LOQ report loads of contaminants (based on upper estimates) which are significantly higher (roughly a factor three) than those using the LOD.

Within the RID-Centre activities, OSPAR Contracting Parties have been invited to clarify whether they used limits of detection (LODs) or limits of quantification (LOQs) for their reporting. The definitions of LOD/LOQ are presented in the Glossary. The information on the LOD/LOQ was reported by Contracting Parties is presented in OSPAR (2008c). It is, however, not always clear from the information submitted by the Contracting Parties to which extent the reported values reflect LODs or LOQs. In some cases both may have been used in parallel. For instance, Spain has submitted an overview showing that LOD is used in País Vasco and Guadalquivir, whereas LOQs are used in Galicia, Guadiana and the Northern region. Most countries preferred, for various reasons, to use LODs, however.

Most Contracting Parties reported ranges of LOD/LOQ values. In most cases the reason for this was that different detection limits were obtained by different laboratories involved (in different regions). It is remarked that detection limits for the analysis of riverine inputs will generally have to be lower and more challenging than those used for direct discharges. Most countries have reported a good overview of the regional laboratories and the detection limits achieved.

The fact that detection limits may vary considerably for river sample analyses within the same country when different laboratories perform the analyses, and also that they use different analytical methods or change analytical method at different moments in time, demonstrate that great care must be taken when undertaking and interpreting trend analyses.

The most recent LOD values reported by the Contracting Parties for the six determinants are listed in Annex II, together with the Region(s) concerned. Some countries appear to have rather high LODs (or LOQs) for determinants that are present at relatively low concentrations, such as for cadmium and mercury. For example, the recommended detection limit of cadmium is 0.01 μ g/l, but there were reports of detection limits of 2, 5 and even 20 μ g/l. This leads to striking differences in input estimates for those substances where either upper or lower estimates were used.

4.5.8 Recommendations

It is important that Contracting Parties have access to detailed information about detection limits and analytical laboratory methods. *Changes in laboratories and detection limits over time should be duly registered* – preferably in a common database. There were examples in this assessment that such changes could influence the trends significantly.

The RID Principles state that it is necessary to choose an analytical method, if available at a reasonable cost, which gives at least 70 % of positive findings (*i.e.* no more than 30% of the samples below the detection limit). This seems problematic especially for countries with relatively clean rivers, even at LOD values that correspond to the OSPAR standards, *e.g.*, in Region I.

In this assessment, it was noted that detection limits have changed over time, vary considerable between countries and laboratories and are, in some cases, far above the OSPAR recommended values. The use of different detection limits is clearly a considerable problem both within countries and for comparison of result between countries (and between years) and should be considered in future reporting. *The detection limits should be at least as low as the limits adopted by OSPAR in 2005.*

Optimally the same accredited laboratory should be used for all rivers within a country. If this is not possible, *intercomparison of results should take place. In cases of changes of laboratories, over time an intercomparison should be carried out during a certain 'transfer of responsibility period'.* This was most likely a problem in some countries both in terms of using several laboratories and in cases where laboratories have changed in the period 1990 to date.

Historical QA, i.e. monitoring results checked against historical data should be undertaken by qualified researchers with experience in assessing water quality data. This should be done as soon as possible after analysis so whenever possible anomalies are detected, the samples can be re-analysed.

Given differences in and quality of reporting between countries and over years (*e.g.* due to reason mentioned above) trustworthy trend analyses may be difficult to carry out for aggregated data such as for the four Regions in this assessment. *Attempts of further harmonisation of data between countries and historical reconstruction should be pursued.*

The implementation of the various trend assessments in this study and also in the 2005 trend assessment (for period 1990 – 2002), highlights the importance of ensuring that *datasets* for inputs and flows are as complete and consistent as possible before applying statistical methods. Ideally, all significant anomalies should be addressed and *Contracting Parties should be urged to complete their datasets prior to the assessment*.

There is, for some *datasets*, an indication of under-reporting in the earlier years of the RID programme, as well as a higher degree of uncertainty in the reported values in the beginning of the time series. This is a factor which could influence the outcome of any assessments by *e.g.*, reducing any downward trends that are identified or increasing any upward trends. This is an issue that should be considered more fully before any further trend analyses are carried out. One approach would be to reduce the period of assessment to include only those years for which data are more consistent.

Recognising that inputs and direct discharge datasets are not so well developed for Region IV as they are for other Regions, one should *consider how best to progressively develop and present the data that is available for this Region*, and seek to undertake assessments when sufficient time series of consistent data become available, either at local or regional scale.

Comments on long-term trends were generally lacking in the country reports for single years; the comments were too scarce and mainly include details about deviations and changes to the previous years. To support future assessments, reporting comments after a *simple visual inspection of the reported annual values in a full time-series perspective is recommended, as well as a short explanation for identified obvious trends.* Explanations could for example be on general trends in improved waste water treatment, changed and implemented policies, changed reporting quality.

5. Part III – Catchment Assessment

5.1 Introduction

The trend assessment at a catchment level was conducted on a selected number of rivers, for which data were provided on a voluntary basis by France, Germany, Iceland, the Netherlands, Norway and Sweden of RID monitoring data. A 'catchment' is defined as the area drained by a river or body of water, also called a 'river basin'. The catchment approach and the regional approach (Part II) are statistically different in character in terms of the level of detail of the input data used for the assessment. The regional assessment was based on aggregated data of a large number of rivers lumped together and was carried out only on annual data, the minimum time-resolution available. The catchment approach was based on the RID data for each single river and was carried out on monthly data before the results were aggregated to annual total inputs. The comparison between the approaches, assessment of *e.g.* similarities or differences in trends, LODs and uncertainties were summarised in Part I of this report. All riverine inputs reflect a mix of natural and anthropogenic sources, with natural sources in many cases being negligible compared to anthropogenic sources. No attempt has been made in this assessment to differentiate between them.

The detailed statistical analysis and catchment data are electronically available as <u>Technical</u> <u>Supplement 2</u>. <u>Technical Supplement 3</u> provides an analyses of data in relation to limits of detection and <u>Technical Supplement 4</u> provides supplementary graphic presentation of catchment data.

5.1.1 Geographic coverage and determinants

The catchment assessment presented covers 10 rivers at 11 sites of the OSPAR Regions I, II and IV. The list of these rivers and sampling sites, as well as some catchment related information, is presented in Table 5.1. The catchment assessment covers 8 rivers with one discharge site. The river Rhine was monitored at two locations (IJsselmeer, Maassluis), while the location Haringvlietsluis is a mix of rivers Rhine (65%) and Meuse waters. Figure 5.1 shows the locations of the 10 catchments. No single river catchment data for Region III was made available by Ireland or the UK.



Figure 5.1: Location of the ten catchments included in the catchment assessment for the period 1990 - 2006

The determinants considered were cadmium (Cd), copper (Cu), lead (Pb), mercury (Hg), total nitrogen (tot-N) and total phosphorus (tot-P). Only the riverine inputs were considered. Information about direct discharges downstream the sampling point were either insufficient or considered as less relevant since the sampling sites, in most cases, are located close to the river mouth.

River (country)	Name of sampling site	OSPAR Region	Total drainage area (km²)	Upstream sampling site (km²)	Agricultural area (%)	Population density (inh/km ²)
Ölfusá (IS)	Selfoss	I	6 100	5 676	2%	0.3
Alta (NO)	Alta	I	8 772	7 373 ¹	2%	0.3
Glomma (NO)	Sarpfoss	П	42 441	41 918 ²	6%	14.4
Göta Älv (SE)	Alelyckan	П	50 119	48 194	14%	17.5
Elbe (DE)	Hamburg- Seemannshöft	II	148 268	139 775	59.8	165
Rhine (NL)	IJsselmeer	П	-	-	-	-
Rhine (NL)	Maassluis	П	-	-	-	-
Rhine/Meuse (NL)	Haringvlietsluis	II	-	-	-	-
Seine (FR)	Amfreville-sous- les-Monts	II	64 953	64 953	66%	215
Loire (FR)	Monjean sur Loire (2002-2006)	IV	110 178	110 178	73%	61
Garonne (FR)	St Léger	IV	38 227	38 227	63%	59

Table 5.1: Characteristics of the 10 catchments

¹ 6 182 km² at water discharge monitoring site

² 40 221 km² at water discharge monitoring site

5.1.2 Data preparation

In order to assure maximum effectiveness of the statistical analysis, the input data were screened for completeness, consistencies and measurements at or below LOD (see also section 2.3). The following table shows the availability of data for the catchment assessment.

Table 5.2: Data availability

- components with complete data between 1990 and 2006
- components with missing values (entire year) or low sampling frequency;

River	Cd	Cu	Pb	Hg	Tot-N	Tot-P	Q
Ölfusá	1996-2007	1996-2007	1996-2007	1996-2007	1997-2007	1997-2007	1996 - Aug.2007
Alta	no data above LOD after 2004						
Glomma							
Göta Älv	until 2007	until 2007	until 2007	1995-2007	until 2007	until 2007	
Elbe	1992-2007	1992-2007	1992-2007	1992-2007			
Rhine (IJsselmeer)	until 2005 only	until 2005 only	until 2005 only	until 2005 only	until 2005 only, 2004 missing	until 2005 only	
Rhine (Maassluis)	until 2005 only	until 2005 only	until 2005 only	until 2005 only	until 2005 only	until 2005 only	
Rhine/Meuse (Haringvlietsluis)	until 2005 only	until 2005 only	until 2005 only	until 2005 only	until 2005 only	until 2005 only	
Seine	1994 + 2005 missing 1993-1996: 4 values per year only	1994 + 2005 missing 1993-1996: 4 values per year only	1994 + 2005 missing 1993-1996: 4 values per year only	1994 missing; 1995: beginning of the year only; 1996: 2 values only	1998-2000 missing		data incomplete
Loire	2002 missing 2003 + 2004: 2 values per year only; no value above LOD after 2000	1992-2002 + 2004 missing 2003: 2 values per year only	1992-2002 missing 2003 + 2004: 2 values per year only; no value above LOD after 2004	2002 missing 2000, 2003, 2004: 2 values per year only; no value above LOD after 2001			
Garonne	4 values only; 2004 + 2005: 2 values per year only	4 values only; 2004 + 2005: 2 values per year only	4 values only; 2004 + 2005: 2 values per year only	4 values only; 2004 + 2005: 2 values per year only		1989-1992 & 1997-2006 only	

With regard to the availability of the data and the trend assessments, the following should be noted in particular:

- For some rivers, the number of analytical results per year was varying and in some cases at a low (annual) sampling frequency; this is especially the case for the trace metals. This may cause varying accuracy and reliability in the temporal trend assessment;
- Spurious trends can be caused by changes in the LOD during the monitoring period as was the case in many rivers, especially for the heavy metals. In the assessment estimated LOESS levels are included when data below the LOD are replaced by both the LOD itself and 0.
- For the river Ölfusá (Iceland), two series of water discharge data (one on a daily basis, one for

the days when concentration measurements were carried out) were available. It was decided to use the daily observations even though they differed from the other data;

- Water discharge values for the IJsselmeer were calculated as a total of the water discharge values to the Wadden Sea at the Afsluitdijk/Kornwerderzand buiten and at the Afsluitdijk/Den Oever buiten monitoring stations. There were several zero water discharge results for these stations. Together they represent the total discharges of the river IJssel, one of the branches of the river Rhine via the lake IJsselmeer to the Wadden Sea. As the adjustment method L1 (see Background document on complete statistical analysis) requires positive water discharge data, the zero values have been replaced by 1m³/s this is the minimum water discharge figure in the data series excluding the zero values. This correction does not influence the assessment, as 1m³/s is negligible compared to the average water discharge;
- The water discharge series for river Seine (France) were not complete. For the missing days, a linear interpolation was carried out to estimate the missing water discharge values. Table 5.3 gives an overview of the percentage of water discharge values missing for the Seine;
- For the river Loire (France), two different sites were used in order to create a merged time series; one site with data for the period 1990 2001, the other with data from 2002 onwards;
- For the river Garonne (France) only total phosphorus was assessed for trends.

Year	% missing	Year	% missing	Year	% missing
1990	42%	1996	57%	2002	0%
1991	0%	1997	0%	2003	0%
1992	44%	1998	14%	2004	0.5%
1993	0%	1999	18%	2005	0%
1994	0%	2000	0%	2006	0%
1995	0%	2001	8%		

Table 5.3. Percentage of missing water discharge values in the river Seine for the period 1990 - 2006

5.2 Results and discussion

5.2.1 General input level characteristics

It should already from the onset be pointed out that the rivers differ in size and therefore also often in magnitude of riverine inputs. The level of concentrations of determinants also varies considerably. Such variations should be borne in mind when the trends are interpreted for each of the determinants in the following sections. As an example, Table 5.4 below shows the variations in the concentrations (long-term averages) of total phosphorus and total nitrogen concentrations in the 10 rivers. They range from 0.07 - 7.33 mg/l for tot-N (factor of more than 100) and 0.012 - 0.559 mg/l for tot-P (factor of nearly 50).

River	tot-N (mg/l)	tot-P (mg/l)
Ölfusá	0.07	0.012
Alta	0.19	0.013
Glomma	0.60	0.021
Göta Älv	0.85	0.020
Elbe	4.97	0.241
Rhine (IJsselmeer)	1.88	0.145
Rhine (Maassluis)	3.12	0.251
Rhine/Meuse (Haringvlietsluis)	3.13	0.169
Seine	7.33	0.559
Loire	4.50	0.228
Garonne	-	0.139

Table 5.4: Long-term average (1990 – 2006) concentrations of total nitrogen and total phosphorus in the ten rivers

5.2.2 River water discharge (Q)

The total river water discharge showed for most rivers an inter-annual variability at least a factor two. For the rivers Göta Älv and Seine this amounted to nearly a factor three, for the river Loire a factor of more than three and for the Rhine/Meuse river (Haringvlietsluis) to a factor of more than 4.

Two exceptions were noted. For the river Ölfusá a difference of less than 30% was reported between the most extreme years in the period 1997 – 2006. The second lowest inter-annual variability was found in the river Rhine (at Maassluis) with only 50% difference between the lowest and highest annual discharge.

Table 5.5:	Years with t	he relatively	lowest and	highest water	discharges in th	ə 11	rivers/sampling sites
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River (site)	Years with low water discharge	Years with high water discharge
Ölfusá	1998, 2001	2004, 2006
Alta	1990, 1994, 2003	1992, 2005
Glomma	1991, 1996	1999, 2000
Göta Älv	1996, 2003	2000, 2001
Elbe	1992, 1993	1995, 2002
Rhine (IJsselmeer)	1996, 1997	1994, 1998, 2002
Rhine (Maassluis)	1998, 2003	2001, 2002
Rhine/Meuse (Haringvlietsluis)	1995, 2004	1995, 2002
Seine	1990, 1991, 1996	2000, 2001
Loire	1990, 1991	1994, 2001
Garonne	2005, 2006	1999, 2004

The high-flow and low-flow years vary substantially between the rivers (Table 5.5). The years 1990, 1996 and 2003 seem to be typical low-flow years while 2001 and 2002 appear to be typical high-flow years. The table illustrates another difficulty in the interpretation when data are aggregated to entire regions.

A visual inspection of the annual water discharges of the rivers could be summarised as:

- No general upward or downward trends in any of the rivers concerned;
- A step-trend (1990 1995, 1996 2002 and 2003 2006) for the rivers Rhine, Meuse and Seine (see also Figure 3.2).

5.2.3 Cadmium (Cd)

Figure 5.2 shows the annual riverine cadmium inputs of the rivers Alta, Ölfusá, Elbe, Glomma, Göta Älv, Rhine/Meuse, Seine and Loire for the period 1990 – 2006, and the total riverine inputs to Regions I, II and IV. There are no riverine input data for river Garonne (Region IV). The details of the statistical analysis are available in the Background document of Statistical Analyses. The monitoring regime varies slightly between the rivers, see also Table 5.2, which provides an overview of each component and the completeness of reported input years, as well as an indication of when the data are based on low sampling frequency.

The inputs were based on monthly data before the results were aggregated to annual total inputs. To illustrate similarities and/or differences in input patterns between Regions and individual Catchments, the total inputs for the three regions are also shown together with the figures of the rivers discharging into the relevant Regions. A more in depth analyses of such comparisons is made in Section I (Synthesis of the two Approaches) of this report on selected parameters, rivers and regions.



Figure 5.2: Annual cadmium inputs for Regions I, II and IV (left column) and the annual cadmium inputs from selected rivers in these Regions (right column). Inputs by the river Ölfusá are not included in the total for Region I

When studying the various charts (also for the other determinants) it is important to note that the scale of the y-axis of the 'Region' charts and that of the 'Catchment' charts are not always the same.

With regard to the inputs of Cd a number of comments can be made:

- Region I:
 - For most years the share of the rivers Ölfusá and Alta represent a relatively low proportion of the total riverine inputs of cadmium to Region I, mostly about 10% or lower; exceptional

is the year 2006 when the reported inputs of cadmium from the river Ölfusá were very high (about 10 times higher than average) and represented close to 90% of the total inputs to Region I. However, as was mentioned above, the inputs of the river Ölfusá were not included in the Region I total;

- There has been a significant reduction in both the total Cd riverine inputs to Region I (in 2006 4 8 times lower than in the early 1990s) and in the inputs from the river Alta (15 20 times lower).
- Region II:
 - For most years the share of the selected rivers represent about 25 to 35% of the total riverine inputs of cadmium to Region II, with the exception of the years 1993 (exceptionally high inputs from the river Seine) and 1995 (exceptionally high inputs from the rivers Rhine/Meuse);
 - The order of magnitude of the estimated inputs of cadmium are reduced by a factor of about two when one considers the total riverine inputs of cadmium to Region II between early 1990s and 2005 – 2006, about 7 for the river Seine, 2 for the river Elbe, 3 – 5 for the river Glomma, whereas for the rivers Rhine, Meuse and Göta Älv the inputs appear to be of the same order of magnitude, with considerable variations between years for each of the rivers.

From the figures in the overview of complementary data and statistics (<u>Technical Supplement 2</u>) it is obvious that it is the concentration and not the water discharge that is the main cause for the identified trends in riverine inputs. However, these trends should be interpreted with caution, e.g. because for Cd in general there is a high percentage of the results that was reported below LOD, and – for some rivers – also a change in LODs during the monitoring period (see Annex II).

As an illustration, the cadmium results from the river Loire show a statistically significant upward trend. However, the analysis is highly affected by the number of samples below the LOD (since 2000 all measurements were below the LOD) and the large fluctuations in LOD ($0.1 - 10 \mu g/l$). Table 5.6 shows the lower and the upper riverine inputs in Loire (and other rivers) for two different years, clearly addressing the problem to assess trends with varying LODs levels.

Another example is for the rivers Rhine/Meuse sampled at Haringvlietsluis. In 1990, the trend of the LOESS is not affected by data below LOD. This is due to low LOD in the first years. As LODs are higher later years, there are spurious upward trends as a result of the number of measurements below LOD.

The analysis for the sampling site IJsselmeer (river Rhine) also indicates that the analyses might be affected by data below the LOD. More specifically, the estimated LOESS level in 1990 was almost the same between lower and upper estimates. However the increase in LOD, from 0.1 μ g/l during the period 1990 – 1995 to 0.5 μ g/l from 1996 onwards, might have caused considerable bias in the trend analyses and create spurious upward trends. In other words, if the LOD had been lower the result might have been an even higher reduction of cadmium inputs.

The river Glomma results show yet another apparent example of the LOD problem. Due to a decrease of LODs a spurious downward trend is to be expected. Therefore the statistically significant downward trend observed in the inputs might be due to the decrease of LODs.

Table 5.6: Lower and upper estimates of cadmium in various rivers in the first and last year of the monitoring period. Lower estimates are when LOD is zero while upper estimate are when the concentration value equals the LOD-value

LOESS Level (t Cd/yr)						
River/Site	Year	Lower Estimate	Upper Estimate			
Loire	1990	3.74	10.97			
	2006	1.02	104.78			
Rhine/Meuse (Haringvlietsluis)	1990	2.31	2.31			
	2005	1.28	1.66			
Rhine (IJsselmeer)	1990	1.45	1.42			
	2005	0.39	0.92			
Glomma	1990	0.08	0.93			
	2006	0.35	0.33			

The reason for the downward trend in concentrations in river Alta is also likely due to changes in LOD over the years. For example, in 1990 the LOD was 0.1 μ g/l while in the period 1992 – 1998 it was 0.01 μ g/l and LODs in the range 0.001 to 0.02 μ g/l in the period 1999-2003. Over the entire monitoring period considered, 91 out of 141 observations were reported at LOD. There is also a considerable change in the number of LODs. For example 22 of the in total 50 values above LOD all occurred during a three-year period (1999 – 2001). This might cause a spurious downward trend. Moreover, concentrations above LOD are generally low (close to LOD in many/most cases; Skarbøvik *et al.*, 2008). Known improvement in sewage treatment in Norway and reduced discharges from industry could certainly also play roles when trying to identify the reasons for the reduction in cadmium inputs. However, it is difficult to determine the influence of reduced discharges to the reduced inputs compared with the importance of changes in LOD over the time period 1990-2006.

The river Göta Älv has a complete concentration series of cadmium no LODs reported. The sudden increase in concentration in the series in mid 1995 (Figure 5.3) is probably due to a change in analytical chemical method (from atomic adsorption spectrometry to ICP).

The time series for the river Ölfusá was relatively short (1997-2006). Unusually high riverine inputs were estimated in 2006. This was due to two single outlier values in September and December, which were several hundred times higher than the normal values. Apart from these, all concentrations in 2005 and 2006 were at the LOD level. The water discharge during these sampling events was normal. Despite these two outliers affecting the annual riverine input value, the overall trend showed a downward non-significant trend (p <0.37), which may give raise to problems in statistical trend assessments.

The river Elbe is probably the best case to assess 'real' trends in cadmium concentrations since for the whole time series there were no (smaller than) LOD-values reported. The conclusion is that there are no obvious trends during the period 1994-2006. High concentration values in 1992 and 1993 give raise to an overall long-term trend (Figure 5.4). The reasons for the considerable decrease in concentrations from 1994 onwards are not known.



Figure 5.3: Observed cadmium concentrations in river Göta Älv, 1990-2006. Note the shift in the series in mid 1995 probably due to a change in the laboratory method



Figure 5.4: Observed cadmium concentrations in the river Elbe, 1992-2006



Figure 5.5: Total annual riverine copper inputs for Regions I and II, together with annual copper inputs from selected rivers in these Regions. Inputs from river Ölfusá were not included in the total for Region I

5.2.4 Copper (Cu)

Figure 5.5 shows the riverine inputs of copper from the rivers Elbe, Ölfusá, Meuse, Seine, Rhine, Alta, Glomma and Göta Älv during the period 1990 – 2006 (right column), and the total riverine inputs to Regions I and II (left column). The copper inputs from the rivers Garonne and Loire in Region IV were too scarce to calculate reliable total inputs. The monitoring regime varied slightly between the rivers, see also Table 5.2. The inputs were based mainly on monthly data before the results were aggregated to annual total inputs. To illustrate similarities and/or differences in input patterns between Regions and individual catchments, the total inputs for the regions are also shown together with the figures of the rivers discharging into the relevant Regions. The details of the statistical analysis are available in the Background document of Statistical Analyses.

With regard to the inputs of Cu a number of comments can be made:

- Region I:
 - For most years the rivers Ölfusá and Alta represent a relatively low portion of the total riverine input to Region I, usually about 10% or lower.
- Region II:
 - The inputs of the rivers Elbe and Rhine/Meuse tend to be the most important rivers when it comes to the riverine input of copper to Region II;

 In 1995 the inputs of copper from the rivers Rhine/Meuse are much higher than any other yearly input (456 t/yr). The second highest is the river Elbe with 186 t/yr in 2002. Both totals were most likely heavily influenced by flood events.

The problem with regard to results below the level of detection (LOD) appears to be less for Cu. Almost all results were above the reported LODs. The exceptions are the rivers Garonne and Loire where no assessment was made due to low sampling frequency (2 - 4 samples/yr) and some years with completely missing data (*cf.* Table 5.3).

The trend analyses detected two upward and three downward trends. Upward trends were detected in the rivers Göta Älv and Seine (Table 5.7). In Göta Älv, the increase in concentrations at mid-1995 (Figure 5.6) is probably due to a change in the analytical method (AAS to ICP). Thus, the upward trend is most likely a reflection of laboratory method change than a real increase in riverine inputs from river Göta Älv.

River (site)	Flow- adjustment method	Trend pattern	Trend method	Trend sign	Significant (p<0.05)
Ölfusá	L1	Non-monotonic	LOESS Level (log analysis)	Downward	No
Alta	A0	Sub-monotonic	Mann-Kendall and Theil slope (log. analysis)	Downward	Yes
Glomma	A0	Non-monotonic	LOESS Level	Downward	No
Göta Älv	A0	Sub-monotonic	LOESS Level	Upward	Yes
Elbe	L1	Non-monotonic	LOESS Level	Downward	No
Rhine (IJsselmeer)	A0	Non-monotonic	LOESS Level	Downward	Yes
Rhine (Maassluis)	No	Non-monotonic	LOESS Level	Downward	Yes
Rhine/Meuse (Haringvlietsluis)	A0	Non-monotonic	Mann-Kendall and Theil slope	Downward	No
Seine	L1	Non-monotonic	Non-monotonic LOESS Level		Yes

Table 5.7: Trend analyses summary results for copper (Cu)



Figure 5.6: Observed copper concentrations in river Göta Älv, 1990-2006. Note the shift in the series in mid 1995 probably due to a change in the laboratory method



Figure 5.7: Observed copper concentrations in the river Seine, 1990-2006. Note the scattered picture in the beginning of the 1990s and the gaps in data

In the river Seine, the upward trend in riverine inputs was estimated to be about 50% (p <0.03). The corresponding analyses of the dataset showed an increase of about 40% (p <0.06). In this context it should be noted that there is a rather scattered concentration distribution in the early 1990s, as there are some gaps in dataset (Figure 5.7). The figure also shows evidence of an upward trend that is due to an increase in the observations in the lower concentration range. More specifically, concentrations were rarely below 1.8 μ g/l after 1999, whereas this was more common in the beginning of the time series.

For the river Rhine, downward trends in riverine input of Cu could be detected. For the location IJsselmeer (Figure 5.8) this was -38% (p <0.014) and for Maassluis -27% (p <0.014); the river Alta (Figure 5.8) showed a decreasing trend as well (-87%, p <0.001).

The considerable decrease in the near pristine river Alta in Northern Norway is worth mentioning. Originally this river was included in the Norwegian RID programme to determine background concentrations and for comparative purposes with more anthropogenically impacted rivers.

In the Rhine/Meuse river at IJsselmer there has been a consistent downward pattern during the monitoring period, combined with substantially lower number of peak values after 1995. In the river Rhine at Maassluis there is a rather peculiar pattern in 2002 and 2003 with elevated concentrations. This could be explained by high-flows in 2002 and low-flows in 2003.



Figure 5.8: Observed copper concentrations in the three rivers/sites with statistically significant decline in copper riverine inputs; river Alta (upper panel) and river Rhine (IJsselmeer, middle panel; Maassluis, lower panel)

5.2.5 Lead (Pb)

Figure 5.9 shows the riverine inputs of lead from the rivers Ölfusá, Alta, Glomma, Göta Älv Elbe, Rhine/Meuse and Seine during the period 1990 – 2006, and the total riverine inputs to the Regions I and II. No lead inputs from the rivers Garonne and Loire have been made available. The monitoring regime varied slightly between the rivers. The inputs were based mainly on monthly data before the results were aggregated to annual total inputs.



Figure 5.9: Total annual riverine lead inputs for the Regions I and II, together with annual lead inputs from selected rivers in the these Regions. Inputs from the river Ölfusá were not included in the total for Region I

With regard to the inputs of Pb a number of comments can be made:

- Region I:
 - For most years the rivers Ölfusá and Alta represent a relatively low portion of lead inputs compared to the total riverine inputs to Region I, usually about 10% or lower;
 - The figures showing the total annual input of lead to Region I is based on data without Iceland;
 - The inputs from Ölfusá in 2006 (8.2 t/yr) exceed the value given for the whole region (6.7 t/yr).

- Region II:
 - The rivers Elbe and Rhine/Meuse tended to be the most important rivers when it comes to the riverine inputs of lead to Region II;
 - The inputs of lead from the rivers Rhine/Meuse in 1995 are the highest estimated annual inputs during the monitoring period (390 t). The second highest registered input was the river Elbe with 135 t in 2002. Both totals are most likely heavily influenced by flood events.

Similarly to cadmium and mercury, the trend analyses were for some rivers/sampling sites limited by concentrations below LODs (and changing LODs over time). Interpretation of results from the rivers Garonne and Loire were disregarded due to low sampling frequency (2-4 samples/yr) and some years with no data (Table 5.3).

In the river Rhine (Maassluis), 154 out of 360 results of riverine Pb were below LOD. In addition, the LOD-values varied substantially in subsequent periods: values of 0.1 μ g/l in the period 1990 – 1994, 0.9 μ g/l in 1995 and early 1996, back to 0.1 μ g/l from early 1996 to mid 1999, 0.05 μ g/l from mid 1999 to mid 2004 and back to 0.1 μ g/l from the end of 2004 onwards.

Also the results from the two Norwegian rivers were affected by data below the LOD, as well as altered LODs over time. Table 5.8 contains the lower and the upper limit of the LOESS level in 1990 and 2006 for these two Norwegian rivers. They clearly indicate that a reliable trend assessment is difficult to perform.

Table 5.8: Lower and upper estimates of lead inputs from two Norwegian rivers in 1990 and 2006. Lower estimates are when LOD=0, while upper estimates are when the concentration value is set to the LOD-value

LOESS Level (t Pb/yr)						
River	Year	ear Lower Estimate Upper Estimate				
Alta	1990	0.02	0.54			
	2006	0.06	0.06			
Glomma	1990	5.27	10.31			
	2006	6.62	6.61			

With regard to the remaining six rivers/sampling sites, they have more or less complete concentration series of lead without none or relatively few LODs reported. The trend test detected a statistically upward trend in one river and a downward trend in two rivers. The only upward trend was detected in the river Göta Älv, like for Cu for the period 1990 – 2006. The sharp increase in concentrations in the series in mid-1995 (Figure 5.10) is probably due to a change in analytical chemical method.

The two rivers/sites with downward trends were river Rhine (IJsselmeer), a decline of about 60% (p < 0.02) and the river Seine with a decline of about 90% (p < 0.006). The observed concentrations are shown in (Figure 5.11).

River/Site	Flow- adjustment method	Trend pattern	Trend method	Trend sign	Significant (p<0.05)
Ölfusá	No	Non-monotonic	Mann-Kendall and Theil slope	Upward	No
Göta Älv	A0	Non-monotonic	LOESS Level	Upward	Yes
Glomma	No	Non-monotonic	Mann-Kendall and Theil slope	Downward	No
Elbe	L1	Non-monotonic	LOESS Level	Downward	No
Rhine (IJsselmeer)	A0	Non-monotonic	LOESS Level	Downward	Yes
Rhine/Meuse (Haringvlietsluis)	A0	Non-monotonic	Mann-Kendall and Theil slope (log. analysis)	Downward	No
Seine	No	Non-monotonic	Mann-Kendall and Theil slope (log. analysis)	Downward	Yes

Table 5.9: Trend analyses summary results for lead (Pb)



Figure 5.10: Observed lead concentrations in river Göta Älv. Note the shift in the series in mid 1995 probably due to a change in laboratory method



Figure 5.11: Observed lead concentrations in the two rivers with statistically significant decline in lead riverine inputs; river Seine (upper panel) and river Rhine (IJsselmeer, lower panel)

5.2.6 Mercury (Hg)

Figure 5.12 shows the riverine inputs of mercury from the rivers Ölfusá, Alta, Glomma, Göta Älv, Elbe, Rhine/Meuse and Seine for the period 1990 – 2006, and total riverine inputs to Regions I and II. Mercury inputs from the river Loire were available (not displayed), but there were no data available for the river Garonne. The monitoring strategies varied slightly between rivers (see also Table 5.2). The inputs were based mainly on monthly data before the results were aggregated to annual total inputs.



Figure 5.12: Total annual riverine mercury inputs for Regions I and II, together with annual mercury inputs from selected rivers in these Regions. Inputs from the river Ölfusá are not included in the total for Region I

With regard to the inputs of Pb a number of comments can be made:

- Region I:
 - For most years the two rivers contributed a relatively low part of the total riverine inputs of Hg to Region I, usually about 10% or less.
- Region II:
 - In most years, the Elbe, of the rivers examined, contributed the highest inputs of mercury, but also the rivers Rhine/Meuse contributed with considerable inputs to Region II;
 - \circ The Hg inputs of Glomma in 1998, with 2.7 t/yr out of 9.9 tonnes in total for Region II, are due to two measurements of extremely high Hg concentrations (0.915 and 0.73 μ g/l, respectively). The reason for these sudden extreme values is not known.

Six rivers/sampling sites of the rivers Ölfusá, Alta, Glomma, Rhine (location Maassluis), Seine and Loire were disregarded due to several reasons, including:

- In river Ölfusá only 74 out of 79 observations were not below LOD;
- In the rivers Alta and Glomma almost 50% of the observations were below LOD. In addition, the LOD has changed during the monitoring period. Moreover, there was a shift in the analytical chemical laboratory during 1995 giving raise to substantially higher concentration

values (OSPAR, 2008c);

- At the Rhine (Maassluis) 220 out of 335 observations were below LOD.
- In the river Seine almost 60% of the mercury concentrations were reported below LOD; in addition, there were to gaps in the series and differences in sampling frequency between years occurred;
- A considerable part of the data in the river Loire was below the LOD. As the LOD is almost always constant over the whole period (0.1 µg/l), no spurious trends are to be expected. However, the level of the riverine inputs may be affected, as well as suppression of any variability in the true concentrations. It should also be kept in mind that the Loire data series represent the combination of two different sampling sites with no overlap in sampling period (station Possonnière for 1990 2001 and station Monjean for 2002 2006).

Trend assessments of concentrations and inputs of mercury could only be undertaken for the rivers Göta Älv, Elbe and Rhine/Meuse. All three showed a significant downward trend (p < 0.05), Table 5.10. Considerable differences in LODs were reported by Contracting Parties, within one country over time as well as between countries (Annex II).

River (Site)	Flow- adjustment method	Trend pattern	Trend method	Trend sign	Significant (p <0.05)	Level of decrease (%)	p-value
Göta Älv	A0	Non- monotonic	LOESS Level	Downward	Yes	58	p <10⁻⁵
Elbe	No	Sub- monotonic	LOESS Level	Downward	Yes	65	p <10⁻⁵
Rhine (IJsselmeer)	No	Non- monotonic	LOESS Level	Downward	Yes	68	p<0.001
Rhine/Meuse (Haringvlietsluis)	A0	Non- monotonic	Mann- Kendall and Theil slope (log. analysis)	Downward	Yes	71	p<0.005

Table 5.10: Trend analyses summary results for mercury (Hg)

The concentration of mercury showed a very variable pattern at the beginning of the monitoring period for all rivers (Figure 5.13). Peak-values are sometimes 30 times above the 'normal' concentrations for all four rivers.



Figure 5.13: Observed mercury concentrations in the four rivers (sites) where a significant decrease in riverine inputs of mercury has been determined

5.2.7 Total nitrogen (total-N)

Figure 5.14 shows the riverine inputs of total nitrogen (total-N) from the rivers Ölfusá, Alta, Glomma, Göta Älv, Elbe, Rhine/Meuse, Seine, and Loire for the period 1990 – 2006, and the total riverine inputs of nitrogen to the Regions I, II and IV. Total-N inputs were not available for river Garonne. The monitoring strategies vary slightly between the rivers, see also Table 5.2. The inputs were based mainly on monthly data before the results were aggregated to annual total inputs.



Figure 5.14: Total annual riverine inputs of nitrogen for Regions I, II and IV, together with annual nitrogen inputs from selected rivers in these Regions. Inputs from the river Ölfusá were not included in the total for Region I

When studying the various charts it is important to realise that the scale of the y-axis of the 'Region' and the 'Catchment' figures are in some case (very) different. With regard to tot-N some comments can be made:

- Region I:
 - For most years the inputs of tot-N from the rivers Ölfusá and Alta represent a relatively small proportion of the total riverine inputs of nitrogen to Region I, usually less than 10%.
- Region II:

- For most years the rivers Elbe, Rhine and Seine are the largest contributors of the examined rivers of tot-N inputs to Region II;
- 1994 and 1995 were wet years in northern Europe, and the high inputs of total-N in these years are most likely heavily influenced by flood events;

Compared to the trace metals, the total nitrogen input datasets were much more complete. The following observations can be made:

- The river Ölfusá dataset suffered from low sampling frequencies, especially for 2001 (2 samples), 2002, 2004 and 2006 each 4 samples; no data were reported for 2003. The length of the time series (1997 2006) is at the lowest possible (10 years) for a sound statistical analyses;
- For the river Seine no results of total-N were reported for the period 1998 to August 2001;
- For the river Loire, no data were made available for the entire years 2004 and 2005;
- No data on total-N inputs were reported for the river Garonne.

Out of the ten datasets tested, four significant downward trends (p<0.05) in the flow-adjusted riverine inputs of tot-N were detected.

The most 'obvious' trends were found for the rivers Elbe and Rhine (IJsselmeer) with a decrease in level of about 40-45% (Table 5.11). The decrease in input for the two other rivers with significant trends, namely the river Rhine (Maassluis) and the Rhine/Meuse river (Haringvlietsluis), was just under 30%. The downward trends for these four rivers are best illustrated with the concentration time series in Figure 5.15. Noticeable is the 'nice' seasonal pattern at all four sites. Particularly spectacular was the extremely high intra-annual variability at the Rhine station IJsselmeer with for all years a consistent close to zero nitrogen concentrations in the summer period, an indication of high biological uptake of nitrogen and/or denitrification. Such a phenomenon of almost absent summer tot-N concentrations is rarely recorded in large rivers, but the IJsselmeer, although being considered as a branch of the river Rhine, is a productive lake.

As has been indicated in Table 5.11, that nearly significant downward trends were observed for the rivers Alta (p < 0.08) and Loire (p < 0.10). One statistically significant upward trend (p < 0.03) of more than 90% in the flow-adjusted riverine inputs from the river Ölfusá was detected. For the river Glomma the estimated 20% increase was nearly significant (p < 0.08).

Table 5.11: Percentage change in level (over period 1990 – 2006) for flow-adjusted riverine inputs of total nitrogen (tot-N) and p-values for the trend (p < 0.05 in bold). The river Ölfusá was included in the overview for the sake of completeness, but should normally have been discarded for statistical reasons, see comments above

River (site)	% change in level	p-value
Ölfusá	+93	<0.02
Alta	-22	<0.09
Glomma	+21	<0.08
Göta Älv	-4	<0.41
Elbe	-45	<10 ⁻⁵
Rhine (IJsselmeer)	-39	<0.02
Rhine (Maassluis)	-27	<0.007
Rhine/Meuse (Haringvlietsluis)	-27	<0.003
Seine	-10	<0.31
Loire	-25	<0.10
Garonne	-	-



Figure 5.15: Observed concentrations of total nitrogen in the four rivers/sites with statistically significant decline in riverine inputs of total nitrogen; river Elbe (upper left panel), river Rhine at IJsselmeer (upper right panel), river Rhine at Maassluis (lower left panel) and rivers Rhine/Meuse at Haringvlietsluis (lower right panel)

5.2.8 Total phosphorus (total-P)

Figure 5.16 shows the riverine inputs of total-P from the rivers Ölfusá, Alta, Glomma, Göta Älv, Elbe, Rhine/Meuse, Seine, Loire and Garonne in the period 1990 – 2006, and the total riverine inputs to Regions I, II and IV. The monitoring strategies varied slightly between the rivers (see also Table 5.2). The inputs were based mainly on monthly data before the results were aggregated to annual total inputs.



Figure 5.16: Total annual riverine inputs of phosphorus (tot-P) for the Regions I, II and IV, together with annual inputs from selected rivers in these Regions. Phosphorus inputs from the river Ölfusá were not included in the total for Region I

Apart from the remark that the scales of the y-axes often differ, a few comments can be made:

- Region I:
 - The rivers Ölfusá and Alta have, in comparison to the other determinants, a larger contribution to the total input in Region I. Especially the river Ölfusá had a relatively high proportion of phosphorus inputs to the total inputs, especially in the latter years of the monitoring period (about 20%).
- Region II:
 - For most years of the monitoring period the rivers Rhine/Meuse and the Seine showed the highest contributions to the total annual phosphorus inputs;
 - 1994 and 1995 were wet years in northern Europe, and the high phosphorous inputs by the Rhine/Meuse in these years may be partially attributed to flood events.

Similar to nitrogen the possible bias in interpretation introduced by values at or below the LOD is of less concern.

The following observations can be made:

- In the river Ölfusá there was a change in laboratory method in 1999;
- The high riverine inputs and concentrations in 1999-2002 for the rivers Glomma and Alta could possibly be related to the fact that there was a change of laboratory in the period 1999 2004. The high riverine inputs in 1995 and 1996 for the river Glomma are primarily an effect of increased sampling frequency during an extreme flood situation in 1995. This significantly increased the probability to hit concentration peaks since P in this river is bound to particles, especially in flooding situations;
- For the river Loire, a very high LOD was used (0.1 mg/l) prior to 2005, with a number of observations at LOD. In 2005 and 2006 the LOD was lowered to 0.05 mg/l. However, there were many observations close to LOD (in the range of 0.05-0.1 mg/l) in the samples from the river Loire in 2005. The merging of data from two sampling sites with non-overlapping time periods (1990 2001 and 2002 2006) could also be a source of uncertainty; a step-trend could, however, not be observed between 2001 and 2002.
- For the river Garonne, it should be noted that only 6 samples were collected in 1997 of which 4 were reported at the LOD (0.1 mg/l). Low sampling frequency was also reported for the period 1998 – 2000. Since 2001, the sampling frequency was increased to monthly intervals and at the same time the LOD was lowered to 0.05 mg/l (see Annex II). The length of the time period used for the trend assessment (1997 – 2006) is at a minimum requirement for a sound statistical test. This implies that the trend results for the Garonne should be interpreted with caution.

Overall, results from four (out of 11) rivers/locations showed statistically significant downward trends (p <0.05) in the flow-adjusted riverine inputs of total phosphorus (Table 5.12). The most obvious trends were observed for the rivers Seine and Loire, with a decrease of about 80 and 75%, respectively (for the monitoring period). The decline in the rivers Elbe and Rhine (Maassluis) was about 45%. No other significant downward trends could be detected. However, for the Göta Älv a nearly significant upward trend was observed (p <0.08). This seems to be related to a higher interannual variability and peak-concentrations in the three last years of the considered period (2004 – 2006). The same phenomenon was noted for the tot-N data.

The downward trends for the four rivers where a significant trend was observed are best illustrated with the concentration time series (Figure 5.17).

Table 5.12: Percentage change in level (over the entire monitoring period) for the flow-adjusted riverine inputs of total phosphorus, and p-values for the trend; significant trends (p<0.05) are in bold). For the sake of completeness the rivers Glomma and Alta are also included in the table although, for reasons of change in laboratory and LOD, the datasets should have been discarded

River (site)	% change in level	p-value
Ölfusá	-8	<0.72
Alta	-41	<0.58
Glomma	-20	<0.49
Göta Älv	+25	<0.08
Elbe	-43	<10 ⁻⁵
Rhine (IJsselmeer)	-23	<0.23
Rhine (Maassluis)	-46	<0.02
Rhine/Meuse (Haringvlietsluis)	-35	<0.14
Seine	-83	<10 ⁻⁵
Loire	-74	<0.001
Garonne	-59	<0.21



Figure 5.17: Observed concentrations of total phosphorus at the four rivers/sites with a significant decline in riverine inputs of total phosphorus. The missing values for Loire in 2005 and 2006 were interpolated

6. Glossary

AAS	Atomic absorption spectrometry (chemical analytical method for trace metals)
A0	Flow-adjustment method for annual load data which takes into account the ratio between the actual annual run-off and the long-term annual run-off.
Catchment	The whole of an area having one common outlet for its drainage water. A catchment area could be subdivided into a monitored and unmonitored area, depending on where the monitoring point is located.
Cd	Cadmium
Cu	Copper
Direct discharges	A mass of a determinant discharged to the maritime area from point sources (sewage effluents, industrial effluents or other) per unit of time at a point on a coast or to an estuary downstream of the point at which the riverine estimate of inputs is made.
Flow-adjustment	A statistical technique that takes water discharge (flow, Q) data into account in trend assessments of riverine inputs (JAMP Guidance on Input Trend Assessment and the Adjustment of Loads (OSPAR agreement 2003-9)
Heavy metals	Refers to the four metals whose direct discharges and riverine inputs were studied in this assessment namely: cadmium, copper, mercury and lead.
Hg	Mercury
ICP	Inductively coupled plasma (chemical analytical method for trace metals)
INPUT	OSPAR Working Group on Inputs to the Marine Environment
JAMP	OSPAR Joint Assessment and Monitoring Programme (OSPAR agreement 2003-22)
JAMP Guidance	JAMP Guidance on Input Trend Assessment and the Adjustment of Loads (OSPAR agreement 2003-9)
LOD	Limit of Detection is, according to the definitions (IUPAC, IS/TR 13530), "the limit of detection (LOD) is, in broad terms, the smallest amount or concentration of an analyte in the test sample that can be reliably distinguished from zero".
LOQ	The limit of quantification (LOQ) is the smallest amount or concentration of analyte in the test sample which can be determined with a fixed precision, <i>e.g.</i> relative standard deviation $s_{rel} = 33.3$ %. This means in other words, that a substance can only be correctly qualified from LODs, while it only can be quantified from LOQs.
LOESS level	Describes a trend assessment technique based on the underlying non-linear trend. LOESS specifically denotes a method that is (somewhat) more descriptively known as locally weighted polynomial regression.
Main river	For the purposes of the RID Study, a river to be monitored at least once a month (12 datasets) every year in accordance with the objectives of the Comprehensive Study. Main rivers should be major load bearing rivers.
Mann-Kendall (MK)	A non-parametric trend assessment technique for detecting monotonic trends.

Monitored area	The catchment upstream of the river monitoring point.
Nutrients	For the purposes of this assessment total Nitrogen and total Phosphorus
OSPAR	OSPAR Commission (established by the Convention for the Protection of the Marine Environment of the North-East Atlantic)
Pb	Lead
p-value	The p-value is associated with a test statistic. It is "the probability, if the test statistics really were distributed as it would be under the null hypothesis, of observing a test statistic (as extreme as, or more extreme than) the one actually observed."
QA	Quality Assurance
RID (Study)	Comprehensive Study of Riverine Inputs and Direct Discharges (OSPAR agreement 1998-5)
Riverine inputs	A mass of a determinant carried to the maritime area by a watercourse (natural river or man-made watercourse) per unit of time
RTrend	Software programme developed by <i>quo data</i> for the statistical analysis of riverine inputs.
Theil slope	A non-parametric, outlier-resistant estimation method for linear trends.
Total inputs	Sum of direct discharges and riverine inputs.
Total-N	Total nitrogen
Total-P	Total phosphorus
Tributary river	For the purpose of the RID Study, a river with separate catchment from a main river and with an outlet directly to the maritime area or to a main river downstream of a river monitoring point. A tributary river should be a minor load bearing river and can be sampled at a frequency determined by each Contracting Party.
Unmonitored area	For the purpose of the RID Study, defined as any sub-catchment(s) located downstream the riverine monitoring points within catchments and any areas between catchments. The unmonitored areas may contribute to the losses/discharges of substances downstream of the monitoring point or directly to the sea (OSPAR maritime area).
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Annex I Flow-adjustment and Trend Assessment Procedure

Flow-adjustment

Decision on adjustment was taken in accordance with the JAMP Guidance Document (OSPAR agreement 2003-9), with the following steps:

Check whether there is a significant flow-dependence of the load. If not, omit adjustment and use reference method CM2 ("OSPAR formula")

Otherwise check whether there is a significant flow-dependence of the concentration. If not, use adjustment method A0.

Otherwise check adjustment method L1. In the case that with this method both increases sensitivity in terms of trend detectability and power, and provides also a reasonable fit of the data (residual analysis), use adjustment method L1.

Otherwise omit adjustment and use the reference method CM2.

Trend assessment

The trend assessment was based on the annual data obtained in the adjustment step, either based on CM2, A0 or L1. For the assessment of the trend the LOESS method was preferred. However, in case of high variability of the data and especially in case of occurrence of outliers, the Theil slope method was applied. Calculation was carried out on a log scale in case of very high variability of the loads.

The statistical methodology is described in detail in the JAMP Guidance (OSPAR agreement 2003-9). All analyses have been carried out with RTrend version 3.3.0.0. This programme is an upgraded version of the programme used for the 2005 interim assessment.

Cd (µg/I)						
	Recommended	Minimum	Maximum	Difference	Region	
BE	0.01	0.06	5	4.94	Ш	
DE	0.01	0.01	0.05	0.04	П	
DK	0.01	0.004	0.005	0.001	П	
ES	0.01	0.001	20	19.999	IV	
FR ¹)	0.01	0.5	2	1.5	П	
FR	0.01	0.005	10	9.995	IV	
IE	0.01	0.05	0.1	0.05	Ш	
IS	0.01	NI	NI	NI	I	
NL	0.01	0.01	0.2	0.19	П	
NO	0.01	0.001	0.1	0.099	I, II	
PT	0.01	0.01	0.03	0.02	IV	
SE	0.01	0.003	0.003	0	II	
UK	0.01	0.02	0.02	0	II, III	

Annex II Limits of Detection (LOD) reported by Contracting Parties

Cu (µg/l)					
	Recommended	Minimum	Maximum	Difference	Region
BE	0.1	0.06	10	9.94	II
DE	0.1	0.1	1	0.9	II
DK	0.1	0.04	0.04	0	II
ES	0.1	0.005	20	19.995	IV
FR ¹)	0.1	2	2	0	II, IV
FR	0.1	1	30	29	II, IV
IE	0.1	0.5	1	0.5	
IS	0.1	NI	NI	NI	I
NL	0.1	0.1	5	4.9	II
NO	0.1	0.01	0.5	0.49	1, II
PT	0.1	0.01	0.34	0.33	IV
SE	0.1	0.004	0.04	0.036	II
UK	0.1	0.27	0.27	0	11, 111

Pb (μg/l)						
	Recommended	Minimum	Maximum	Difference	Region	
BE	0.01	0.35	11	10.65	II	
DE	0.01	0.1	0.5	0.4	Ш	
DK	0.01	0.02	0.025	0.01	II	
ES	0.01	0.01	50	49.99	IV	
FR ¹)	0.01	4	5	1	II	
FR	0.01	0.01	10	9.99	IV	
IE	0.01	0.5	1	0.5	111	
IS	0.01	NI	NI	NI	I	
NL	0.01	0.1	5	4.9	Ш	
NO	0.01	0.005	0.5	0.495	I, II	
PT	0.01	0.1	0.41	0.31	IV	
SE	0.01	0.02	0.02	0	II	
UK	0.01	0.03	0.03	0	11, 111	

Hg (µg/l)						
	Recommended	Minimum	Maximum	Difference	Region	
BE	0.005	0.003	0.2	0.197	II	
DE	0.005	0.001	0.01	0.009	П	
DK	0.005	0.0005	0.0005	0	II	
ES	0.005	0.001	6.8	6.079	IV	
FR ¹)	0.005	0.05	0.5	0.45	П	
FR	0.005	0.005	1	0.995	IV	
IE	0.005	0.5	1	0.5	Ш	
IS	0.01	NI	NI	NI	I	
NL	0.005	0.001	0.02	0.019	П	
NO	0.005	0.001	0.005	0.004	I, II	
PT	0.005	0.05	0.05	0	IV	
SE	0.005	0.0001	0.0001	0	II	
UK	0.005	0.02	0.02	0	11, 111	

Tot-N (mg/l)						
	Recommended	Minimum	Maximum	Difference	Region	
BE	0.05	NI	NI	NI	II	
DE	0.05	0.05	1	0.95	Ш	
DK	0.05	0.06	0.06	0	П	
ES	0.05	0.02	1.455	1.435	IV	
FR ¹)	0.05	1.44	14.29	12.85	II	
FR	0.05	1.027	11.756	10.729	IV	
IE	0.05	0.02	0.02	0	Ш	
IS	0.05	NI	NI	NI	I	
NL	0.05	0.1	0.1	0	Ш	
NO	0.05	0.001	0.01	0.009	I, II	
PT	0.05	NI	NI	NI	IV	
SE	0.05	0.05	0.05	0	II	
UK	0.05	0.002	0.021	0.019	II, III	

tot-P (mg/l)						
	Recommended	Minimum	Maximum	Difference	Region	
BE	0.005	0.1	1	0.9	Ш	
DE	0.005	0.05	1	0.95	П	
DK	0.005	0.01	0.01	0	П	
ES	0.005	0.01	0.1	0.09	IV	
FR ¹)	0.005	0.02	0.1	0.08	II	
FR	0.005	0.02	0.105	0.085	IV	
IE	0.005	0.005	0.005	0	Ш	
IS	0.005	NI	NI	NI	I	
NL	0.005	0.01	0.1	0.09	П	
NO	0.005	0.001	0.01	0.009	I, II	
PT	0.005	NI	NI	NI	IV	
SE ¹)	0.005	0.005	0.005	0	II	
UK	0.005	0.003	0.03	0.027	11, 111	

¹) For the year 2006



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OSPAR's vision is of a clean, healthy and biologically diverse North-East Atlantic used sustainably

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