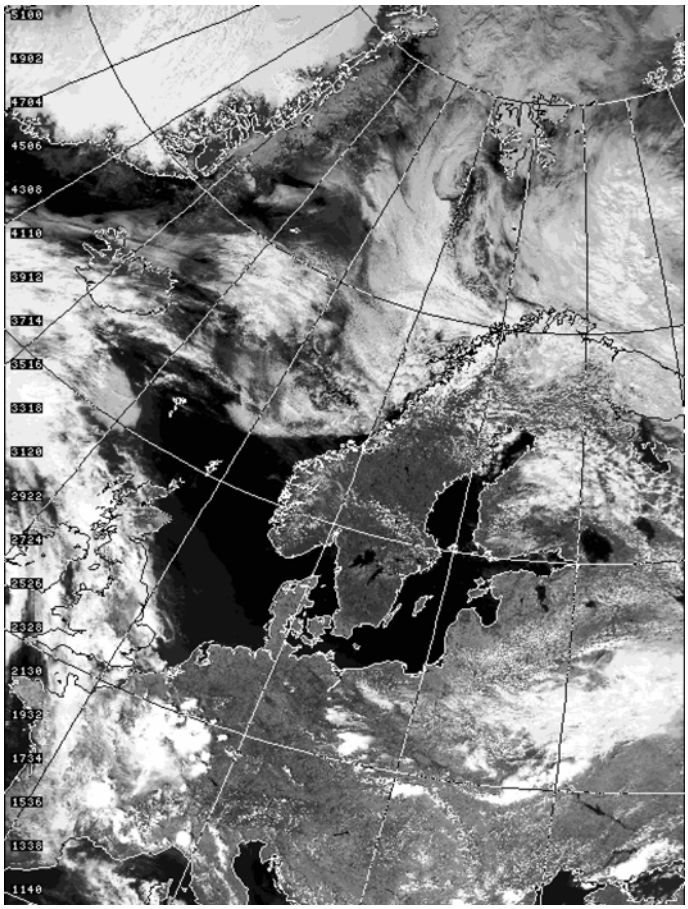
Assessment and Monitoring Series

Assessment of trends in atmospheric concentration and deposition of hazardous pollutants to the OSPAR maritime area

Evaluation of the CAMP network



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OSPAR Commission
2005

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.

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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Front page picture: satellite image of the north-eastern part of the OSPAR region. This image has been made on 11 July 1995, roughly halfway the period 1989-2002 studied in this report.

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Executive Summary

AIRBORNE INPUTS OF CONTAMINANTS TO THE SEA ARE DOWN, BUT STILL IMPORTANT

A scientific assessment of atmospheric monitoring data shows that generally over the past 15 years there have been statistically significant downward trends in direct atmospheric inputs of all the contaminants monitored. Direct atmospheric inputs of contaminants remain important, since they account for up to half the total inputs from land of some heavy metals and up to about a third of those of nitrogen. Shipping also remains a significant source of inputs of nitrogen.

1. Through the Comprehensive Atmospheric Monitoring Programme (CAMP), OSPAR has been monitoring airborne inputs of contaminants to the sea since 1987. The monitoring has involved 49 stations in the 12 OSPAR coastal states[1]. It has covered nitrogen compounds (which are important because they increase the nutrients in the sea and can thus contribute to eutrophication), some heavy metals and lindane (a pesticide). The aim is to estimate the direct input from the air to the marine environment (indirect atmospheric inputs through run-off are covered by the Riverine and Direct inputs study (RID). Regrettably, there have been, and continue to be, gaps in delivering the agreed coverage of monitoring. As a result, there is a marked lack of observations for much of the coastline.

2. Monitoring is both of concentrations in rain or other precipitation and of the level of substances found in air, either attached to particles suspended in air (aerosols) or in gaseous form. There are problems in assessing time-series, since data of both kinds is heavily affected by the weather. Air quality varies considerably with air temperature, humidity and wind direction. Deposition from the air varies with precipitation amount and frequency and the source regions of the precipitation. All this variability makes it difficult to establish statistically significant trends. A further complication for heavy metals is the large proportion of samples where the concentrations are below the level which can be detected.

3. Nevertheless, ***heavy metals*** (arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc) show generally decreasing trends both in precipitation and on aerosols. The assessment examined 108 time-series for concentrations in precipitation. 38 time-series showed statistically significant trends (though this dropped to 14 when normalised by the annual amount of precipitation). All but one of the trends were downward. 60 times series were examined for concentrations on aerosols. 29 time-series showed statistically significant trends. Again, all except one were downward. The two upward trends (at different locations) were for zinc.

4. For ***nitrogen,*** time-series for concentrations of nitrates and ammonia in precipitation were examined for eleven stations in seven countries[2]. Deposition of total amounts of nitrogen was lower at all stations in 2001 than in 1990. Clear trends could not be detected because of the large inter-annual variability. However, when the data was adjusted for interannual rainfall and wind-sector variations, statistically significant downward trends were found in eight time-series. There were no upward trends. For aerosols, time-series of concentrations of ammonia and nitrate were examined for seven stations in six countries[3] and for concentrations of nitrous oxide for four stations in four countries[4]. Statistically significant trends could be seen at all stations before and after normalisation. With one exception, trends in the time-series at all stations were downwards. The one exception was ammonia at Spitsbergen (Norway), which was upwards.

5. For ***lindane*** (γ-hexachlorocyclohexane (HCH)), the problem of concentrations (if any) in samples being below the detection limit is particularly significant. Suitable time-series are only available for stations in Iceland, Norway (for precipitation and aerosols) and Sweden (aerosols only). Statistically significant downward trends are present in five of the six time-series. This is consistent with other work in the Netherlands, and is to be expected given the increasing controls on the use of the substance.

6. Estimates have been made of the total amounts of contaminants directly deposited from the air into Arctic Waters, the Celtic Seas (nitrogen) and the North Sea (nitrogen and the heavy metals monitored).

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1 Belgium, Denmark, France, Germany, Iceland, Ireland, the Netherlands, Norway, Portugal, Spain, Sweden and the United Kingdom. Finland, Luxembourg and Switzerland do not participate.

2 Suitable time-series were not available for Belgium, Denmark, Germany, Iceland and Ireland.

3 Suitable time-series were not available in Belgium, France, Germany, Iceland, the Netherlands and Portugal.

4 Suitable time-series were not available in Belgium, Denmark, France, Germany, Iceland, Ireland, Portugal and the United Kingdom.

Some comparisons have been made with the results of the OSPAR Comprehensive Study of Riverine and Direct Inputs (RID)[5]

7. For ***heavy metals***, in the main body of the North Sea, the atmospheric deposition of cadmium and lead is estimated to be roughly of the same magnitude as the inputs from riverine inputs and direct discharges. There are statistically significant downward trends in the total amounts of copper and lead estimated to have been deposited. With one partial exception, the other heavy metals monitored also show downward trends in the amounts estimated to have been deposited, but these are not statistically significant. The partial exception is zinc, where the upward trend appears to be due to data from one station in Belgium.

8. For ***nitrogen***,

   a. in Arctic Waters, atmospheric deposition is predominant (about six to ten times the riverine inputs and direct discharges). Atmospheric deposition is, however, estimated to have reduced by about 25% between 1990 and 2001. Riverine inputs (being largely natural) have remained more or less constant. Direct inputs (although even now only about 5% of the total) have increased by about 175%;

   b. in the Celtic Seas, atmospheric deposition of nitrogen is estimated to provide about one-third of all inputs of nitrogen. There are no significant trends in inputs by any route;

   c. in the Greater North Sea, atmospheric deposition of nitrogen is also estimated overall to provide about one-third of all inputs of nitrogen. However, although (after normalisation for flow rates) riverine and direct inputs have reduced significantly between 1990 and 2001 (by about 10% and 30% respectively), there has been no similar reduction in the total amount of nitrogen deposited from the atmosphere.

9. Comparisons have been made between the CAMP deposition estimates for nitrogen and the results of modelling carried out under a co-operation programme between OSPAR and EMEP[6]. The CAMP data in general shows smaller reductions in deposition than the modelling results, but is generally consistent with them. The modelling results emphasise the importance of the inputs of nitrogen from shipping – the estimates of total inputs to all OSPAR regions from shipping are larger than the individual inputs of all but the largest OSPAR countries.

10. In conclusion, the assessment finds that it is time to review the CAMP system – a review which OSPAR is now putting in hand.

11. A summary of the trends determined at each CAMP station is given in the attached summary table.

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5 An assessment of the results of this study is being published at the same time.

6 EMEP is a co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air pollutants in Europe under the UN ECE Convention on the Long-Range Transport of Atmospheric Pollution.

Récapitulatif

LES APPORTS ATMOSPHERIQUES DE CONTAMINANTS A LA MER SONT EN BAISSE TOUT EN RESTANT IMPORTANTS

Une évaluation scientifique des données de surveillance de l'atmosphère met en évidence, d'une manière générale et pendant les 15 dernières années, des tendances statistiquement significatives à la baisse des apports atmosphériques directs de tous les contaminants contrôlés. Les apports atmosphériques directs restent importants, car ils peuvent s'élever à la moitié des apports totaux d'origine terrestre de certains métaux lourds et à environ un tiers des apports d'azote. Le transport maritime reste une source importante d'apport d'azote.

1. Grâce au Programme exhaustif de surveillance de l'atmosphère (CAMP), OSPAR a surveillé les apports atmosphériques de contaminants à la mer depuis 1987. La surveillance a été effectuée à 49 stations implantées dans les 12 Etats côtiers d'OSPAR[7]. Elle a porté sur les composés d'azote (qui sont importants car ils augmentent la quantité de nutriments en mer et peuvent ainsi contribuer à l'eutrophisation), sur certains métaux lourds et sur le lindane (un pesticide). Le but est d'estimer l'apport atmosphérique direct au milieu marin (les apports atmosphériques directs par ruissellement font l'objet de l'étude des apports fluviaux et directs (RID). Malheureusement, il y a eu, et il subsiste, des lacunes dans la réalisation de la couverture qui avait été convenue pour la surveillance, en conséquence de quoi un net manque d'observations est constaté sur une grande partie du littoral.

2. La surveillance porte tant sur les teneurs dans la pluie ou autres précipitations que sur le niveau des substances décelées dans l'atmosphère, qu'elles soient fixées sur des particules en suspension dans l'air (aérosols) ou qu'elles se présentent sous forme gazeuse. L'évaluation des séries chronologiques pose des problèmes, car les données des deux types subissent fortement l'influence du climat. La qualité de l'air varie considérablement en fonction de la température de l'air, de l'humidité et de la direction du vent. Les retombées atmosphériques varient selon la quantité des précipitations et leur fréquence, ainsi que selon les régions à l'origine des précipitations. Dans son ensemble, cette variabilité fait qu'il est difficile de déterminer des tendances statistiquement significatives. La forte proportion des échantillons dans lesquels les teneurs sont inférieures au niveau qui peut être décelé complique plus encore la situation dans le cas des métaux lourds.

3. Néanmoins, les ***métaux lourds*** (arsenic, cadmium, chrome, cuivre, mercure, nickel, plomb et zinc) présentent généralement des tendances à la baisse aussi bien dans les précipitations que dans les aérosols. Dans l'évaluation, l'on a étudié 108 séries chronologiques dans les précipitations. Dans 38 séries chronologiques, l'on a constaté des tendances statistiquement significatives à la baisse (bien que ce chiffre soit retombé à 14 après normalisation en fonction de la hauteur annuelle des précipitations). A l'exception d'une seule, toutes les tendances étaient à la baisse. L'on a par ailleurs étudié 60 séries chronologiques des teneurs dans les aérosols. Dans 29 cas, les séries chronologiques présentaient des tendances significatives en baisse. Là encore, à l'exception d'une seule, toutes étaient en baisse. Les deux tendances à la hausse (qui ne se situaient pas aux mêmes emplacements géographiques) concernaient le zinc.

4. Dans le cas de ***l'azote,*** les séries chronologiques des teneurs en nitrates et en gaz ammoniacal dans les précipitations ont été étudiées à onze stations réparties dans sept pays[8]. A toutes les stations, les retombées des quantités totales d'azote se sont avérées inférieures en 2001 à ce qu'elles étaient en 1990. Il a été impossible de mettre en évidence des tendances claires en raison de la forte variabilité d'une année à l'autre. Toutefois, après correction des données en fonction des variations de la pluviosité annuelle et des secteurs éoliens, l'on a constaté des tendances statistiquement significatives à la baisse dans huit des séries chronologiques. Aucune tendance à la hausse n'a été observée. Dans le cas des aérosols, les séries chronologiques des teneurs en gaz d'ammoniac et en nitrate ont été étudiées à sept stations dans six pays[9] tandis que les teneurs en oxyde azoté ont été examinées à quatre stations dans quatre pays[10]. Des

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7 Belgique, Danemark, France, Allemagne, Islande, Irlande, Pays-Bas, Norvège, Portugal, Espagne, Suède et Royaume-Uni. La Finlande, le Luxembourg et la Suisse ne participent pas.

8 Il n'existe pas de séries chronologiques adéquates dans le cas de la Belgique, du Danemark, de l'Allemagne, de l'Islande et de l'Irlande.

9 Il n'existe pas de séries chronologiques adéquates dans le cas de la Belgique, de la France, de l'Allemagne, de l'Islande, de l'Irlande, du Portugal ni du Royaume-Uni.

10 Il n'existe pas de séries chronologiques adéquates dans le cas de la Belgique, du Danemark, de la France, de l'Allemagne, de l'Islande, de l'Irlande, du Portugal et du Royaume-Uni.

tendances statistiquement significatives ont été constatées à toutes les stations avant et après normalisation. A toutes les stations, et à une exception près, les tendances des séries chronologiques étaient à la baisse. L'exception concernait le gaz d'ammoniac dans l'île de Spitzberg (Norvège), laquelle était à la hausse.

5. Dans le cas du ***lindane*** (γ-hexachlorocyclohexane (HCH)), le problème des teneurs (s'il en est) inférieures au seuil de détection dans les échantillons est particulièrement aigu. Il n'existe de séries chronologiques adéquates que dans le cas de stations situées en Islande, en Norvège (précipitations et aérosols) et en Suède (uniquement dans les aérosols). Des tendances statistiquement significatives à la baisse sont présentes dans cinq des séries chronologiques. Ceci coïncide avec les résultats des autres recherches effectuées aux Pays-Bas, et devrait permettre de renforcer les contrôles exercés sur l'utilisation de cette substance.

6. L'on a procédé à des estimations des quantités totales de contaminants retombant directement de l'atmosphère dans les eaux arctiques, dans les mers celtes (azote) et en mer du Nord (l'azote et les métaux lourds ont été contrôlés). Quelques comparaisons ont été faites avec les résultats de l'Etude exhaustive OSPAR des apports fluviaux et des rejets directs (RID)[11].

7. En ce qui concerne les ***métaux lourds,*** dans le corps aquatique principal de la mer du Nord, l'on estime que les retombées atmosphériques de cadmium et de plomb sont à peu près du même ordre de grandeur que les apports fluviaux et les rejets directs. L'on observe des tendances statistiquement significatives à la baisse dans les estimations des retombées totales de cuivre et de plomb. A une exception partielle, les autres métaux lourds qui ont été contrôlés présentent eux aussi une tendance à la baisse dans les quantités estimées des retombées, ces tendances n'étant toutefois pas statistiquement significatives. L'exception partielle est celle du zinc, quoique la tendance à la hausse paraisse être due aux données d'une station située en Belgique.

8. Dans le cas de ***l'azote,***

a. dans les eaux arctiques, les retombées atmosphériques sont prédominantes (environ six à dix fois les apports fluviaux et les rejets directs). Cependant, l'on estime que les retombées atmosphériques ont baissé d'environ 25% entre 1990 et 2001. Les apports fluviaux (qui sont en grande partie naturels) sont restés plus ou moins constants. Les apports directs (bien que même à l'heure actuelle ils ne représentent que 5% à peu près du total) ont augmenté d'environ 175%;

b. dans les mers celtes, les retombées atmosphériques d'azote sont estimées représenter environ un tiers de l'ensemble des apports d'azote. Quelle que soit la voie de pénétration, les apports ne présentent aucune tendance significative;

c. dans la "grande" mer du Nord, l'on estime là encore que les retombées atmosphériques d'azote représentent environ un tiers de l'ensemble des apports d'azote. Toutefois, bien que (après normalisation des flux) les apports fluviaux et directs aient été significativement réduits entre 1990 et 2001 (respectivement d'environ 10 et 30%), il n'y a pas eu de baisse analogue des quantités totales des retombées atmosphériques d'azote.

9. Des comparaisons ont été faites entre les estimations CAMP des retombées d"azote et les résultats de la modélisation réalisée dans le cadre d'un programme de coopération entre OSPAR et l'EMEP[12]. D'une manière générale, les données CAMP mettent en évidence des baisses inférieures dans les retombées aux résultats de la modélisation, quoique d'une façon générale, elles coïncident avec ces résultats. Les résultats de la modélisation soulignent l'importance des apports d'azote par le transport maritime – les estimations des apports totaux du transport maritime à toutes les régions OSPAR sont supérieures aux apports individuels de tous les pays d'OSPAR excepté les plus grands pays.

10. En conclusion, il est constaté, selon l'évaluation, que le moment était venu de réformer le système CAMP – réforme qu'OSPAR a maintenant amorcée.

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[11] Une évaluation des résultats de cette étude est publiée en même temps.

[12] L'EMEP est un programme de coopération dans le domaine de la surveillance et de l'évaluation de la transmission à longue distance des polluants en Europe dans le cadre de la Convention ECE des Nations Unies sur la pollution atmosphérique transfrontière à longue distance.

11. Un récapitulatif des tendances déterminées à chaque station CAMP suit.

Summary table: Overview of trends observed at CAMP stations

				precipitation											aerosol									(a)	(b)	
code	name	lat	lon	as	cd	cr	cu	pb	hg	ni	zn	NH4	NO3	HCH	as	cd	cr	cu	pb	ni	zn	hg	Ngas	Naer		HCH
BE0002R	Oostende	51,22	2,92																							
BE0003R	Brugge	51,25	3,18									-	-													
BE0004R	Knokke	51,35	3,33		-	-		↘	+	-	+/-										-					
BE0011R	Moerkerke	51,25	3,35																				+			
BE0013R	Houtem	51,02	2,58																				+			
BE0014R	Koksijde	51,12	2,50																							
BE0090R	Bredene	51,23	2,95																							
BE3a	Brugge	51,25	3,18	station operational in the initial phase of CAMP; no data in database																						
DE0001R	Westerland	54,93	8,32	-	-	+	-	↘	-	-	+	-	-	-		↘		↘	↘	↘			-			
DK?	Husby	56,28	8,20	station operational in the initial phase of CAMP; no data in database																						
DK0008R	Anholt	56,72	11,52															↘	↘	-	↘			↘	↘	↘
DK0020R	Pedersker	55,02	14,95									+	+													
DK0031R	Ulborg	56,28	8,43	-	↘	↘	↘	↘		↘	↘	-	-		↘		↘	↘	↘	↘	↘		↗(c)	-	↘	
ES0005R	Noya	42,73	-8,92										+													
ES0008R	Niembro	43,45	-4,85																							
FR0005R	Le Hague	49,62	-1,83									↘	↘													
FR0090R	Porspoder	48,50	-4,77	-	-	-	-	↘		-	↘	↘	-													
GB0006R	Lough Navar	54,43	-7,90									-	-													
GB0013R	Yarner Wood	50,60	-3,71																							
GB0014R	High Muffles	54,33	-0,80	↘	-	-	-	-		-	-	↘	↘		-	+	+	-	↘	-	↗			-	-	
GB0016R	Glen Dye	56,97	-1,47									+	+													
GB0090R	East Ruston	52,80	1,47	↘	↘	-	↘	↘		↘	-				↗	-	+	↘	↘	↘	-					
GB0091R	Banchory	57,08	-2,53	↘	-	-	↘	↘		↘	-				↗	↘	+	+	↘	↘	↗					
GB0092R	Isle of Wight	50,67	-1,30																							
GB0093R	Staxton Wold	54,18	-0,43		-		-			-																
GB0094R	Lough Erne	54,40	-8,05																							
GB0095R	Driby	53,23	0,07									-	-													
GB0096R	Chilton	51,57	-1,32																							
IE0001R	Valentia	51,93	-10,25								↗															
IE0002R	Turlough Hill	53,03	-6,40								-	-	↘													
IS0002R	Irafoss	64,08	-21,02				↘				↘		+													

				precipitation											aerosol									(a)	(b)	
code	name	lat	lon	as	cd	cr	cu	pb	hg	ni	zn	NH4	NO3	HCH	as	cd	cr	cu	pb	ni	zn	hg	Ngas	Naer		HCH
IS0090R	Reykjavik	64,13	-21,90				-				↘		+													
IS0091R	Storhofdi	63,40	-20,28											-	-	+	↗	-	-	-	-				+	↘
NL0009R	Kollumerwaard	53,33	6,28		-		-	-			-	-	-		↘	↘			↘		↘		↘			
NL0091R	de Zilk	52,30	4,52		↘		-	-	↘			↘	-											-	-	
NL01	Vlissingen	51,45	3,58	station operational in the initial phase of CAMP; no data in database																						
NL02	Leiduin	52,33	4,58	station operational in the initial phase of CAMP; no data in database																						
NO0001R	Birkenes	58,38	8,25		↘			↘			↘	↘	↘										↘	↘	↘	
NO0039R	Karvatn	62,78	8,88		-			-			-	+	-										-	↗	-	
NO0042R	Zepellinfjell	78,90	11,88												+	⇔	-	-	-	-	-	-		↗	-	↘
NO0057R	Ny Aalesund	78,92	11,92									-	+											↗	+	
NO0099R	Lista	58,10	6,57	-	↘		-	-	-		↘	+	-	↘				+	-	⇔	+		↗	+	↗	↘
PT0003R	Viana do Castelo	41,70	-8,80				-				↘	+	-													
PT0004R	Monte Velho	38,08	-8,80								↘	-	-													
PT0010R	Angra do Heroismo	38,67	-27,22										+													
SE0002R	Rorvik	57,42	11,93						↘			↘	↘	-								+	↘	↘	+	-
SE0014R	Rao	57,40	11,92																							
SE0097R	Gaardsjoen	58,05	12,02		↘			↘			-															
SE0098R	Svartedalen	57,98	12,10	-	↘		-	-			↘	-	↘													

According to INPUT(2)95/3/2 Rev.1-E another station was operational in Iceland: Irafoss, coded as IS1

This station can not be traced in the NILU/CAMP data base

note (a) : sNH4

note (b) : sNO3

note (c) refers to NH3

	no monitoring data
	less then 5 years with valid data, not included in trend analysis
+	not significant upwards
↗	upwards, $\alpha = 0.1$
↗	upwards, $\alpha = 0.01$
+/-	opposite (but non-significant) trend in bulk an dwet only samplers
⇔	no change over time
-	not significant downwards
↘	downwards, $\alpha = 0.1$
↘	downwards, $\alpha = 0.01$

for precipitation data, significancy refers to analysis before normalisation (see report)

1. Introduction

Within the framework of the OSPAR Convention a *Comprehensive Atmospheric Monitoring Programme* (CAMP) has been established. The objectives of the CAMP are to monitor the concentrations of selected contaminants in precipitation and air and their depositions in order:

- to assess, as accuratly as relevant, the atmospheric input of the selected contaminants to the maritime area and regions thereof on an annual basis;
- to determine long-term trends in atmospheric inputs;
- to validate atmospheric transport models used for assessments of atmospheric inputs to the maritime area.

Each Contracting Party bordering the maritime area, with the exception of the EU, should have at least one monitoring station on the coast to be included in the joint monitoring programme. The station should be a so-called *background station*, that is, a station which is not directly influenced by local sources. The station should be located not more than 10 km from the coast line. At the station simultaneous measurements of the chemical composition of air and precipitation are to be performed. The programme calls for mandatory monitoring of a range of heavy metals, organic pollutants and nutrients in precipitation and air and encourage the monitoring on a voluntary basis for additional pollutants in precipitation and air. Data collected at the CAMP stations is submitted by the Contracting Parties to the database manager at NILU. Submitted data is deemed to have been quality controlled and validated by Contracting Parties.

The CAMP network has been operational since 1987 although over the years the number and location of monitoring stations has varied. Figure 1 and Table 1 give an overview of all stations which have participated in the network. Overview of information on CAMP monitoring stations is currently not available.

Table 1 has been constructed on the basis of a number of sources.

- Recent information is taken from Barrett (2004).
- Historical data is taken from the following OSPAR-documents :
  1. Proposed update of the principle of the Comprehensive Atmospheric Monitoring Programme, INPUT(2) 95/3/2 Rev.1-E;
  2. An overview of existing data on inputs of substances reported in the framework of OSPAR, INPUT(2) 95/6/2 Add.1-E;
  3. Oslo and Paris Commission (1994);
- The personal archives of the authors.

This report is based on the data available in the CAMP data base at www.nilu.no/camp. For some stations (for example, Brugge in Belgium, Leiduin in the Netherlands) concentration data has been obtained in old INPUT-documents or in the overview report on “Method 3a” (Oslo and Paris Commission, 1994) but information is either absent or only partly available in the current CAMP database. In this evaluation these stations or time series have not been included.

In this report the CAMP air quality data are evaluated with an emphasis on trends in atmospheric concentrations and deposition. The report focuses on nutrients, heavy metals and γ-HCH (lindane). Other organic pollutants (PCB, PAH) are measured on a voluntary basis (see Chapter 4) but the availability of monitoring data is too limited for an OSPAR-assessment. To assist the interpretation of the observed changes in ambient concentration, information on atmospheric emissions in Contracting Parties has been collected (Chapter 3). The atmospheric input to the OSPAR maritime area in relation to other inputs is also discussed (Chapter 7). Note that in this report only the *direct* atmospheric input, that is, the deposition directly into the maritime area is considered. Indirect contributions via rivers, caused by deposition in the catchment area followed by run-off or leaching, have not been included.

For the preparation of this assessment report, a number of background documents have been written. For more detailed information the reader is therefore referred to the following documents which are available from the OSPAR Secretariat on request:

- Wängberg I. and Munthe J. (2004) Evaluation of mercury data reported to the OSPAR-CAMP data base, 1999-2002. IVL, Göteborg.
- Wängberg I. and Munthe J. (2005) Trends in air concentrations and deposition of mercury in the marine environment of the North East Atlantic. IVL, Göteborg.

- Wängberg I. and Munthe J. (2001) Evaluation of mercury data reported to the OSPAR-CAMP data base 1989-1998. IVL, Göteborg.
- Libiseller C. and Grimvall A. (2004) Quality assessment of CAMP data for nitrogen wet and dry deposition. Linköping University, Sweden.
- Libiseller C. and Grimvall A., Hallberg L. (2005) Meteorological normalisation of time series of wet and dry deposition. Linköping University, Sweden.
- Libiseller C. and Grimvall A. (2005) Trend analysis of CAMP data regarding wet and dry nitrogen deposition. Linköping University, Sweden.
- Institute for Energy and Environment (2005) Expert examination of quality of the CAMP data on organics. INPUT 05/2/06-E.

Table 1. Stations reporting precipitation and air quality to OSPAR

code	name	latitude		longitude			Alt (a)	D(b)	period
BE0002R	Oostende	51	13	2	55	E	-99	-99	1994, 1996; coordinatess estimated
BE0003R	Brugge	51	15	3	11	E	3	-99	1987-2000
BE0004R	Knokke	51	21	3	20	E	0	1	1996-2003
BE0011R	Moerkerke	51	15	3	21	E	0	9	1990-2003
BE0013R	Houtem	51	1	2	35	E	2	12	1990-2003
BE0014R	Koksijde	51	7	2	30	E	7	1.5	2001-2003
BE0090R	Bredene	51	14	2	57	E	10	2	1990,1991, 1993
BE3a	Brugge	51	15	3	11	E	10	8	no data archived; at least 1987-1993
DE0001R	Westerland	54	56	8	19	E	12	0.09	1987-2003
DK?	Husby	56	17	8	12	E	-99	-99	no data archived; operational in 1993-? ; coord estimated
DK0008R	Anholt	56	43	11	31	E	40	0.5	1991-2003
DK0020R	Pedersker	55	1	14	57	E	-99	-99	1998-2002
DK0031R	Ulborg	56	17	8	26	E	40	20	1987-2003
ES0005R	Noya	42	44	8	55	W	685	0.4	1992-2000
ES0008R	Niembro	43	27	4	51	W	134	<1	1999-2003
FR0005R	Le Hague	49	37	1	50	W	133	-99	1987-2002
FR0090R	Porspoder	48	30	4	46	W	30	0.5	1989-2002
GB0006R	Lough Navar	54	26	7	54	W	130	28	1987-1999
GB0013R	Yarner Wood	50	36	3	43	W	119	-99	1996
GB0014R	High Muffles	54	20	0	48	W	265	22	1987-2002
GB0016R	Glen Dye	56	58	1	28	W	85	24	1989-2000
GB0090R	East Ruston	52	48	1	28	E	5	8	1987-2002
GB0091R	Banchory	57	5	2	32	W	130	26.5	1989-2002
GB0093R	Staxton Wold	54	11	0	26	W	35	10	1987-1992
GB0094R	Lough Erne	54	24	8	3	w	160	25	1989-1993
GB0095R	Driby	53	14	0	4	E	47	16	1989-2000
GB0096R	Chilton	51	34	1	19	W	109	-99	1994
GB0092R	Isle of Wight	50	40	1	18	W	50	6.4	1993-1995
IE0001R	Valentia	51	56	10	15	W	9	0	1992-2003
IE0002R	Turlough Hill	53	2	6	24	W	420	19	1992-2003
IS0002R	Irafoss	64	5	21	1	W	65	-99	1992-2001
IS0090R	Reykjavik	64	8	21	54	W	61	1	1992-2003
IS0091R	Storhofdi	63	24	20	17	W	118	0.5	1995-2003
NL0009R	Kollumerwaard	53	20	6	17	E	1	7.5	1989-2003
NL0091R	de Zilk	52	18	4	31	E	4	2.5	1996-2003
NL01	Vlissingen	51	27	3	35	E	-99	-99	no data archived, operational in 1986; coords estimated
NL02	Leiduin	52	20	4	35	E	-99	6.3	no data archived; at least operational in 1987-1992
NO0001R	Birkenes	58	23	8	15	E	190	20	1987-2003
NO0039R	Karvatn	62	47	8	53	E	210	70	1987-2003
NO0042R	Zepellinfjell	78	54	11	53	E	474	2	1989-2003
NO0057R	Ny Alesund	78	55	11	55	E	8	0.3	1987-2003
NO0099R	Lista	58	6	6	34	E	13	0.1	1988-2003
PT0003R	Viana do Castelo	41	42	8	48	W	16	4	1987-2003
PT0004R	Monte Velho	38	5	8	48	W	43	1.5	1991-2003
PT0010R	Angra do Heroismo	38	40	27	13	W	74	1	1995-2003
SE0002R	Rorvik	57	25	11	56	E	10	0.65	1987-2001
SE0014R	Rao	57	24	11	55	E	-99	-99	2002-2003
SE0097R	Gaardsjoen	58	3	12	1	E	113	12	1993-2003
SE0098R	Svartedalen	57	59	12	6	E	120	16	1987-2003

(a) station altitude (in m); -99 indicates missing information;
(b) distance from sea (in km) -99 indicates missing information.

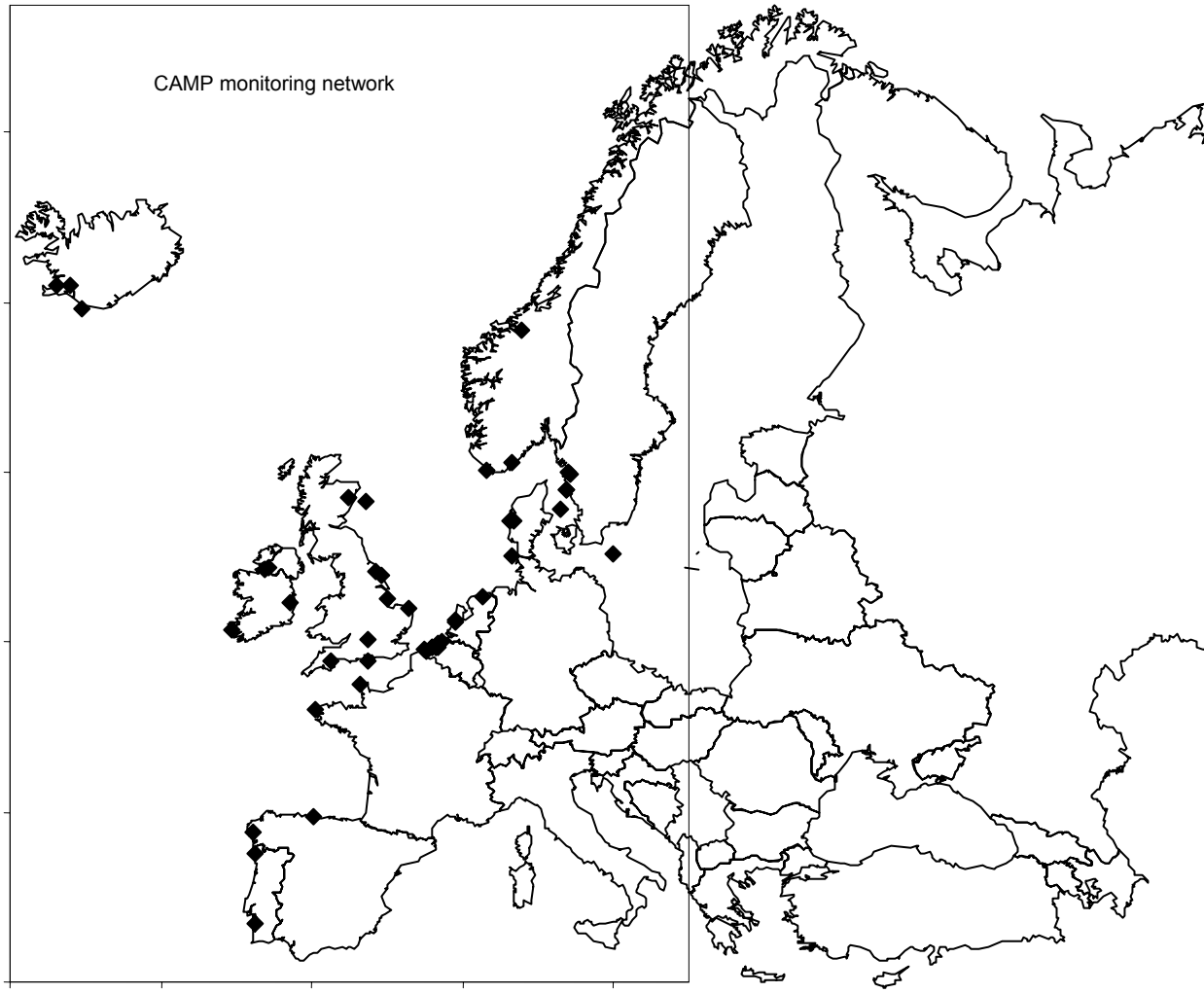


Figure 1. The OSPAR CAMP monitoring network . (Note that the stations Zeppellinfjell, Ny Alesund and Angra do Heriosmo are located outside the plotted area).

2. Atmospheric emissions

2.1. Nitrogen compounds[13]

Nitrogen oxides (NO_x) and ammonia (NH_3) emissions as officially reported under the Convention on Long Range Transboundary Air Pollution (CLRTAP) to EMEP are presented in Annex 1 for the period 1990-2001 (Vestreng, 2003).

An additional and very important source of atmospheric NO_x emissions to the OSPAR Convention Waters is the international ship traffic. Official data for 1990 give 1,266 kt NO_x (as NO_2) annual emissions from the international ship traffic on the North-East Atlantic and 648 kt NO_x (as NO_2) from the international ship traffic on the North Sea. This is the largest source of NO_x emissions in the OSPAR area of interest. Official information about this source is rather old and exists only for 1990. There is a need for ship traffic emission data for more recent years to be developed as soon as possible. According to recent estimates (EEB, 2004), nitrogen oxides emissions from the international ship traffic on the European seas increased by more than 40% from the year 1990 to 2000. Unfortunately, these recent estimates are not available to EMEP yet.

For most of the OSPAR Contracting Parties a reduction in emissions of nitrogen oxides (NO_2) has been reported for the period 1990 – 2001. Out of 18 countries relevant for OSPAR, only in four countries (Portugal, Spain, Iceland and Ireland) were annual emissions of nitrogen oxides reported to be higher in 2001 than in 1990, by 35 %, 8 %, 8 % and 6 % respectively. Moreover, in two of these countries (Iceland and Ireland) emissions in both years were low (9 and 38 kt N a^{-1}, respectively) compared to the rest of the countries.

[13] Extracted from Bartnicki and Fagerli (2004).

Germany, France and the United Kingdom were the largest emitters of nitrogen oxides among the OSPAR Contracting Parties. In these three countries, emissions were significantly lower in 2001 than in 1990 by 42 %, 26 % and 39 % respectively. The three largest contributors to nitrogen deposition in the OSPAR Convention Waters from outside the OSPAR area were Poland, Italy and the part of the Russian Federation included in the EMEP area. In these three countries, emissions in 2001 were reported to be lower than in 1990 by 37 %, 35 % and 29 %, respectively.

Annual emissions of nitrogen oxides, as a sum of emissions from all OSPAR Parties and the additional three countries identified above from outside OSPAR, were 29 % lower in 2001 than the annual emissions in the year 1990. Taking into account the uncertainties regarding the reporting of emission data, there seems to be evidence that nitrogen oxides emissions are being reduced in the OSPAR area.

Ammonia (NH_3) emissions for the period 1990 - 2001 are shown in Annex 1, Table 2. Annual emissions of ammonia (NH_3) were also lower in 2001 than in 1990 for most of the OSPAR Parties and the selected three countries outside OSPAR. Out of 18 countries relevant for OSPAR, annual emissions in 15 countries were lower in 2001 than in 1990. In the Russian Federation, Poland and the Netherlands this emission reduction was significant (45 %, 39 % and 36 %).

In the Russian Federation, Poland and Germany ammonia (NH_3) emissions decreased in the beginning of the period investigated (approximately until 1994). From 1994 to 2001 emissions remained on the same level in almost all countries except Spain, where emissions were slightly rising from 1994 to 2001.

The rate of reduction of ammonia (NH_3) emissions was slightly lower than the rate of reduction of nitrogen oxides (NO_2) emission in the period 1990 – 2001. Annual emissions of ammonia, as a sum of emissions from all OSPAR Parties and the three selected countries outside OSPAR were 20 % lower in 2001 than the annual emissions in the year 1990.

Annual emissions of nitrogen oxides (NO_2), as a sum of emissions from all OSPAR Parties and the three selected countries outside OSPAR were 13 % higher than the corresponding annual emissions of ammonia (NH_3) in the year 2001, in terms of nitrogen emitted.

2.2 Heavy metals

Emissions of heavy metals are, to a limited extent, officially reported under the CLRTAP. In Annex 2, an overview of emissions in Contracting Parties is presented. This overview is based on the official submissions (ECE, 2003; 2004), or in cases where no official number is available, emissions are based on expert estimates (as presented by the EMEP Meteorological Synthesizing Centre East, see Ilyin and Travnikov (2003) and http://www.msceast.org; or taken from Berdowski *et al.* 1997) or by interpolation/ extrapolation between data of the nearest years.

For all metals a decrease in emissions has occurred between 1990 and 2000, the largest change is observed for lead emissions. For the heavy metals the following observations can be made for the changes between 1990 and 2000:

- Arsenic (As): emission reduction in Europe as a whole are slightly less than 40%. With a reduction of 45% the OSPAR region performs better than the rest of Europe. However, Spain forms an exception with increasing emissions.
- Cadmium (Cd): in Europe the emissions are about halved during this period. Emissions in Spain do not follow this trend and show an increase.
- Chromium (Cr): an overall emission reduction of about 35% but with increases on the Iberian Peninsula. For the rest of Europe a reduction of 30% is estimated.
- Copper (Cu): changes in copper emissions within the OSPAR region are highly variable (ranging from a 50% increase to a 80% decrease) but as a whole the emissions have been reduced by one third. In the rest of Europe emissions are more or less stable in this decade.
- Mercury (Hg): significant reductions in the OSPAR-region (average 60%) and in the rest of Europe (average 40%) have taken place.
- Nickel (Ni): the emission reduction in the OSPAR-region is about 25% which is less than in the rest of Europe (75%).
- Lead (Pb): in Europe as a whole and in the OSPAR-region the emissions have been reduced by about 70-80%; in Portugal the reduction is substantially less (*c.* 25%).
- Zinc (Zn): the emission reduction in the OSPAR region is about 25% which is more than in the rest of Europe. The reduction in the OSPAR region is not uniform; on the Iberian Peninsula an increase in emission is noted.

2.3 Organic pollutants: γ-HCH

Only a few European countries report annual sales or consumption data for individual insecticides, such as lindane and technical HCH. Overviews of European emission data are presented by Breivik *et al*. (1999) and Holoubek *et al.* (2000).

Recent emission inventories suggest that emissions in Europe as a whole have declined from 2332 to 790 tons during the 1980 to 1996 period, that is, a decrease of 75% (Holoubek *et al.,* 2000). Nowadays, use of lindane is severely restricted or banned in most European countries. Until recently, major use of lindane was made in France and, to a lesser extent, Spain (Breivik *et al.,* 1999), with France accounting for 70% of the European emissions. In Annex 3 emissions of γ-HCH in Contracting Parties are presented.

3. Concentration in precipitation

3.1 Mandatory pollutants

The following contaminants are to be measured on a mandatory basis in precipitation:

- heavy metals: arsenic (As)
  cadmium (Cd)
  chromium (Cr)
  copper (Cu)
  lead (Pb)
  mercury (Hg)
  nickel (Ni)
  zinc (Zn)
- persistent organic contaminants: γ-HCH[14] (lindane)
- nutrients: ammonium (NH_4^+)
  nitrate (NO_3^-)

The yearly averaged concentration in precipitation, $\overline{C}$, is estimated by precipitation amount weighted averaging of the concentrations for each of the sampling periods:

$$\overline{C} = \frac{\sum_{i=1}^{N} P_i C_i}{\sum_{i=1}^{N} P_i} \qquad [1]$$

where N is the number of sampling periods in one year, P_i is the precipitation amount and C_i is the reported concentration in period i. Reported data are frequently below detection limits (see, for example, the latest summary report on CAMP-2003 data (Barrett, 2004)) As one of the objectives of this assessment is the analysis of possible trends in concentrations, more stringent criteria than given in the CAMP-principles, have been applied in the calculations of annual mean values for heavy metals: values below detection limit are excluded and an annual mean is judged to be valid when temporal coverage is more than 75%. By applying these criteria the numbers presented in this report might differ slightly from earlier published data.

In Annex 4, an overview of the annual mean concentration in precipitation of the mandatory heavy metals is given. Annex 5 gives an overview of the availability of data on nitrogen wet deposition. Data on γ-HCH is given in Annex 6.

The location of stations where heavy metals, nutrients and γ-HCH in precipitation are measured is presented in Figure 2 and 3. For the metals cadmium, lead and zinc a reasonable spatial coverage is found; for the other metals – and in particular for mercury – coverage is less good and for large stretches of coastline there is a marked lack of observations. Nutrients are well covered but lindane in measured only in the more northern parts. The maps clearly show that, although a 17 year period is considered, some Contracting Parties have not reported on one or more of the mandatory pollutants during this period.

[14] In view of the low concentrations, the monitoring of α-HCH is no longer mandatory. When γ-HCH is not measured as a separate component, it should be clearly stated in the data reports that total HCH has been measured.

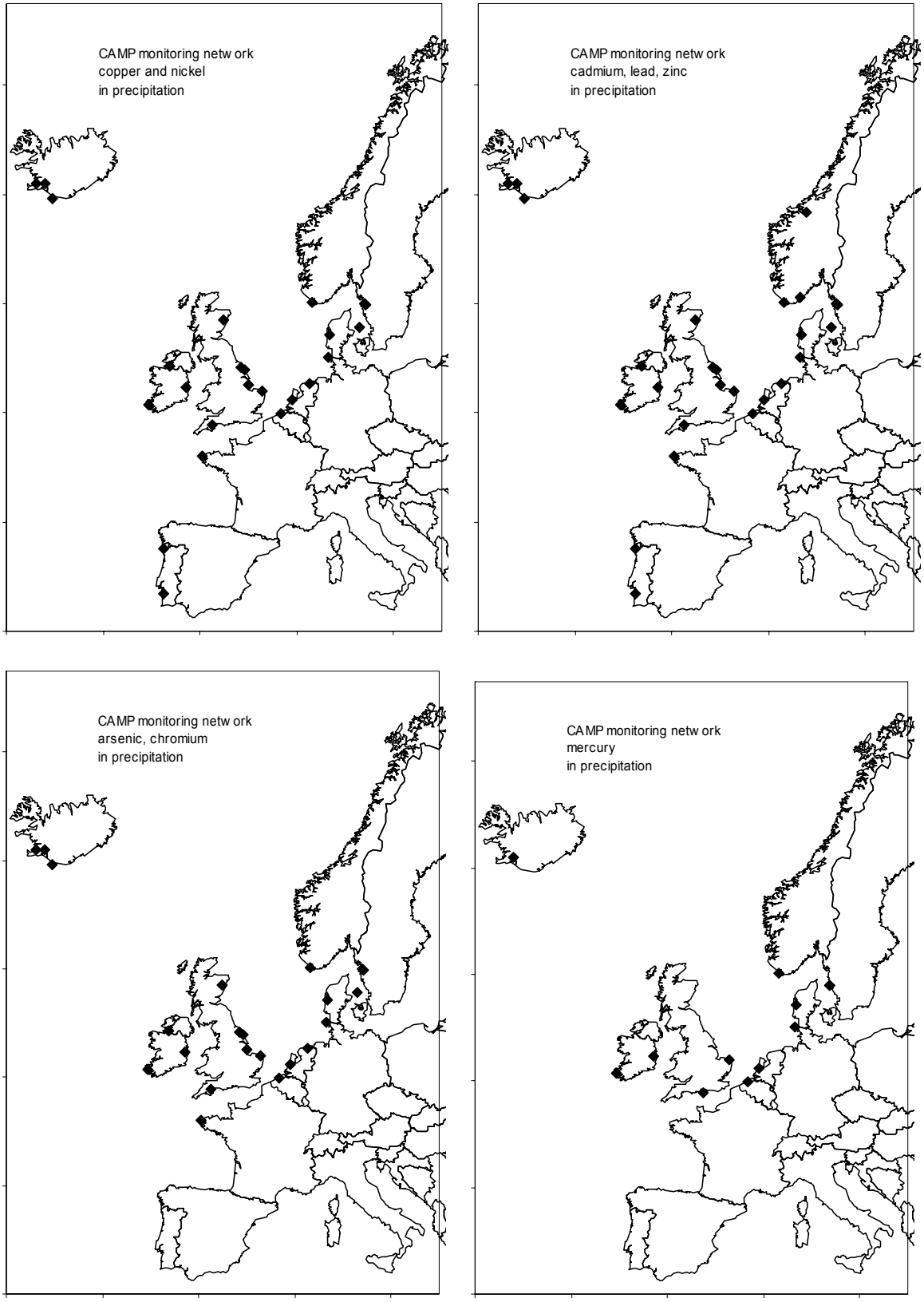


Figure 2. Location of stations measuring heavy metals in precipitation during one or more years in the period 1987-2003. (Note that the station Angra do Heriosmo (monitoring of cadmium, copper, nickel, lead and zinc) is located outside the plotted area).

The maps therefore give a rather optimistic view of the spatial coverage: all stations providing data for at least one year in the period 1987-2003 are shown. In a specific year the spatial coverage may be less than shown. Moreover, observed concentrations of heavy metals and γ-HCH are frequently below the detection limit. This further reduces the spatial coverage in the assessment.

Contracting Parties are responsible for quality control and validation of the data. This is an issue of major concern. Quality flags are frequently missing. While analysing the data outliers have been noted. Also rather constant concentration levels are seen, suggesting that detection limit values are reported without the correct quality flag.

Annex 4 shows that both bulk and wet only samples are used to collect precipitation. According to the CAMP-principles the use of wet-only samplers is recommended. The use of bulk collectors is acceptable when on a regular basis, an intercomparison of results of wet-only and bulk samplers actually in use in the network takes place. Except for the station BE0004R where parallel measurements using bulk and wet only samplers are made, the CAMP data base does not contain information on intercomparison studies as mentioned above. As bulk samplers contain an (unknown) contribution from dry deposition, a direct comparison between stations is strongly hampered.

To illustrate the differences between samplers, in Figure 4 data from the bulk and wet-only samplers at BE0004R are compared. The sampling periods of the two samplers are not identical which complicates a direct comparison. However, the total precipitation amounts correlates well although the bulk sampler measures systematically a larger amount. This suggests that the wet only sampler is not opening sufficiently quickly. The cadmium concentrations – presented here as a typical example - for the bulk sampler show a few large peaks which are flagged as valid measurements. These peaks are not seen in the results of the wet only sampler. The “constant” values seen during some periods are mostly flagged as “below detection limit”. Therefore, for those periods no reliable concentration estimate can be made.

3.2 Voluntary pollutants

The CAMP Principles recommend measurement of the following contaminants on a voluntary basis in precipitation:

- persistent organic contaminants:
    - the PCB-congeners 28, 52, 101, 118, 138, 153 and 180;
    - the following PAHs: phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo[*ghi*]perylene, indeno[*1,2,3-cd*]pyrene.

Precipitation data for these pollutants is reported for a very limited number of stations. Germany are preparing an expert examination of the quality of the CAMP data on organics. According to a progress report presented to the INPUT Working Group in 2005, benzo(a)pyrene in precipitation is measured only at 4 stations (located in Germany and Sweden). The reported data for about 10% of the samples are below the detection limit of the analytical devices. Due to the scarcity of data, the organic contaminants are excluded in the further assessments in this report.

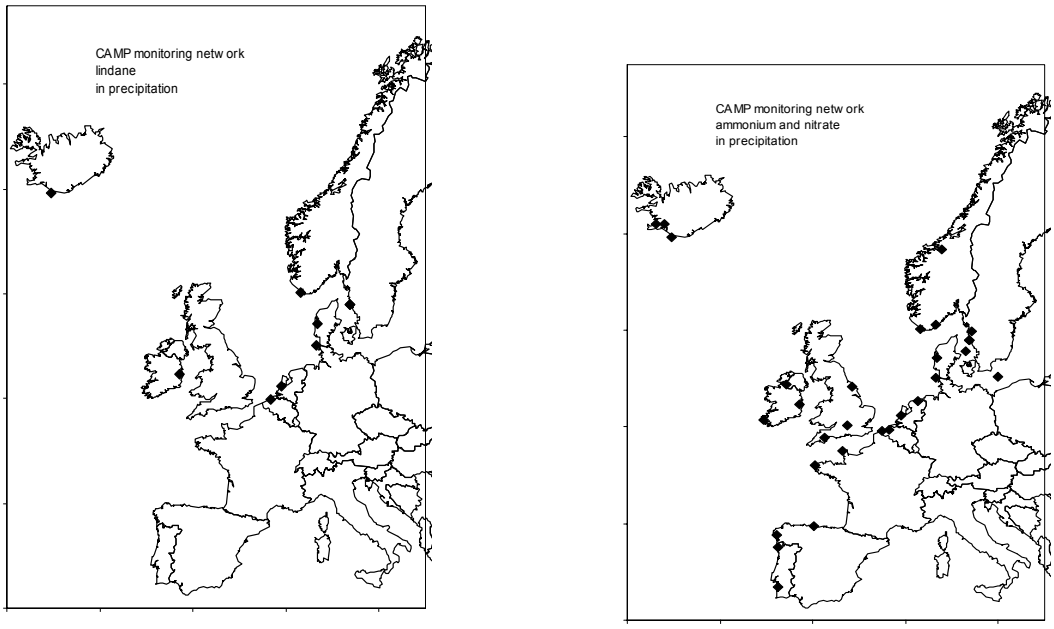


Figure 3. Location of stations measuring lindane (left) and nutrients (nitrate, ammonium; right) in precipitation during one or more years in the period 1987-2003. (Note that the stations Zeppellinfjell, Ny Alesund and Angra do Heriosmo (monitoring of nutrients in precipitation) are located outside the plotted area).

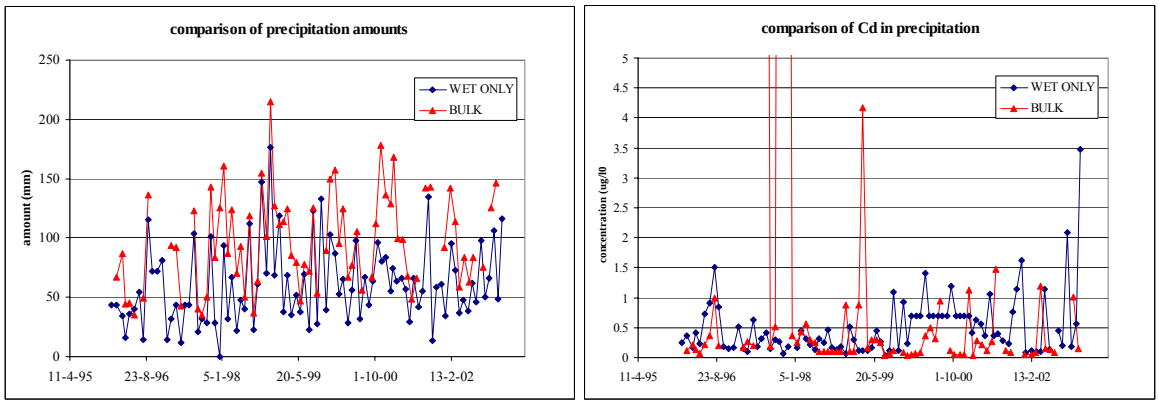


Figure 4. Comparison of precipitation amounts and cadmium concentrations in bulk and wet only samplers at the station BE0004R, Knokke.

4. Concentration in air

4.1 Mandatory pollutants

The CAMP Principles require monitoring of the following contaminants on a mandatory basis in air:

- nutrients: in gaseous phase: NO_2, HNO_3, and NH_3
  in aerosol phase[15]: ammonium (NH_4^+) and nitrate (NO_3^-)

An overview of the availability of data on the mandatory pollutants is given in Annex 7. Figure 5 shows the stations where nutrients in air have been measured at least during one year over the period 1987-2003. Again, a clear lack of observations in large parts of the OSPAR coast lines is evident.

[15] As an alternative total nitrate (sum of gaseous HNO_3 and particulate NO_3) and total ammonium (sum of gaseous NH_3 and particulate NH_4) can be measured.

4.2 Voluntary pollutants

The CAMP Principles recommend measurement of the following contaminants on a voluntary basis in air:

- heavy metals: arsenic (As)
  cadmium (Cd)
  chromium (Cr)
  copper (Cu)
  lead (Pb)
  Mercury (Hg)
  Nickel (Ni)
  Zinc (Zn)
- nutrients: nitrogen monoxide (NO)
- persistent organic contaminants:
  - γ-HCH (lindane);
  - the PCB-congeners 28, 52, 101, 118, 138, 153 and 180;
  - the following PAHs: phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo[*ghi*]perylene, indeno[*1,2,3-cd*]pyrene.

Of this voluntary list, a substantial amount of data is available for the heavy metals and, to a lesser extent, for γ-HCH. Availability and observed concentrations of the heavy metals are presented in Annex 8. Information on nitrogen monoxide (NO) and lindane in air is included in the overviews of the mandatory pollutants (Annex 7 and 6, respectively).

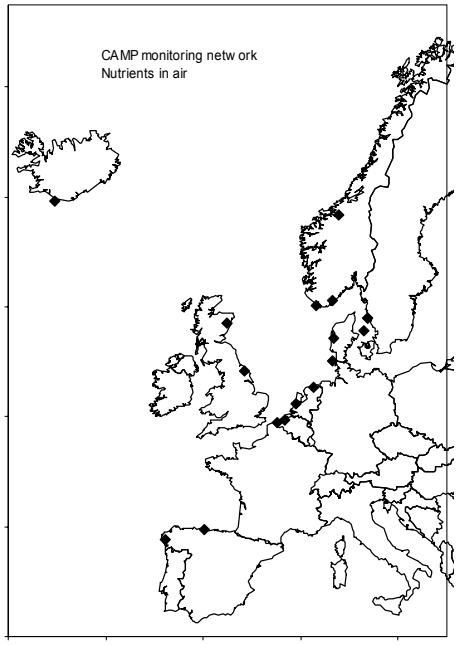


Figure 5. Location of stations measuring nutrients (nitrate, ammonium) in air during one or more years in the period 1987-2003. (Note that the stations Zeppellinfjell and Ny Alesund are located outside the plotted area).

5. Estimation of trends

5.1 Introduction

There is a widespread conception that temporal trends in air quality and atmospheric deposition of environmentally active substances must be rather large or persist for a long time before they can be detected by statistical trend tests. However, several research reports have demonstrated that a substantial fraction of the variation in the collected data can be attributed to random fluctuations in weather conditions. Air quality can vary strongly with air temperature, humidity and wind direction. Deposition can vary strongly with precipitation amount, rainfall frequency, and the source regions of the sampled precipitation. When annual

mean values are computed, this meteorological variation is partly but not fully suppressed. Hence, important anthropogenic trends can be concealed, and the natural interannual variation in weather conditions can also give rise to spurious trends.

When riverine inputs of nutrients to the sea are analysed for trends, it is generally accepted that flow-normalisation (or flow-adjustment) is a helpful tool to clarify the human impact on the environment, and an OSPAR Guidance document for this type of normalisation has been prepared. Also meteorological normalisation of air quality is in widespread use. For example, there are numerous research reports about meteorological normalisation of tropospheric ozone concentrations (Thompson *et al*., 2001). Normalisation of wet deposition was not scientifically examined until recently (Libiseller *et al*., 2005).

In general, a rather simple normalisation can remove a substantial part of the variation in collected data. For example, normalisation of annual riverine loads of nitrogen to a long-term average annual runoff can often clarify important anthropogenic trends. Likewise, the wet deposition of a variety of substances can be normalised to a long-term average precipitation amount. However, it is also well-known among statisticians that more advanced normalisations can give added value to collected data. In particular, it can be a great advantage to undertake the normalisation on daily or monthly data, even if the final goal is removal of irrelevant variation from annual summaries (Hussian *et al.,* 2004). Moreover, wet deposition and air quality data can be more effectively normalised, if measured data are adjusted for random fluctuations in the source regions of the sampled air or precipitation (Libiseller *et al*., 2005). A detailed analysis of trends in the wet deposition of nitrogen at selected EMEP stations illustrates this very clearly. When unadjusted data were analysed, not a single significant trend was detected. However, after a simple normalisation a few downward trends in both oxidized and reduced Nitrogen emerged, and after a more advanced normalisation the trend was even more clear.

In this report, a modified version of the EMEP trend tool (Salmi *et al.*, 2002) was used as the major instrument for trend assessment of precipitation data. The original tool was used for the aerosol data. This tool, originating from the Finnish Meteorological Institute, is designed for trend analysis of annual data. The well-known Mann-Kendall test is used to determine the statistical significance of downward or upward tendencies, and the trend slope is computed according to the method proposed by Cen. In our modified version of the EMEP tool, the measured wet deposition was adjusted prior to the Mann-Kendall test. Deposition values representing unusually wet years were decreased and values representing unusually dry years were increased in attempt to remove a substantial part of the weather-dependent fluctuations in the collected deposition data. How much the original values had to be decreased or increased was determined by fitting a straight line to the observed annual values of the deposition and the precipitation amount. The normalised values represent the deposition that would have been observed if the precipitation amount had been identical to its long-term annual mean. The mathematical formulae for this simple type of normalisation are given by Libiseller *et al*. (2005).

The software package RTrend (see INPUT 05/3/4-Add.3), which has been developed for trend assessments of riverine inputs, is in many respects more advanced than the EMEP tool used in the present study. For example, RTrend contains 'smoother-based' trend tests that can be utilised as alternatives to the Mann-Kendall test. Furthermore, RTrend contains various modules for data preparation and adjustments (normalisations). However, the latter modules are constructed specifically for riverine loads and runoff, and this makes the present version of RTrend less suitable for atmospheric deposition and air quality data. By its construction, RTrend is also less convenient for trend analysis of a large number of time series of data.

In summation, the time-constraints for the assessment and the lack of resources for modifying a complex software package, made the EMEP tool a more realistic option (see also the more in-depth comparison between Rtrend and the EMEP tool in Annex 9). The software that was developed specifically for the in-depth study of nitrogen trends (Libiseller *et al*., 2005) would also require modifications before it can be recommended for the relatively simple adjustments and trend assessments in the present report. Finally, it is worth mentioning that the ICES working group WGSAEM (Working Group on Statistical Aspects of Environmental Monitoring) discussed various normalisation methods at its meeting in Nantes, Feb 28 – Mar 4, 2005. In particular, it was concluded that there is a need for a common framework for all normalisation methods that are now being applied to different types of environmental data.

5.2 Heavy metals

5.2.1 Concentration in precipitation

The EMEP trend tool (Salmi *et al.*, 2002) has been applied to time series of heavy metals in precipitation. Trends for both the observed concentrations alone, and for the concentrations after normalising with annual

precipitation amount, have been calculated. Whilst 38 time series show significant trends in their observed pollutant concentrations alone, only 14 showed significant trends once concentrations were normalised for precipitation amounts.

Table 2. Results of trend analysis of heavy metals in precipitation.

Component/ station		First year	Last year	N (a)	Signif: (b) (Conc. Alone)	Signif: (b) (Normalised with precipitation amount)	Slope- µg/l/yr	Av. Con. µg/l last 3ys (Hg ng/l)	Rel. slope (%/yr)
As	**GB0014**	1994	2000	6	*(neg)				
	GB0090	1991	2000	9	*(neg)				
	GB0091	1991	2000	8	+(neg)				
Cd	**DK0031**	1989	2002	12	*(neg)	**	-0,003	0,04	-7
	GB0090	1990	2000	10	*(neg)				
	NL0091	1989	2002	6	*(neg)				
	NO0001	1987	2002	12	**(neg)	**	-0,003	0,04	-8
	NO0099	1990	2002	11	+(neg)				
	SE0097	1996	2002	6	*(neg)				
	SE0098	1987	1994	8	*(neg)				
Cr	**DK0031**	1989	2002	12	* (neg)	*	-0,019	0,19	-10
Cu	**DK0031**	1989	2002	12	*(neg)				
	GB0090	1987	2002	13	*(neg)	+	-0,081	2,00	-4
	GB0091	1989	2002	11	*(neg)				
	IS0002	1992	2001	10	**(neg)				
Hg	**NL0091**	1990	2001	9	***(neg)				
	SE0002	1991	2002	12	**(neg)	**	-1,094	9,04	-12
Ni	**DK0031**	1990	2002	11	*(neg)				
	GB0090	1994	2002	7	*(neg)	+	-0,083	0,49	-17
	GB0091	1989	2002	12	*(neg)				
Pb	**BE0004**	1996	2002	7	*(neg)	*	-2,107	22,18	-9
	DE0001	1990	2002	10	*(neg)				
	DK0031	1989	2002	12	***(neg)	***	-0,170	1,07	-16
	FR0090	1989	2002	11	*(neg)				
	GB0090	1989	2002	12	***(neg)	*	-0,329	2,38	-14
	GB0091	1990	2002	11	*(neg)				
	NO0001	1990	2002	13	***(neg)	***	-0,195	1,52	-13
	SE0097	1996	2002	6	*(neg)	*	-0,198	1,39	-14
Zn	**DK0031**	1990	2002	11	*(neg)				
	FR0090	1989	2002	12	**(neg)				
	IE0001	1989	2002	11	+(pos)				
	IS0002	1993	2001	9	+(neg)				
	IS0090	1992	2002	14	**(neg)				
	NO0001	1987	2002	14	***(neg)	***	-0,306	4,04	-8
	NO0099	1991	2002	12	*(neg)	+	-0,313	7,61	-4
	PT0003	1989	2002	13	+(neg)				
	PT0004	1997	2002	5	*(neg)	*	-4,429	8,85	-50
	SE0098	1987	1994	8	*(neg)				

a) number of years with valid data in given period
b) levels of significance *** α=0.001 ** α=0.01 * α=0.05 + α=0.1
c) slope (in % per year) relative to average concentration for last three years

The results (Table 2) are summarised below:

Arsenic: Significant trends are only seen in concentrations at three UK stations. Concentrations at the UK stations declined since the beginning of the nineties by a factor of two, in line with reported emission changes. An exception to this is for the Atlantic coast station FR90 where no apparent tendency is

observed, a non-significant downward tendency was seen at the other five stations reporting for five or more years. After normalisation against precipitation quantities none of the stations shows a significant trend.

Cadmium: Significant downward trends of a factor of 2 are seen in the concentrations at stations on the southern/eastern coastline of the North Sea (sub-regions 6-9, see Chapter 7.3 for definition of the North Sea sub-regions). Remaining stations commonly showed no apparent directional tendency. Significant downward trends of approximately a factor of two over the period are found at the stations DK0031 and NO0001 (sub-regions 8 and 9) after the concentrations are normalised for precipitation amounts.

Chromium: Only one station (DK0031) had significant downward trends in observations before and after normalisation against precipitation. Despite a halving of emissions in the reporting period, there were no clear trends for the other six stations with time series of at least five years.

Copper: Significant downward trends were found in concentrations at four stations across the region; nine other stations with long time series show non-significant downward tendencies. A non-significant, upward tendency is seen at the Portugese station PT0003. This accords with emission changes. The UK and Germany have seen substantial declines in copper emissions, while smaller changes have occurred elsewhere. The exception is the Iberian peninsula where emissions have risen notably. When data is normalised for precipitation a significant downward trend is found at only one station (GB0090).

Mercury: Whilst concentrations alone showed significant downward trends at Dutch (NL0091) and Swedish (SE0002) stations, when normalised for precipitation the only significant trend was at SE0002. Although emission changes have been substantial across much of the OSPAR region, this is not reflected in changes in concentrations. The absence of an observed decline in mercury levels may in part be due to the high detection limits resulting from the techniques chosen for several sites, namely in Ireland, Belgium and the UK (Wängberg and Munthe, 2001, 2004).

Nickel: Significant downward trends in concentrations are found at GB0090, GB0091 and DK0031; when normalised against precipitation amount, a significant downward trend remains only at GB0091. Whilst European wide decreases in emissions have been modest, those in neighbouring countries to these monitoring sites have been more substantial. A non-significant downward tendency is observed, both before as well as after normalisation, at five other stations with long time series. At three UK-stations (GB0014, GB0090, and GB0091) a possible discontinuity is noticed: data is missing for 1996, the concentration data before this gap are clearly higher than following 1996. This discontinuity may explain the observed downward trends.

Lead: Significant downward trends were found at five sites for normalised observations (BE0004, DK0031, GB0090, NO0001, SE0097), and at three additional stations for the observations of concentration alone (DE0001, FR0090, GB0091). Across the remaining stations included in the analysis the tendency also appeared to be downward. Emission changes have been substantial across Europe, and hence such a picture is not surprising.

Zinc: Whilst ten stations were found with significant downward trends in concentrations alone, and three with downward trends in normalised observations, there is also some uncertainty. One station, IE0001, is found to have had significant increasing trend in concentrations alone, while other stations show no apparent tendency whatsoever, e.g. NL0009, and the second Irish station IE0002. Some stations sometimes indicate opposite or changing tendencies over time, e.g. IS0090 where trends were downward until 1998 and upward after step-change from 1999. At station BE0004 the wet only and bulk sampler show opposite trends. Although not flagged as invalid, both samplers show suspiciously high values.

5.2.2. Concentration on aerosol

The EMEP-trend tool (Salmi *et al.,* 2002*)* has also been applied to time series of heavy metals on aerosol which cover at least a period of 5 years. Of the 60 time series fulfilling these criteria, 29 show a trend at a α=0.1 level of significance or better. Table 3 summarizes the results, graphs showing the trend lines are included in Annex 8. In all cases a linear trend is assumed.

Table 3. Results of trend analysis of heavy metals on aerosol

Component/station		First year	Last Year	N (a)	Signific. (b)	Slope ($ng/m^3.yr$)	Aver Conc (ng/m3) last 3 years	rel slope (%/yr) (c)
As	**DK0031**	1992	2002	9	***	-0.170	0.37	-46
	GB0090	1992	2002	11	+	0.077	1.08	7
	GB0091	1992	2002	11	*	0.026	0.40	7
	NL0009	1994	2002	9	**	-0.179	0.69	-26
Cd	**DE0001**	1987	2001	14	**	-0.019	0.16	-12
	GB0091	1989	2002	13	+	-0.007	0.083	-9
	NL0009	1994	2002	9	*	-0.021	0.19	-11
Cr	**DK0031**	1990	2000	7	+	-0.042	1.05	-4
	IS0091	1995	2002	8	+	0.597	8.19	7
Cu	**DE0001**	1987	1999	13	**	-0.137	1.56	-9
	DK0008	1996	2002	7	+	-0.048	1.21	-4
	DK0031	1990	2002	13	**	-0.072	1.14	-6
	GB0090	1989	2002	14	+	-0.112	2.32	-5
Ni	**DE0001**	1995	1999	5	*	-0.095	1.12	-8
	DK0031	1990	2002	13	***	-0.078	1.05	-7
	GB0090	1989	2002	13	+	-0.110	1.70	-6
	GB0091	1989	2002	13	*	-0.038	0.52	-7
Pb	**DE0001**	1987	2000	14	***	-1.430	6.47	-22
	DK0008	1996	2002	7	*	-0.497	4.42	-11
	DK0031	1990	2002	12	***	-1.137	4.03	-28
	GB0014	1994	2002	9	*	-0.739	5.71	-13
	GB0090	1989	2002	14	**	-1.143	9.8	-12
	GB0091	1989	2002	14	***	-0.457	2.13	-21
	NL0009	1990	2002	13	***	-2.181	7.94	-27
Zn	**DK0008**	1996	2002	7	+	-0.220	10.2	-2
	DK0031	1990	2002	13	***	-1.322	9.72	-14
	GB0014	1994	2002	9	*	3.758	47.1	8
	GB0091	1989	2002	14	+	0.709	20.5	3
	NL0009	1994	2002	9	+	-1.86	31.4	-6

(a) number of years with valid data in given period;
(b) level of significance: *** $\alpha = 0.001$
** $\alpha = 0.01$
* $\alpha = 0.05$
+ $\alpha = 0.1$
(c) slope (in % per year) relative to the concentration averaged over the 3 last monitoring years.

Inspecting the time series and the summary table leads to the following conclusions:

Arsenic: Seven time series were included in the analysis, four of them have a significant trend. Two stations in the UK show a positive trend which is not in line with the changes in emissions in the UK and in other Contracting Parties. No clear reason for this increase can be given. The decrease at the stations in Denmark and the Netherlands is slightly larger (observed reduction by factor of four) than would be expected from the changes in emission (reduced by factor of two).

Cadmium: Seven time series were analysed of which three showed a significant trend. The three time series with a significant trend show a consistent picture: a decrease of about 10% per year relative to the average concentrations over the period 2000-2002. This trend is in line with the trend in European emissions. The data from GB0091 for 1993 may be invalid although the quality flags indicate valid numbers. Deleting the suspicious year has no effect on the conclusions related to the trend.

Chromium: Two (out of six) time series have a significant trend, one upward, the second downward. The small downward trend in Denmark is in line with the emission estimates. In the OSPAR region as a whole emissions are constant between 1995 and 2000 but in the southern part of the OSPAR area (France,

Spain) increasing emissions are observed. Observations from the southern part of the area are lacking so no empirical confirmation of the positive emission trend can be given. The station in Iceland measures by far the highest Cr concentration of all operational CAMP stations. No clear reason for the positive trend is evident other than that there is an increasing impact from local emission sources and/or a measured artefact.

Copper: Copper in air has been measured at nine stations during a period of 7 to 14 years. At four stations a small (-4 to -9 % per year) but significant downward trend is observed. The emissions (Annex 2) show a 30% reduction between 1990 and 1995 and a levelling after 1995 (the slight increase in emissions is mainly caused by the increase in Spanish emissions).

Mercury: Gaseous mercury has been measured at 4 stations. No decreasing trend could be observed during the period 1995-2002. The measurements suggest that total gaseous mercury concentrations measured in the OSPAR area are dominated by the global background concentration to a larger extent than before (Wängberg and Munthe, 2005).

Nickel: For nickel, nine long time series have been analysed; four of them show a significant downward trend of 6-8% per year. Emissions between 1990 and 2000 reduced by about 3% per year.

Lead: As expected from the large reductions in emissions between 1990 and 2000, the ambient data at a relatively large number of stations show a significant downward trend of 10-25% per year.

Zinc: In contrast to the stations in Denmark and the Netherlands, two UK stations show a positive trend in Zn-concentrations. The emission data would suggest that, at least in the first half of the nineties, a decrease in concentrations could be expected.

5.3 Nutrients

5.3.1 Trends in wet nitrogen deposition

Wet deposition data for a total of ten sampling sites representing seven nations were analysed for trends in oxidized and reduced nitrogen. Considering that the emissions to the atmosphere of chemically bound nitrogen have decreased over the past decade, one could expect several significant downward trends in the collected data. Such trends were also found, but not until after adjustment for random variation in weather conditions. When observed wet deposition data were analysed without any prior adjustment, Mann-Kendall's nonparametric test did not reveal any statistically significant trends at the 5% level. After meteorological normalisation that eliminated the variation in deposition that could be attributed to interannual variation in precipitation amount, significant downward trends emerged at the EMEP stations of Birkenes (NO0001) and Rörvik (SE0002). However, it was not until the data were analysed with a higher temporal resolution and adjustments were made for random fluctuations in wind sector values that downward trends were revealed also at LaHague (FR0005), High Muffles (GB0014), and Kollumerwaard (NL0091). To be more precise, both nitrate and ammonium trends were found to be statistically significant at Birkenes, Rörvik and LaHague, whereas the decrease at High Muffles was significant only for nitrate and the trend at Kollumerwaard only for ammonium. Figures 6 and 7 illustrate the downward trends at Birkenes before and after normalisation.

An examination of possible errors in data or other signs of poor data quality revealed frequent inconsistent coding of missing values. Furthermore, the trend analysis was hampered by incomplete reporting. However, there were no signs of obvious errors in the reported concentration values.

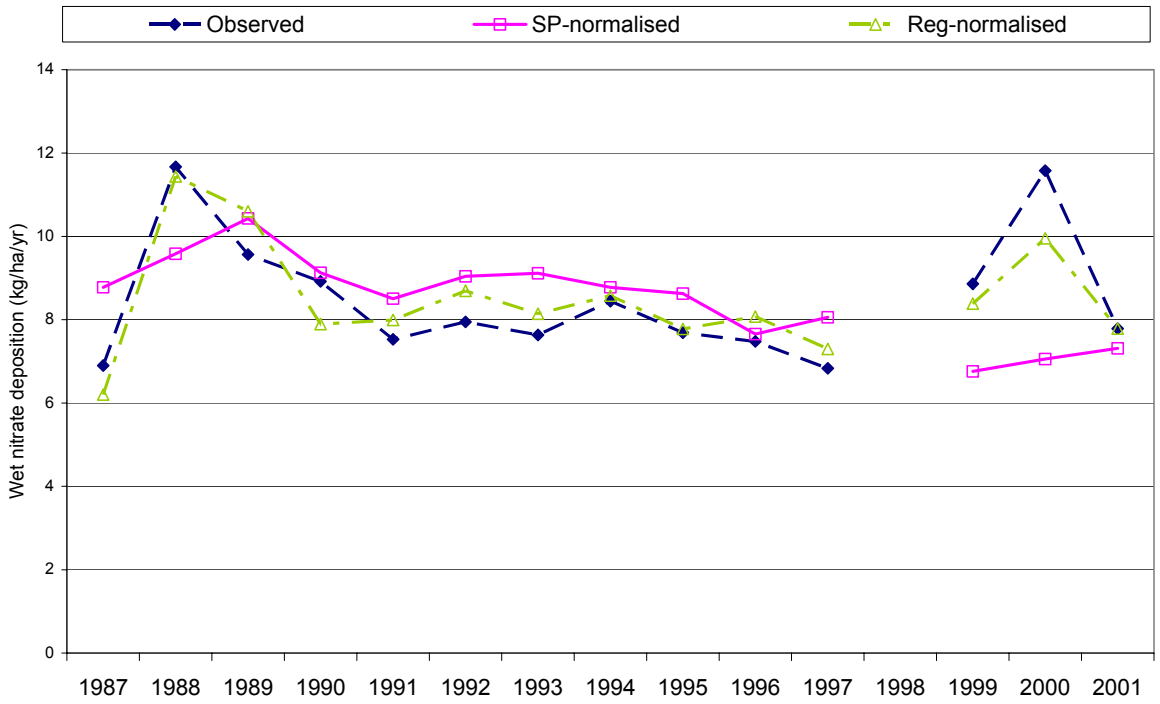


Figure 6: Time series of observed and normalised wet nitrate deposition at Birkenes (NO01). 'Reg-normalised' refers to simple normalization with respect to precipitation amount. 'SP-normalised' refers to the more advanced normalisation process developed by Libiseller et al. (2005)

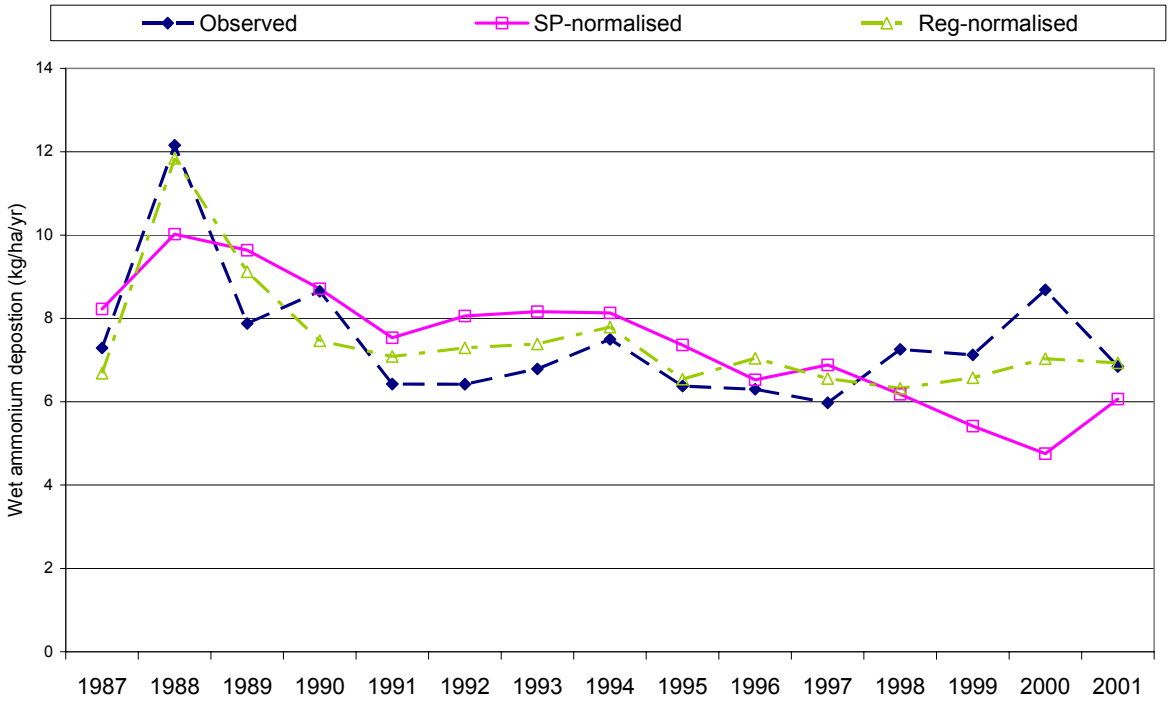


Figure 7: Time series of observed and normalised wet ammonium deposition at Birkenes (NO0001)

5.3.2 Trends in air concentrations of nitrogen species

Air quality data representing ammonia on aerosol (sNH4) and nitrate on aerosol (sNO3) concentrations for seven stations and NO2 data for four stations were analysed for trends before and after meteorological normalisation. In contrast to the wet deposition, normalisation of air quality had only a minor effect, and

almost identical trends were found in the unadjusted and adjusted time series of data. To be more precise, trend analysis revealed statistically significant downward trends in the concentrations of sNH4 and sNO3 at the EMEP stations of Anholt (DK0008) and Birkenes (NO0001), and in the NO2 concentrations at all EMEP stations for which time series for such data were available, i.e. at Kollumerward (NL0009), Noia (ES0005), Birkenes (NO0001), Kårvatn (NO0039) and Rörvik (SE0002). The station at Spitzbergen (NO0042) differed from all other stations in the sense that there was an upward trend in sNH4.

Incomplete reporting was found to be the greatest obstacle for a more thorough and reliable trend analysis of nitrogen species in air. In addition, some data quality problems were noted for the measurements of NO_2 in Norway. In particular, it can be seen that the detection level has varied over time (Fig. 8). Similar data quality problems seem to have affected the measurements of sNH_4 at Rörvik (SE0002).

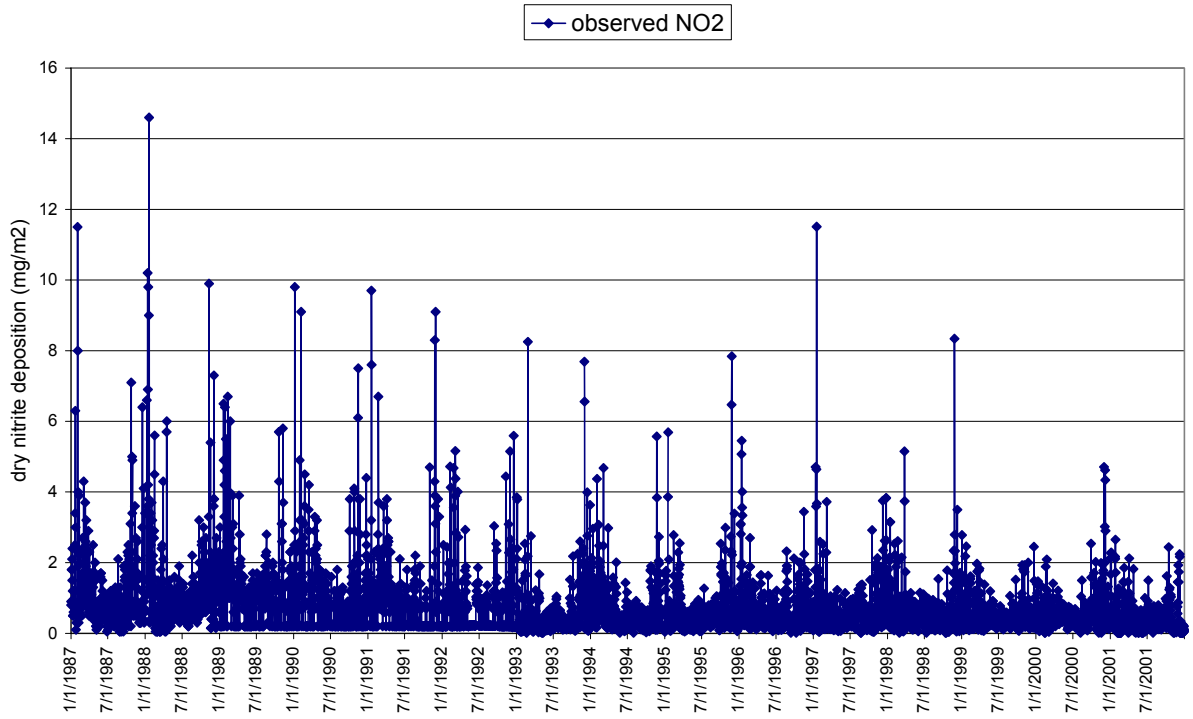


Figure 8: Daily NO_2 concentrations at Birkenes (NO01). Data quality problem can be seen by comparing data collected before and after 1993.

5.4 γ-HCH in precipitation and air

As γ-HCH in precipitation is frequently below the detection limit, long time series are available only at the stations IS0091R and NO0099R. Time series plots , both for concentration in precipitation as well as for total deposition, are given in Annex 6. The estimated trend at the Norwegian stations are significant at an α=0.01 level. These findings are in agreement with a more detailed analysis of measurements in the Netherlands (Buijsman and van Pul, 2003).

γ-HCH in air is measured at four stations (annual mean concentrations and time series plots are given in Annex 6). A significant downward trend is observed at three stations (IS0091R, NO0042R and NO0099R) but not at the Swedish station SE0002R. The concentrations in air show a strong downward trend, similar to the data in precipitation; the estimated linear trend in air and precipitation is similar. The initially high concentrations and the strong trend found at the station Lista is noteworthy (NO0099R). Changes in sampling strategies (from monthly sampling to irregular two day sampling) might have contributed to the observed trend (Institute for Energy and Environment, 2005).

6. Deposition to the Convention Waters

6.1 Nitrogen deposition

The atmospheric deposition of nitrogen to the OSPAR convention area over the period 1990 – 2001 has been estimated by EMEP using atmospheric dispersion models (Bartnicki and Fagerli, 2004). Here some main findings, taken from Bartnicki and Fagerli (2004), will be summarized.

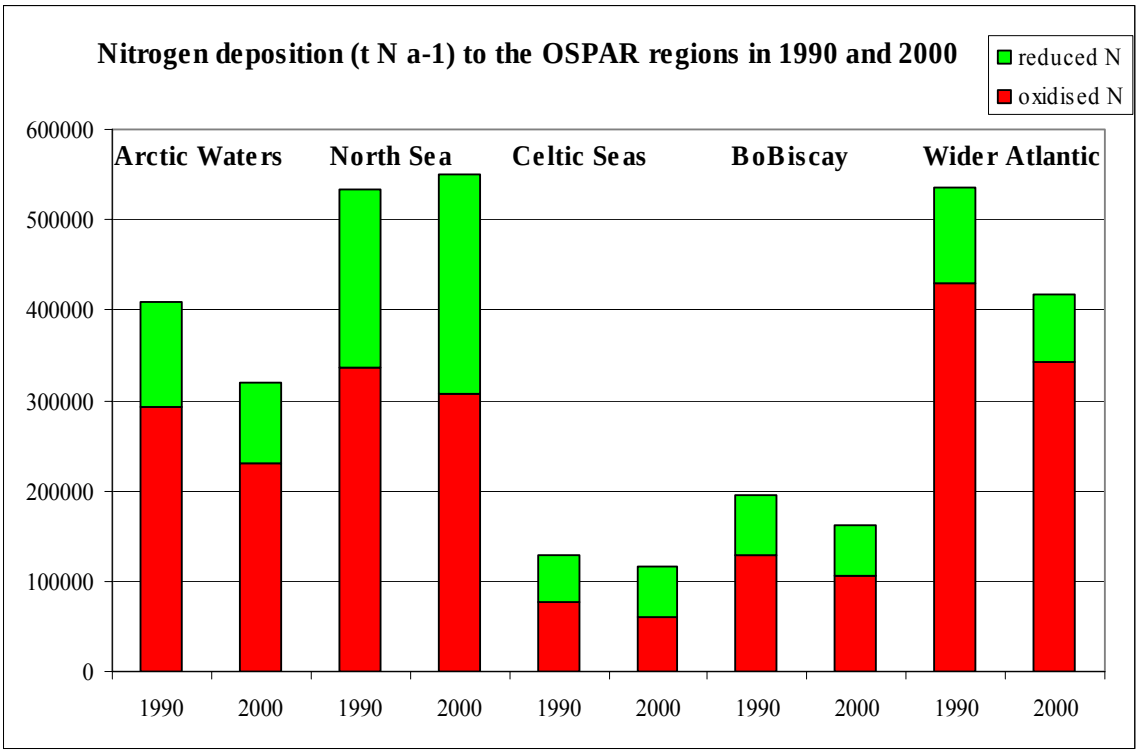


Figure 9. Nitrogen deposition (unit t N a^{-1}) to the main regions of the OSPAR maritime area, modelled data for 1990 and 2000.

Figure 9 shows the modelled total nitrogen deposition to the Convention Area for 1990 and 2000. Compared to 1990 the total N-deposition is in 2000 lower except for the Greater North Sea. In the OSPAR Contracting Parties and in countries strongly contributing to the deposition in the OSPAR region, emissions of nitrogen oxides are in total reduced between 1990 and 2001 (see section 3.1). For reduced nitrogen (NH3) a reduction in emissions – although to a lesser extent - has also been noted. The higher deposition to the North Sea in 2000 must be ascribed to the weather conditions; the deposition in 2000 is the highest in the period 1997-2001. Over the period 1990-2001 no clear change in N-deposition is modelled for the Greater North Sea. Figure 6 further shows that the contribution of oxidized nitrogen is by far more important to the total N input that reduced nitrogen.

The relative contribution of the Contracting Parties to the N-deposition in the five main regions (the relative contribution is estimated as the deposition caused by emissions in the specified country relative to the deposition in the area caused by all sources) is given for the situation in 2000 in Figure 10. The relative contribution of a country depends on the total emissions within the country and its orientation to the receptor area (where distance and orientation with respect to the prevailing wind direction are the most important). With this is mind, an interpretation of Figure 10 can be made. In the more remote areas (Arctic waters, Wider Atlantic) the contribution of remaining sources is mainly the contribution from the initial and boundary contribution of the dispersion model. In the other regions contributions of the remaining sources are much lower. International shipping on the North Sea (indicated by NOS) and on the Wider Atlantic (indicated by ATL) are important contributors to the deposition in all region; it should be noted that the ship traffic emissions are based on 1990 data and will be underestimated for the 2000 situation. The main land-based sources of nitrogen emissions responsible for the nutrient input to the OSPAR area are located in the large countries: United Kingdom, France, Germany, Spain. Portugal and Ireland are important contributors to the deposition in the Bay of Biscay and the Celtic Seas, respectively; not surprisingly in view of their location.

As with the contribution of individual countries, the contribution of individual emission sectors can be defined. Agriculture, (responsible for more than 80% of the ammonia emissions), and transport (road transport and other mobile sources) are the most important contributing sectors.

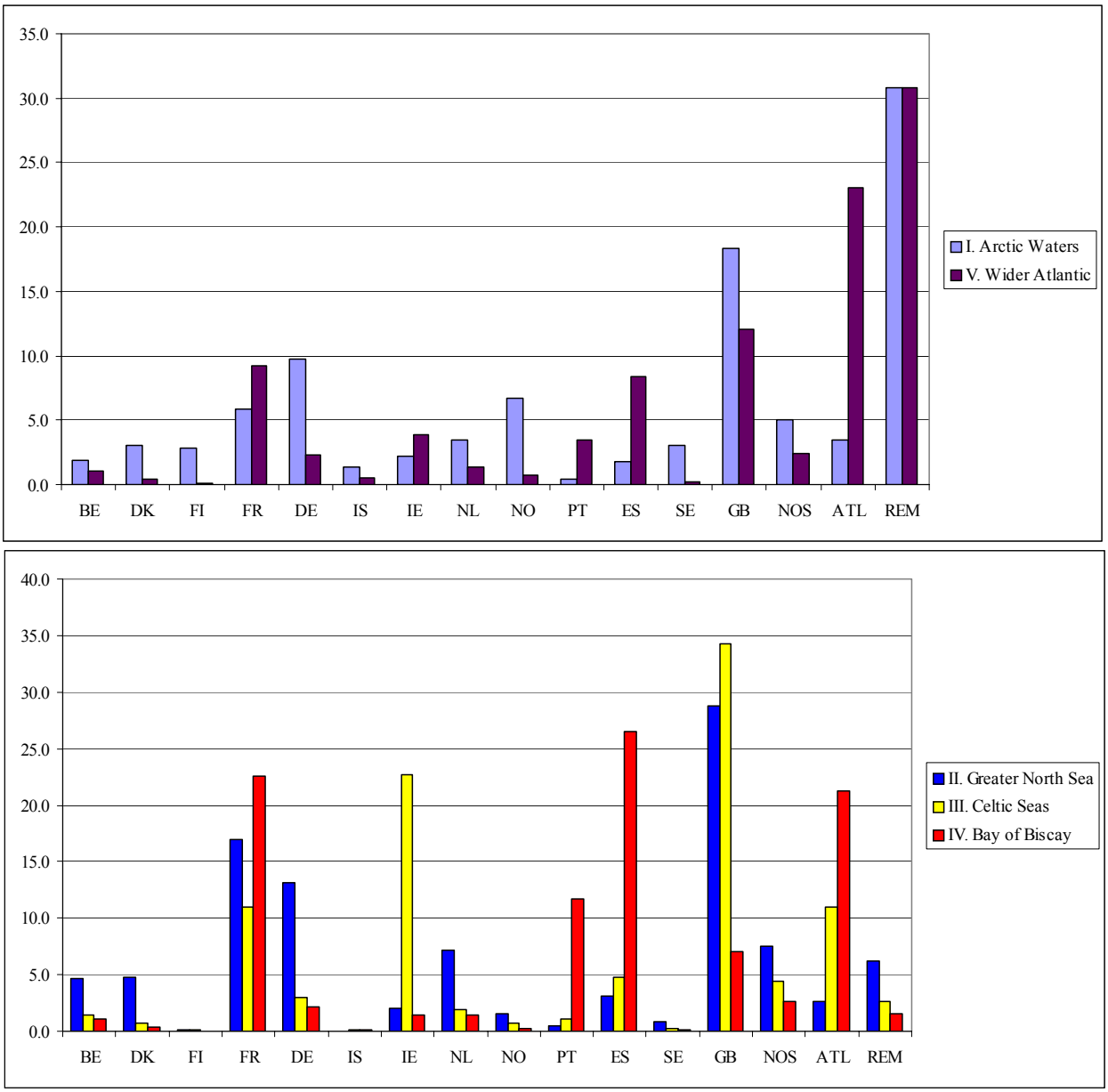


Figure 10. Relative contribution (in % of total deposition) of Contracting Parties to the total deposition in the OSPAR regions. The countries are indicated by their ISO-3166 alpha-2 code; the contributions of ship traffic emissions on North Sea and Wider Atlantic are indicated by NOS and ATL, respectively. REM stands for the contribution from all other European countries and from undefined sources.

6.2 Deposition to the North Sea: "Method 3a"

The atmospheric input is a mass of a contaminant carried to the maritime area via the atmosphere. Total deposition consists of the sum of wet and net dry deposition; for (semi)-volatile compounds the net dry deposition term includes the mass which has re-volatilised from the sea surface[16].

[16] Note that the net dry deposition term might be negative. This might be the case when some material which entered the maritime system by other pathways (e.g. wet deposition, riverine input, direct dumping) re-volatises.

Although the locations of CAMP monitoring stations have been carefully selected and measurements are not directly influenced by inland sources, the atmospheric input to the marine area can not be calculated directly from the (land-based) measurements. Deposition estimates based on averaging the deposition flux over all monitoring stations will be in error for several reasons:

- As sources of contaminants are mainly land-based one can expect that the concentrations in the more remote parts of the Convention waters will be lower than at coastal stations; the use of coastal data might lead to an overestimation of atmospheric inputs.
- For (semi)-volatile POP the dry deposition estimates based on ambient data will not include the effect of re-volatilisation.
- It is generally assumed that precipitation amounts at open sea are lower than at nearby land areas; the use of coastal precipitation data might lead to an overestimation of atmospheric inputs. Precipitation rates at coastal stations are extremely variable; for example, coastal stations in the United Kingdom have generally less precipitation than the stations in Norway, most probably due to orographic effects. To take this into account, and to correct for the smaller precipitation amount at open sea, a method which allows variable amounts of precipitation to be used based on 70% of the median values for coastal stations has been proposed for estimation of inputs to the North Sea.
- The CAMP monitoring stations are not regularly spaced along the coast line of the maritime area. For example, for the North Sea an averaging of deposition fluxes measured at coastal stations may over-estimate the atmospheric input due to the relatively higher network density in the southern, more polluted part of the North Sea. Other interpolation methods e.g. by inverse distance weighting, may overcome this problem but any concentration gradients from land to sea due to removal processes or due to dilution (the first point discussed above) are still not accounted for.

Since in the CAMP-network measurements are available on a monthly or 4-weekly basis only, it is not possible to separate on-shore and off-shore winds and estimate direction dependent concentrations. However, it is possible to apply a bulk gradient correction factor for each station derived from calculation with an atmospheric transport model. These *dispersion factors f(i)* are defined as the ratio of the modelled deposition flux at station *i* to the modelled deposition flux for the Convention Waters as a whole or sub-regions of it. This model-based dispersion factor can now be applied to extrapolate the measured deposition at the coastal station to the whole North Sea area and/or its sub-regions; total deposition is calculated by:

$$d_{(wet+dry)} = A\frac{\sum_{i=1}^{i=n_w} C_w(i)\, pa_{land}\; f_w(i)}{n_w} + A\frac{\sum_{i=1}^{i=n_a} C_a(i)\, v_d\, t\; f_a(i)}{n_a} \qquad [2]$$

where A is the total area of the North Sea (525,000 km^2) and/or sub-regions, n_w the number of stations with wet deposition measurements, n_a the number of stations with air concentrations measurements, v_d is the dry deposition velocity, t is the length of the period considered (here one year = 365*24*3600 s), $C_w(i)$ the weighted mean concentration in precipitation, pa_{land} is the precipitation amount and $C_a(i)$ the mean concentration in air at station i. $f_w(i)$ and $f_a(i)$ represent the dispersion factors for wet and dry deposition, respectively as calculated by the RIVM/TREND model. These dispersion factors have been modelled several years ago. After that date, some changes in the CAMP network were made. For new stations dispersion factors from the nearest existing station were selected. As discussed above it is suggested to take for pa_{land} the median precipitation value of all the stations rather than using station specific precipitation amounts.

The procedure described above and usually indicated as “Method 3a” (see Oslo and Paris Commissions (1994) for a more detailed description) is applied here on the CAMP data for the period 1987-2002. An overview of the wet and dry deposition loads and total atmospheric input to the North Sea is presented in Table 4 and Figure 11. Monitoring data sets for validation of “Method 3a” are not available; it can only be compared with other atmospheric input estimates , some of them based on measurements, others on model calculations. Such a comparison between five model based and two measurement based studies has been made by Nijenhuis et al. (2001), see Table 4. Although the definition of the *North Sea* may differ in the various studies resulting in different area and the base year of the emission estimates differs, some general observations can be made:

- the difference between the five model based methods are relatively small.
- the results of “Method 3a” are in the range of the two other measurement based estimates;
- measurement based methods show higher estimates of the deposition than model based methods.

Table 4. Deposition of heavy metals (tonne/a) to the North Sea estimated using "method 3a".

	As	Cd	Cr	Cu	Ni	Pb	Zn
1987		53					
1988							
1989		29	65	554	176	1040	2949
1990		39	99	417	243	1123	3436
1991		29	78	684	269	1222	2791
1992	86	27	342	477	249	893	3088
1993	129	33	332	514	228	1017	3362
1994	61	22	56	370	136	983	3037
1995	101	28	140	663	203	833	3419
1996	85	21	279	440	183	1314	4901
1997	95	21	136	372	103	853	3362
1998	41	18	314	405	169	583	4047
1999	36	36	60	451	117	557	2862
2000	67	22	322	425	263	635	3721
2001	54	23	56	366	114	448	3429
2002	86	37	93	309	176	530	3760
Reduction(1)	46	26	6	32	43	56	-22
Emission reduction (2)	47	44	38	33	23	82	27
Comparison with other input studies based on:							
Monit (3)	130-580	50-250		500-2300	230-1100	2300-12000	2700-12000
Monit (4)	220-720	110-430	70-1400	1400-10000	360-3600	3600-13000	7200-58000
Models (5)	42-49	9-15	26-74	46-130	49-222	690-2600	550-1200

(1) Percentage reduction over the 14 year period 1989-2002 (for As, 11 years, 1992-2002);
(2) Percentage reduction in emissions in OSPAR Contracting Parties over the period 1990-2000, see Annex 2.
(3) GESAMP, 1989;
(4) Van Aalst *et al*., 1983;
(5) Nijenhuis *et al.,* 2001; Baart *et al*., 1995; Van den Hout, 1994; Wulffraat *et al*., 1993; Van Jaarsveld *et al*., 1986.

One reason for this latter point might be that the wet deposition measurements are easily contaminated and tend to give overestimates of wet deposition. Another reason may be that there seems to be a systematic underestimation of the emissions.

For most of the metals the data do not suggest a significant trend. The EMEP trend tool reveals significant downward trends for copper and lead. Non-significant downward tendencies are calculated for the other metals except for zinc where a positive tendency is found. The total deposition is dominated by the wet contribution. The changes in deposition over time are reasonably in line with the observed changes in concentrations in precipitation. The upward tendency for zinc deposition is surprising; with the exception of the bulk sampler at BE0004, all stations around the North Sea show a decreasing tendency – although mostly not significant. The increase in Zn deposition is most likely caused by the introduction of the station BE0004 in 1996. The concentrations of Zn and other heavy metals, measured at this station are very high when compared with neighbouring CAMP stations. It might well be that this station is influenced by a local source and therefore not representative for the North Sea. If this station is not included in the "Method 3a" procedure, the Zn deposition decreases non-significantly with 11% over the 1989-2002 period. With the exception of Cr and Zn, there is a fair correspondence between changes in emissions and deposition.

Model estimates (Nijenhuis *et al.,* 2001) indicate strong reductions in deposition between 1990 and 1995 for all heavy metals considered. For arsenic, lead, copper and nickel a reduction of 20-35% over this 5 year period has been modelled. The observations of lead and copper are not in contradiction with the model results but the expected reduction is not seen in the observations of the other heavy metals. The large scatter (and uncertainties) in the precipitation data may play a role here. Further, the applicability of "Method 3a" should be re-assessed. Although this method overcomes some shortcomings related to the

representativeness of the CAMP network and stations (see above) , it is essentially based on the situation around 1985. The changes in the network and the –sometimes dramatic – changes in source strength and location of emissions are not correctly included in the dispersion factors.

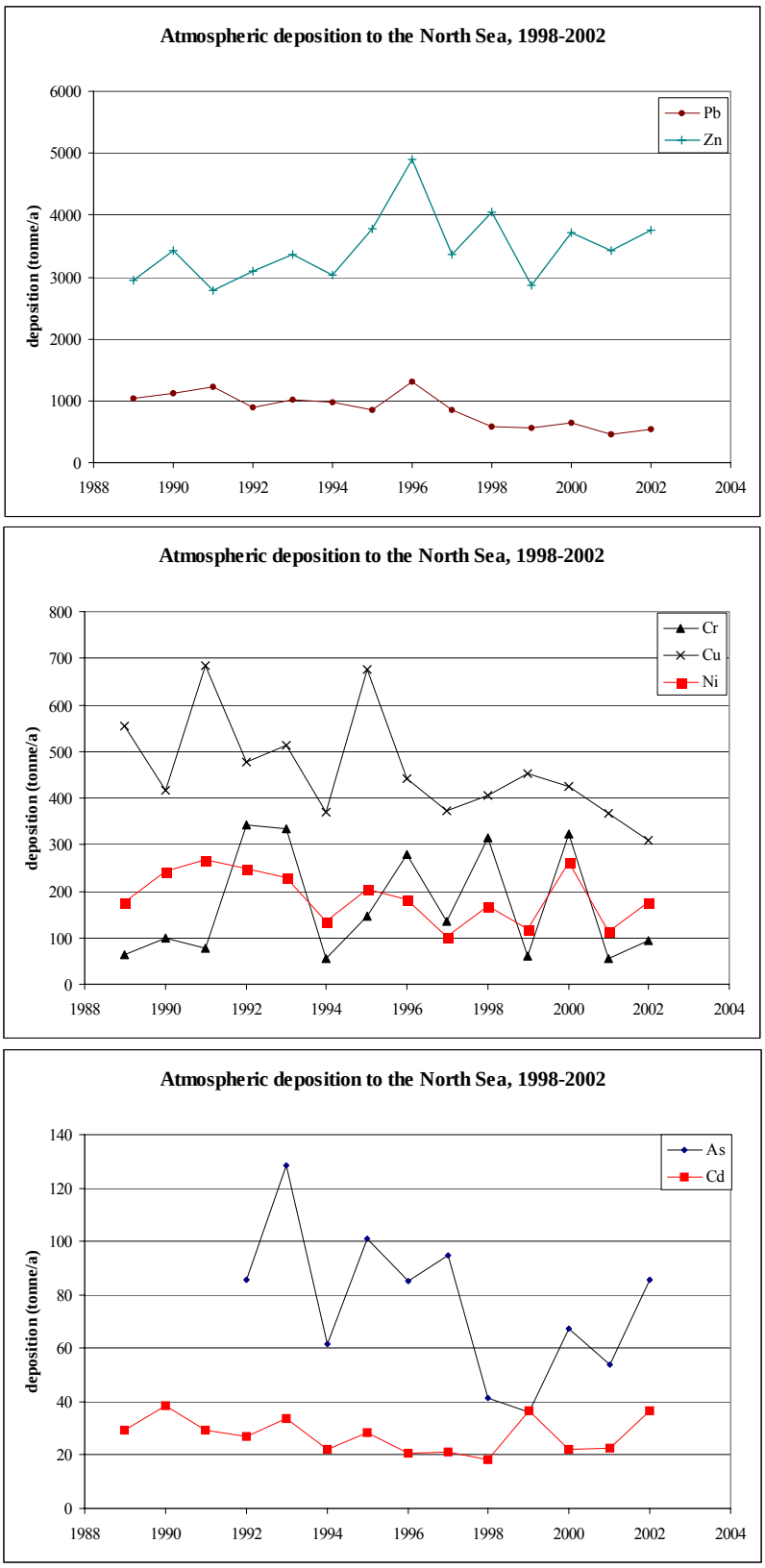


Figure 11. Atmospheric deposition of heavy metals to the North Sea (estimations based on "method 3") ; 1989 – 2002.

Application of "Method 3a" in the estimation of the atmospheric load of nutrients and some persistent organic pollutants is, in principle, possible. However, the appropriate dispersion factors for nutrients have not been

evaluated (Oslo and Paris Commission, 1994). Here we will estimate the wet nitrogen deposition with a modified version of "Method 3a" (Oslo and Paris Commission, 1994):

$$d_{(wet)} = A \frac{\sum_{i=1}^{i=n_w} C_w(i)\ pa_{sea}}{n_w} \qquad [3]$$

where pa_{sea} is the precipitation amount at open sea estimated as 70% of the median values for coastal stations. The contribution of dry deposition to the total atmospheric input can not be estimated because:

- There is limited information on concentrations in air;
- No representative dry deposition velocity can be estimated. For the N-species the sum of gaseous ammonia and particulate ammonium or the sum of gaseous nitric acid and particulate nitrate is measured. Deposition velocities for the gaseous and aerosol bound species may differ up to an order of magnitude.

As for the North Sea area the total atmospheric input of N-species is dominated by wet deposition (see, e.g. van Jaarsveld, 1992) the application of Eq. [3] will lead to a slight underestimation of the total atmospheric N-input. Final results are given in Figure 12. Equal contributions of reduced (ammonium) and oxidised (nitrate) nitrogen are estimated. The total N-input varies between 250 and 400 ktonne N/a. In total-N and ammonium deposition no trend can be detected; the nitrate deposition shows a significant (α = 0.05 level) downward trend. A comparison with the EMEP results and riverine input is given in the next section.

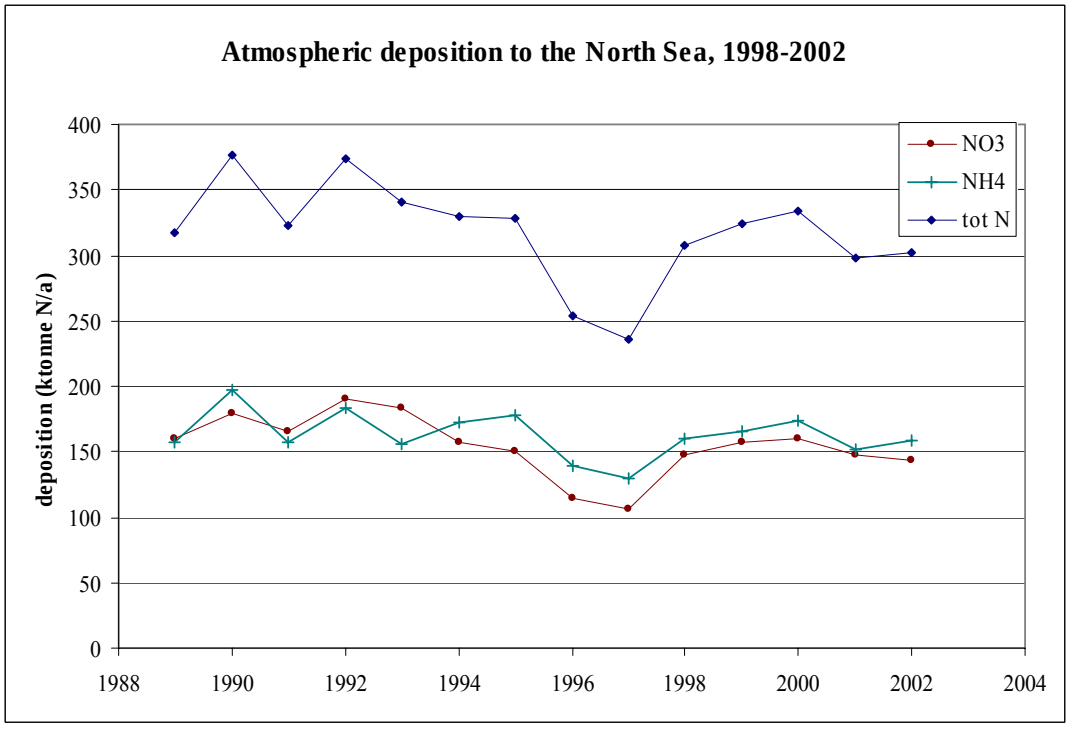
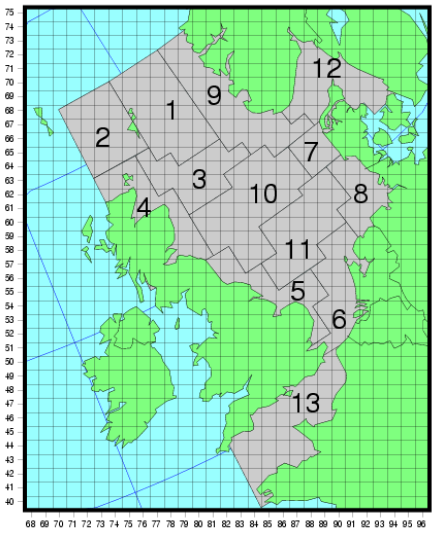
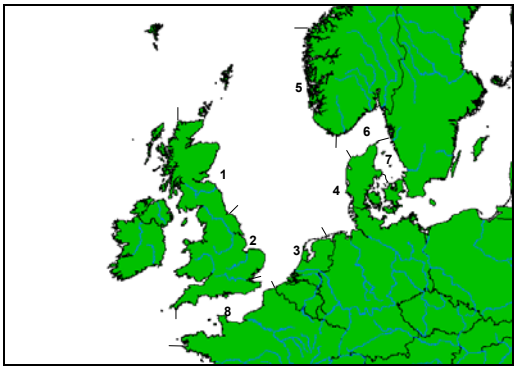


Figure 12. Atmospheric deposition of nitrate (NO3) and ammonium (NH4) to the North Sea (estimates based on wet deposition only using a modified "Method 3a")

6.3 Atmospheric input in relation to other inputs

The modelled atmospheric nitrogen deposition to regions of the OSPAR maritime area provided through the programme of cooperation with EMEP (section 7.1) have been compared with trends in riverine inputs and direct discharges of nitrogen calculated from RID data. A separate assessment of data collected under the RID study will be published parallel to this report.

Main characteristics, similarities and differences between both assessments are:

EMEP-assessment	RID assessment
• NO_x, NH_3 and total nitrogen; • for North Sea Main Body total nitrogen, lead and cadmium deposition estimated using "Method 3a". • five OSPAR Regions and 13 subregions (according to the ICES Boxes) to OSPAR Region II (Greater North Sea), see figure.  • comparison for three sub-regions : o North Sea Main Body (sub-region 1-11) o Skagerrak and Kattegat (subregion 12) o Channel (subregion 13); excluded in the analysis as RID data set are incomplete • Deposition calculated for eight years: 1990, 1995-2001 • all results (maps, tables, ascii files with data) are available from www.emep.int/index_facts.html • published as INPUT 05/2/2 Add.1	• total nitrogen, cadmium, lead and mercury; • three OSPAR Regions (I, II and III) and eight sub-regions to OSPAR Region II (Greater North Sea), see figure;  • for Regions IV no assessment could be made due to data gaps; • no waterborne inputs exist for Region V • comparison for three sub-regions : o North Sea Main Body (sub-region 1-5) o Skagerrak and Kattegat (subregions 6 and 7) o Channel (subregion 8); excluded in the analysis as RID data set are incomplete • data available for period 1990-2002 • published (as draft final report) as INPUT 05/3/5 Add.1

The comparison between the two assessments can for the moment only be made for total nitrogen for the OSPAR Regions I, II and III for the years 1990-2001. For Region II, Greater North Sea, more detailed information is available and a further sub-division is used. Note that the waterborne total nitrogen input comprise the inorganic as well as organic part, the atmospheric total nitrogen deposition only comprise the inorganic part. The organic nitrogen load via the atmosphere is assumed to be small.

For the sub-region North Sea Main Body a comparison between the atmospheric and waterborne inputs of heavy metals can be made. In this case the atmospheric loads are based on the "Method 3a"-evaluation. The documentation of "Method 3a" for the *North Sea* is not well defined. In view of the selected stations used in the method and on basis of the quoted area (525 000 km^2) "Method 3a" is most likely better representative for the North Sea Main Body than for the Greater North Sea.

Because of the major influence that riverine flow rate has on the absolute values of some riverine inputs, it is useful to apply flow adjustment to riverine inputs data in order to reduce meteorological induced variabilities and, thus, more reliably evaluate any underlying trend in inputs. In order to filter out these variabilities as effectively as possible so that the anthropogenic input pattern becomes clearer, adjustment procedures have

been developed (JAMP "Guidance on Input Trend Assessment and the Adjustment of Loads" and associated software RTrend) and applied in the RID assessment.

Results[17]

I Arctic Waters

Figure 13 gives a comparison between the atmospheric deposition and waterborne input of total nitrogen. As the load by riverine input is not significantly flow-dependent, no adjustment on the riverine load is needed. It is clear that for the Arctic Waters the atmospheric deposition exceeds the load by direct discharges and riverine inputs by a factor 6-10. Trends over time shows a decreasing trend for the atmospheric deposition (about 25% reduction over the period 1990-2001) whereas the load via rivers is more or less stable over time. Direct discharges increase significantly (165%) over time; there is a need to evaluate the reasons for the steady increase.

II Greater North Sea

Nutrients

The comparison for the nitrogen load to the Greater North Sea is given in Figure 14. For this OSPAR Region, the riverine load data has been adjusted for variation in flow rates. The largest contribution to the total nitrogen load is given by riverine inputs. The direct discharges are about 1/10 of the riverine inputs and about 1/5 of the atmospheric total N deposition. Atmospheric deposition contributes about one third of the total nitrogen load. In contrast to the atmospheric input which shows no significant change in the period 1990-2002, the direct discharges show a significant reduction of 30% in this period whereas the total waterborne input is significantly reduced by 12%.

For the sub-region North Sea Main Body (ICES regions 1-11) and Skagerrak and Kattegat (ICES region 12) the air- and waterborne inputs are compared in Figure 15 and 16. Results for the Main Body are very similar to those for the Greater North Sea: waterborne input dominates; the atmospheric input (both the EMEP- and "Method 3a" results) shows no clear changes over time while the total waterborne input and direct discharges are significantly reduced by 14% and 40%, respectively, over the period 199o – 2002. The EMEP and "Method 3a"results are very similar; as expected, the EMEP results are systematically higher as in "Method 3a" the contribution of dry deposition has been neglected. The EMEP model estimates the NO3/NH4 ratio as 60/40 whereas "Method 3a" gives a more 50/50 distribution.

Also in the Skagerrak/Kattegat region the waterborne input dominates; it is about twice as high as the atmospheric deposition. The atmospheric deposition shows no clear trend; the reduction of 10% in the Skagerrak and 20% in the Kattegat sub-region of the waterborne inputs of total nitrogen is not significant. Only the direct discharges into the Skagerrak and Kattegat, which comprise about 1/5 of the riverine inputs, are decreasing significantly with 30% and 55%, respectively.
In general it could be concluded that for coastal waters the direct atmospheric input is substantial but smaller than the riverine input and direct discharges.

Heavy metals

For the heavy metals lead and cadmium the comparison is given in Figure 17. For both metals the RID and atmospheric inputs are roughly of the same magnitude. However, as mentioned above, it might well be that "Method 3a" overestimates the atmospheric deposition.

III Celtic Seas

Figure 18 shows the results for total nitrogen for the Celtic Seas. As the load by riverine input is not significantly flow-dependent, no adjustment on the riverine load data is needed. Note that for three years (1999-2001) data on riverine loads from Ireland (estimated to be around 150 ktonne N/a) are missing. No realistic change in input - neither via the atmosphere, nor by direct discharges and riverine inputs – is observed. Waterborne input is roughly a factor of two larger than the atmospheric deposition.

[17] Annex 10 provides a numerical comparison of the loads for all regions and pollutant considered here.

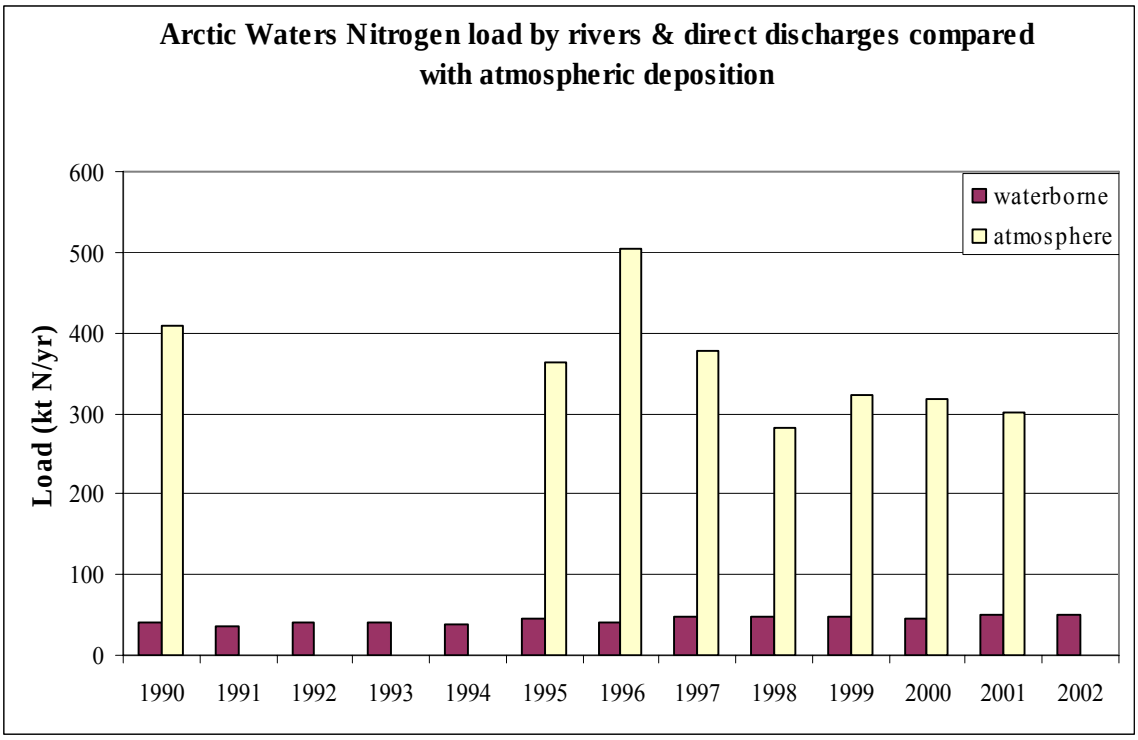


Figure 13. Total nitrogen load into the Arctic Waters by direct discharges and riverine inputs compared to the total atmospheric deposition (EMEP model results).

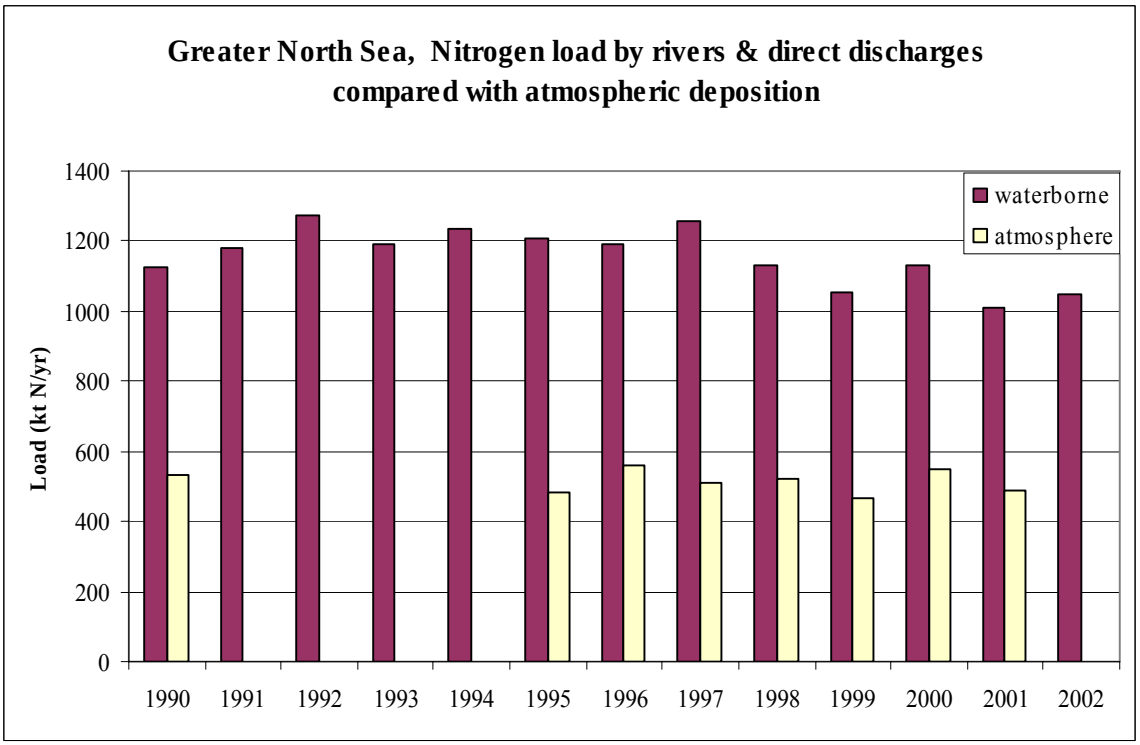


Figure 14. Total nitrogen load into the Greater North Sea by direct discharges and riverine inputs compared to the total atmospheric deposition (EMEP model results).

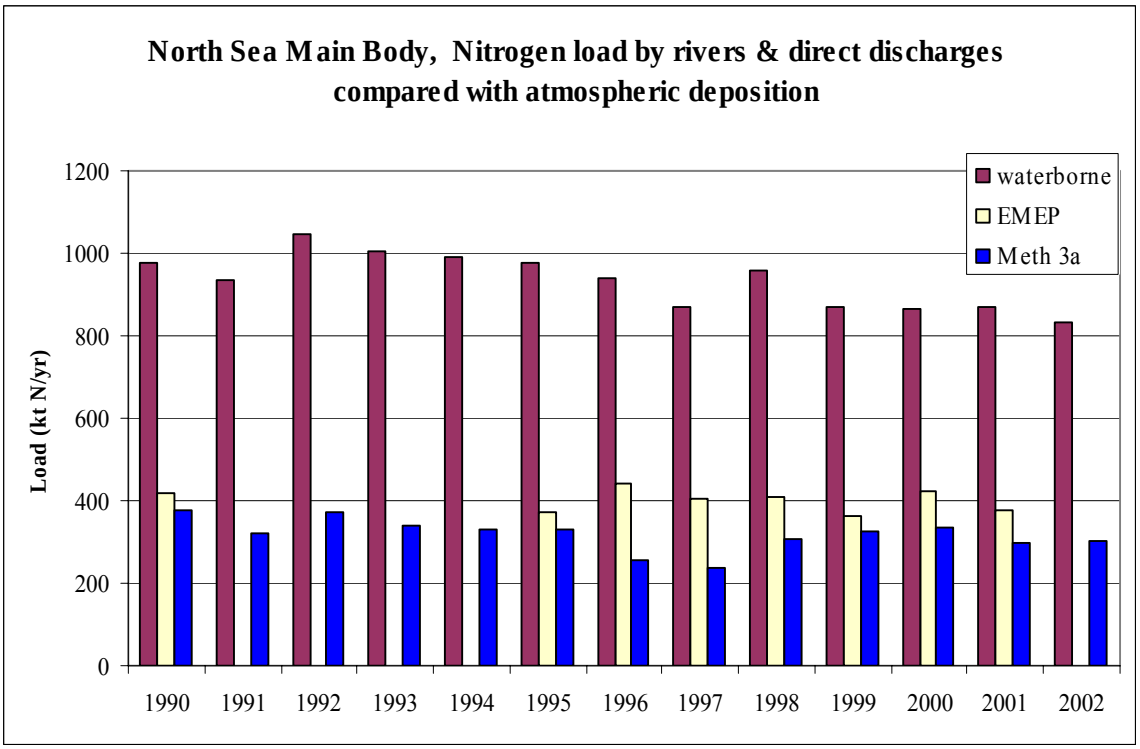


Figure 15. Total nitrogen load into of the sub-region North Sea Main Body by direct discharges and riverine inputs compared to the total atmospheric deposition based on EMEP model estimates and on estimates by "Method 3a".

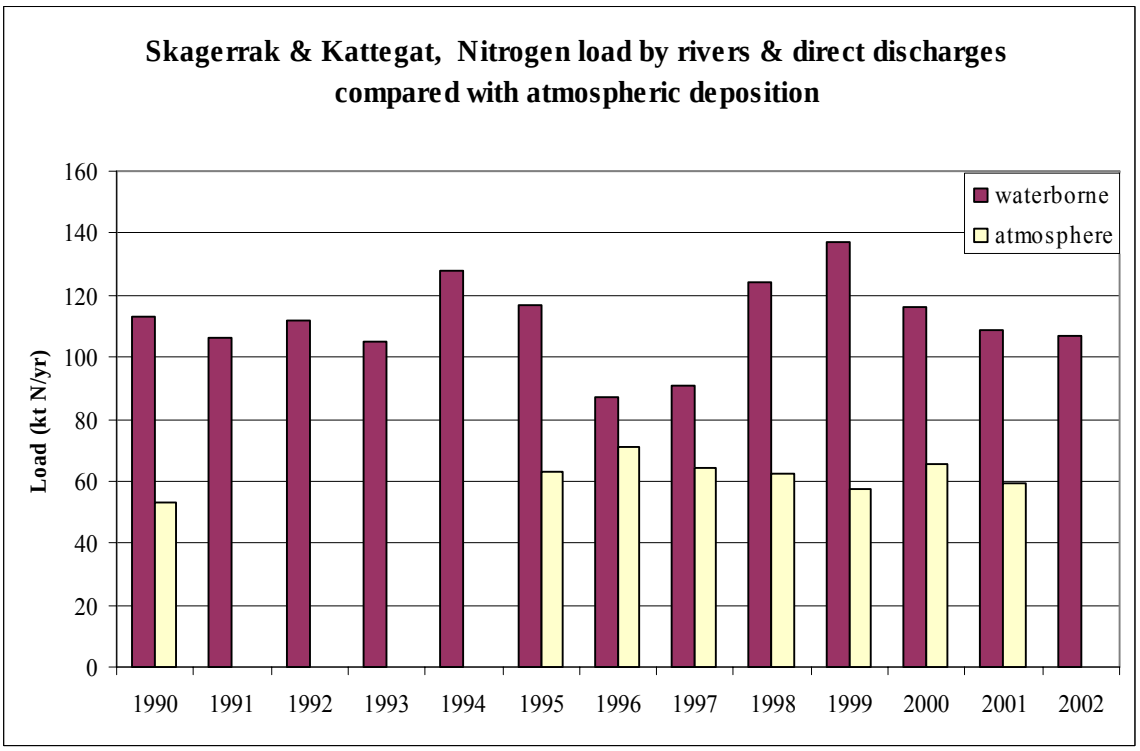


Figure 16. Total nitrogen load into the sub-region Skagerrak and Kattegat by direct discharges and riverine inputs compared to the total atmospheric deposition (EMEP model results).

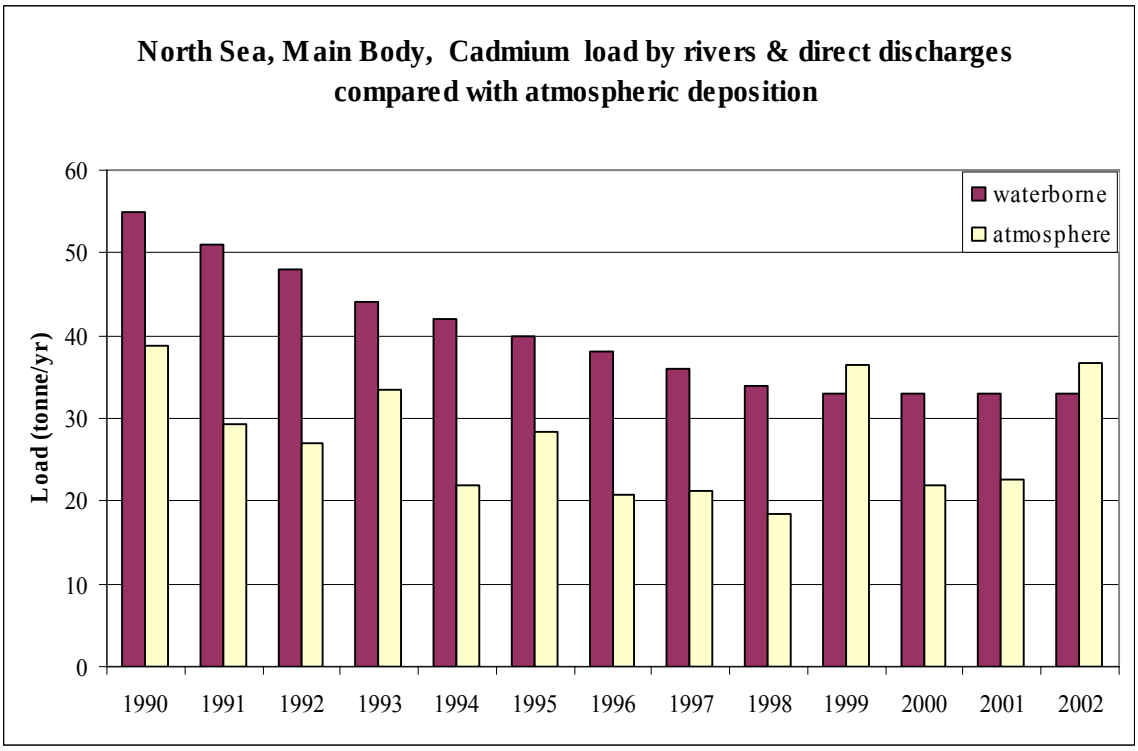


Figure 17a. Total load of cadmium into the sub-region North Sea Main Body by direct discharges and riverine inputs compared to the total atmospheric deposition ("Method 3a").

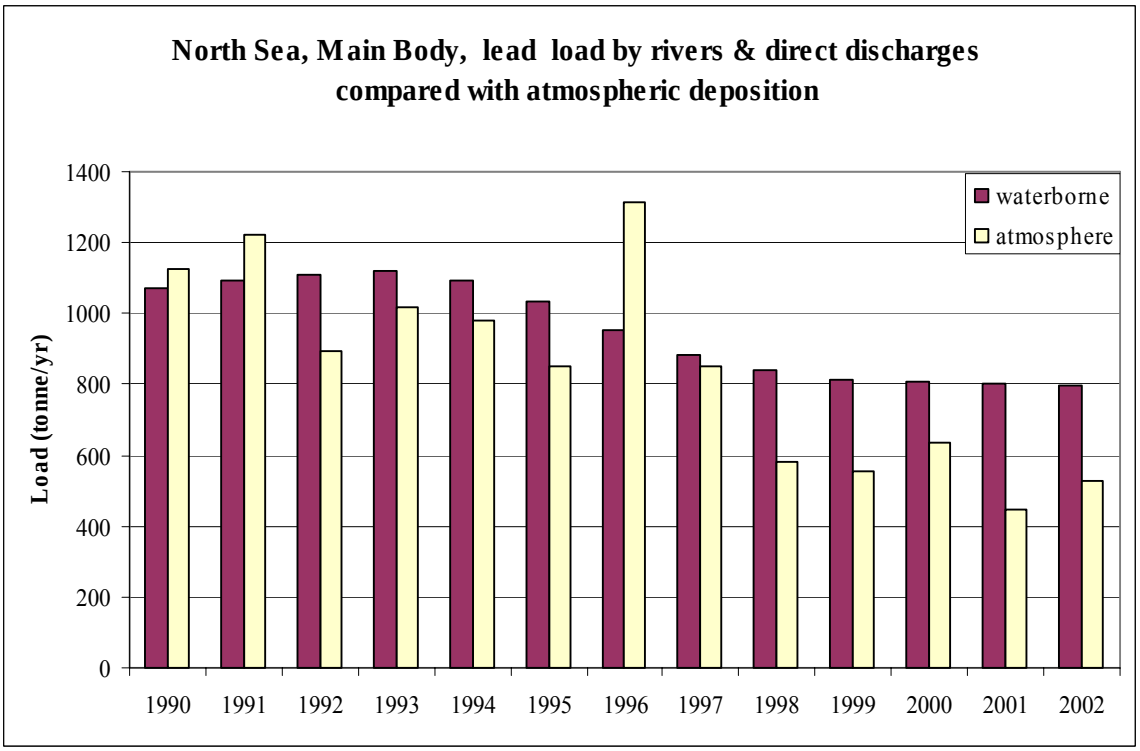


Figure 17b. Total load of lead into the sub-region North Sea Main Body by direct discharges and riverine inputs compared to the total atmospheric deposition ("Method 3a").

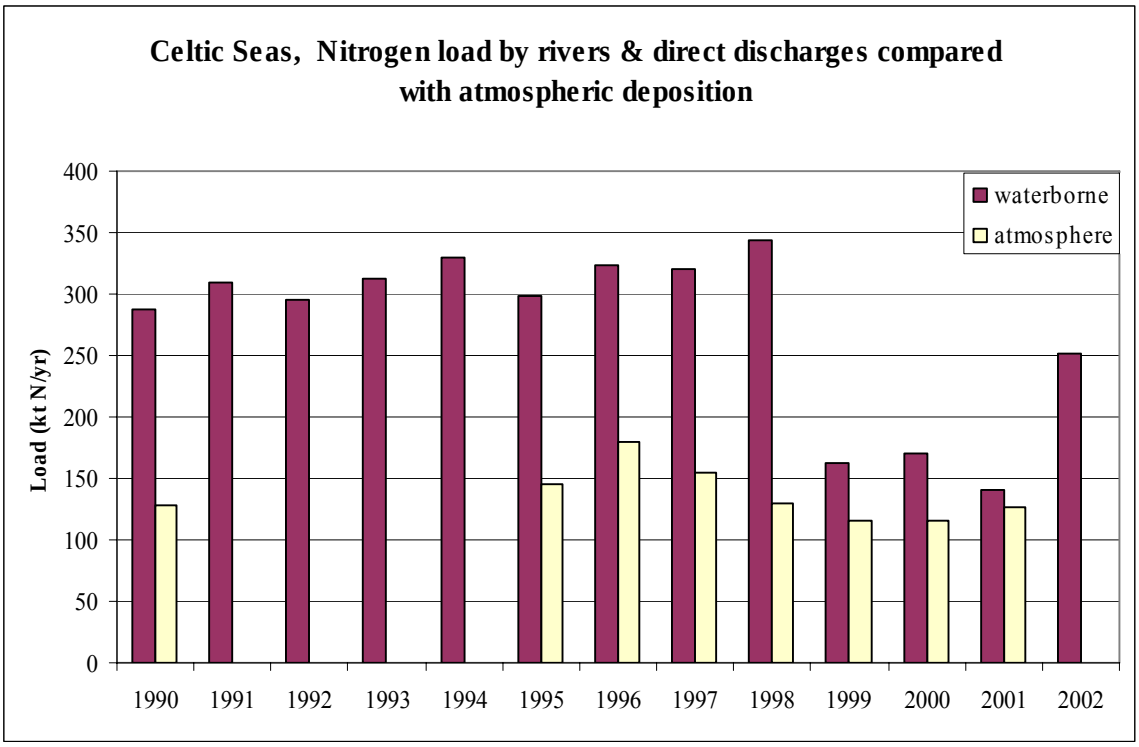


Figure 18. Total nitrogen load into the Celtic Seas by direct discharges and riverine inputs compared to the total atmospheric deposition (EMEP model results).

7. Discussion & Conclusions

CAMP network

The CAMP network was initiated in 1987 and is now more than 15 year old. It was one of the first international networks for toxic air pollutants and has given large contributions to current knowledge on transport and dispersion of these pollutants.

Over the period since 1987 the station density has varied and the coverage for various pollutants has also varied. The maps shown in Chapter 4 and 5 clearly show that there is a marked lack of observations for many pollutants for large stretches of the coastline. Some Contracting Parties have never reported on one or more of the mandatory pollutants over the whole period. In case a Contracting Party does not report on all mandatory pollutants, it is recommended that in the data reporting to OSPAR, this Contracting Party gives information on the reason for not reporting and gives an indication of when the measurements will start.

According to the CAMP principles a monitoring station should be a so-called *background station* and it should not be located more than 10 km from the coast line. Figure 19 shows – for the stations for which this information is available – the distance to the sea and the station elevation. One must conclude that one quarter of the stations are more than 10 km from the coast line and some of them are at high altitudes.

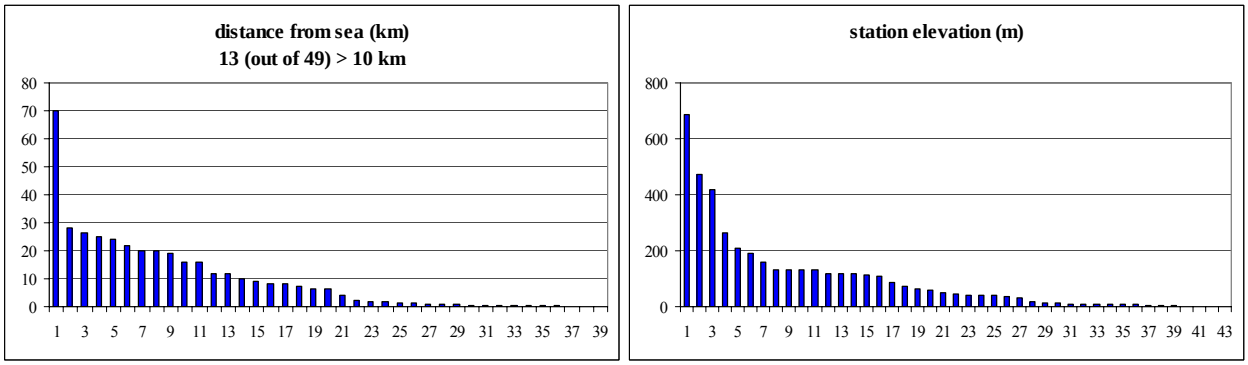


Figure 19. Distance of CAMP monitoring station to the coastline (in km) and station elevation (in m).

A priori it can not be stated that the inland and/or high altitude stations are not representative for the marine environment but a re-evaluation of the representativeness of the stations might be considered.

According to the CAMP principles each Contracting Party bordering the maritime area should have at least one CAMP monitoring station. As the objectives of CAMP are to assess the atmospheric inputs to the maritime area, it might be considered to rephrase this requirement in terms of a station density per km coastline. This will improve the representativity of the network for the maritime area. Note that the required density might be pollutant-dependant. If current concentrations are well below the levels at which effects are expected, a lower density can be accepted. Both as a result of a revision of the EMEP monitoring strategy (Tørseth and Hov, 2003) and the introduction of the 4th Daughter Directive on As, Cd, Hg, Ni and PAH (EU, 2005) national monitoring networks might be reconstructed. In this action the requirements of CAMP should be considered as well.

Evaluating the CAMP data shows two typical problems with measuring heavy metals in precipitation. The first is a wide scatter in results and the frequent observation of outliers. It is known that precipitation measurements of heavy metals are frequently contaminated (Ross, 1986; Slanina *et al.,* 1990). By installing three or more samplers at one station, outliers can be easily detected. Installing more samplers would strongly reduce the measurement error but it would also increase the operational cost. This might partly be compensated for by allowing a longer sampling period which would reduce the number of samples to be analysed. The second problem is the frequent occurrence of concentrations below the detection limit. If this is the case a reliable annual mean can not be calculated. A remedy might be either to improve the detection limit or to stop the monitoring as the data have only limited value. Such an evaluation depends on the ratio between current ambient levels and the levels at which adverse effects on the marine ecosystem are expected. Based on an ecotoxicological assessment, one might define a target value for concentrations in precipitation or air in such a way that if ambient levels are below this target value no adverse effects are expected. The monitoring strategy will depend on the ratio of ambient and target value:

- When the ambient levels are well below the target value, there is less urgency to monitor. It is sufficient to perform indicative measurements to ensure that ambient levels are not increasing. Spatial and/or temporal resolution of the measurements can be reduced. If the detection limit is below the target value, there is no need to improve it.
- When ambient levels are around or above the target values, the monitoring strategy should be adjusted in such a way that an assessment of the atmospheric input, as accurate and relevant can be made.

Database

Most of the data is now relatively easily available from the database hosted by NILU. However, not all data reported under CAMP is available in the database .In particular data from the early years is missing. It might be considered, with assistance of Contracting Parties, to complete the database.
Meta-information on the stations (that is, documentation on station location (latitude, longitude, altitude), the institute responsible for the station management, information on measurement method, information describing the surroundings of the station etc; see for example the *Exchange of Information* decision of the European Commission (2001, 1997) for relevant items) is not archived on a central place. For a number but not for all stations this information is available either from the EMEP database (http://www.nilu.no/projects/ccc/) or from AirBase (http://airbase.eionet.eu.int/). It is recommended to develop and maintain a database on meta information.

The observed time series both for precipitation and aerosols show in general a downward tendency although a significant trend is seen only for about half of the analysed time series. In more detailed look, sometimes inconsistencies between precipitation and aerosol data can be observed, for example, the trends in arsenic concentration in precipitation and on aerosol for two stations in the UK have opposite sign. The reason for this different behaviour is not clear. By visual inspection of the time series suspicious data can be detected; checking the data quality flags showed that the date is marked as valid by the data supplier. As the trend results are sensitive for these outliers a re-examination of the data is recommended.

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Annex 1 Atmospheric emission of nitrogen oxides and ammonia

Table 1. Annual nitrogen oxides emissions (as NO_2) from the OSPAR Contracting Parties. Emission from three selected countries (Italy, Russian Federation and Poland) with the largest deposition of nitrogen in the OSPAR Convention Waters, are also included. Unit: kt N a^{-1}.

Country	Year											
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Belgium	102	99	102	100	101	109	96	93	95	88	100	96
Denmark	84	99	85	86	87	82	95	82	74	69	64	62
Finland	91	88	86	86	86	79	82	79	77	75	72	68
France	577	598	584	547	532	520	511	490	485	462	439	429
Germany	830	765	707	672	625	604	577	543	510	493	482	485
Iceland	8	8	9	9	9	9	9	9	9	9	9	9
Ireland	36	37	40	33	35	35	37	36	37	36	38	38
Luxembourg	7	7	7	8	7	6	7	5	5	5	5	5
Netherlands	173	173	169	163	155	148	152	138	130	131	126	125
Norway	68	65	65	68	67	67	70	71	72	72	68	67
Portugal	83	87	94	92	93	97	96	98	105	111	117	115
Spain	367	380	389	381	383	387	375	389	387	400	406	397
Switzerland	47	44	42	39	38	37	34	33	32	30	29	28
Sweden	102	102	97	93	97	90	90	85	81	79	77	75
UK	840	801	771	720	700	662	659	612	584	551	529	511
Italy	590	604	612	606	544	538	531	506	485	452	418	418
Poland	390	367	344	341	336	341	351	339	302	289	255	245
Russia	1096	1045	950	929	812	782	751	724	757	759	717	717

Table 2. Annual ammonia emissions (NH_3) from the OSPAR Contracting Parties. Emission from three selected countries (Italy, Russian Federation and Poland) with the largest deposition of nitrogen in the OSPAR Convention Waters, are also included. Unit: kt N a^{-1}.

Country	Year											
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Belgium	82	77	77	80	79	82	82	82	84	82	67	67
Denmark	110	106	105	102	99	93	90	90	91	86	86	84
Finland	31	33	34	32	30	29	29	31	31	29	27	27
France	642	637	630	623	628	631	640	645	646	648	646	642
Germany	606	538	524	506	490	497	501	493	497	497	491	500
Iceland	2	2	2	2	2	2	2	2	2	2	2	2
Ireland	92	95	96	96	98	99	100	101	105	105	100	100
Luxembourg	6	6	6	6	6	6	6	6	6	6	6	6
Netherlands	191	188	48	157	137	159	120	155	140	137	125	122
Norway	19	19	21	20	21	21	22	21	21	21	21	21
Portugal	87	84	88	82	77	82	80	79	81	85	84	84
Spain	269	260	259	244	260	250	278	278	293	303	303	313
Switzerland	59	58	58	58	58	57	57	57	56	56	56	56
Sweden	44	45	45	51	51	51	51	49	49	47	47	44
UK	281	282	270	270	271	263	265	268	264	260	245	239
Italy	384	371	362	370	378	380	354	365	361	369	360	360
Poland	418	371	368	315	316	313	300	288	306	281	265	254
Russia	981	956	893	744	636	679	617	601	556	541	535	535

Annex 2 Atmospheric emissions of heavy metals

Atmospheric emissions of arsenic, cadmium, chromium, copper, mercury, nickel, lead, and zinc in Contracting Parties in 1990, 1995 and 2000. Units: tonne per year.

	Arsenic			Cadmium		
	1990	1995	2000	1990	1995	2000
Belgium	5.7	5.7	2.6	7.8	5.1	2.4
Denmark	1.45	0.9	0.9	1.2	0.9	0.6
Finland	33.2	3.5	4.6	6.3	1.7	1.4
France	28	25	25	16	13	10
Germany	122	32	32	31	11	11
Iceland	0.134	0.134	0.134	0.166	0.166	0.166
Ireland	2.3	1.6	0.9	1.6	1.04	0.49
Luxembourg	0.935	0.507	0.079	0.6	0.4	0.051
Netherlands	1.46	1.19	1.24	1.95	1.01	1.15
Norway	3.10	2.90	2.46	1.64	1.01	0.73
Portugal	2	2	2	2	2	1
Spain	35	43	55	14.9	16.7	19.8
Sweden	5.5	1.4	0.7	2.47	0.76	0.52
Switzerland	1.25	1.25	1.25	4.2	2.5	2.18
UK	62	46	28	20.3	12	7.25
Total OSPAR	242	121	129	91.9	57.3	51.5

	Chromium			Copper		
	1990	1995	2000	1990	1995	2000
Belgium	32.0	27.0	17.0	34	29	24
Denmark	6.5	4.4	1.6	10	10	8.6
Finland	31.6	21.7	28.0	94.4	26.7	18.7
France	396	211	259	168	175	177
Germany	253	115	115	361	79	79
Iceland	0.4	0.4	0.4	2.14	2.14	2.14
Ireland	5.18	4.1	2.9	10.2	9.4	8.6
Luxembourg	3.3	1.8	0.3	14.7	8.0	1.3
Netherlands	11.2	8.2	5.5	19.4	20.5	21.2
Norway	12.8	11.4	8.8	21.9	18.6	19.3
Portugal	5.0	6.0	6.0	13	15	18
Spain	38.9	44.0	50.0	145	158	225
Sweden	22.9	12.2	6.7	34.32	19.21	15.00
Switzerland	6.7	6.7	6.7	17.8	17.8	17.8
UK	174.0	122.0	72.0	126.9	87.9	48.2
Total OSPAR	826	474	508	945	588	636

	Mercury			Nickel		
	1990	1995	2000	1990	1995	2000
Belgium	8.86	3.76	2.88	76	76	49
Denmark	3.46	2.66	1.16	25	22	12
Finland	1.1	0.7	0.58	67	34	33
France	25.3	21.0	13.4	318	259	222
Germany	113	31	28.7	278	158	158
Iceland	0.05	0.05	0.05	4.73	4.73	4.73
Ireland	1.6	1.57	1.53	37	31.6	26.2
Luxembourg	0.3	0.1	0.27	14.7	7.7	0.68
Netherlands	3.02	1.06	0.6	76	87	53
Norway	1.7	1.09	1	37	37	37
Portugal	0.23	0.23	0.23	69	69	58
Spain	21.5	21.1	23.4	266	325	323
Sweden	4.73	1.09	0.77	30	32	16
Switzerland	6.8	3.3	2.63	27.5	27.5	27.5
UK	36.68	22.24	9.04	393	300	108
Total OSPAR	191.6	88.7	77.1	1325	1171	1020

	Lead			Zinc		
Country	1990	1995	2000	1990	1995	2000
Belgium	566	361	114	219	181	167
Denmark	123	18	6.9	36	25	23
Finland	326	57	38	571	322	70.7
France	4264	1446	247	2031	1371	1442
Germany	2323	632	519	1323	451	451
Iceland	12	3.9	0.4	0.4	0.4	0.4
Ireland	130	81.5	32.91	35.8	23.8	14.7
Luxembourg	77	30	1.6	67.4	52.2	37
Netherlands	335	159	44	220.9	143.9	126
Norway	186	21	6.2	109	109	109
Portugal	631	631	485	25	30	35
Spain	2833	975	672	1380	1557	2025
Sweden	474	23	15	178	129	94
Switzerland	520	226	114	841	607	558
UK	2809	1549	184	1229	1096	650
Total OSPAR	12800	4664	2297	7041	5007	5157

Annex 3 Emissions of γHCH

Emissions of γ-HCH in Contracting Parties for 1980, 1985, 1990 , 1996 in tonne per year, data taken from Holoubek et al. (2000), see http://www.recetox.muni.cz)

	year			
Country	1980	1985	1990	1996
Belgium	19.1	23.0	15.4	15.9
Denmark	4.3	2.9	2.9	0
Finland	5.0	1.8	0	0
France	322	294	270	560
Germany	111	49.6	27.6	0
Iceland	0.035	0.039	0.073	0.109
Ireland	2.2	2.5	1.8	2.2
Luxembourg	0.317	0.191	0.135	0.151
Netherlands	32.6	36.6	3.8	6.3
Norway	1.1	1.2	0.13	0
Portugal	1.6	2.3	3.4	13.2
Spain	221	275	180	123
Sweden	3.1	1.6	2.7	1.1
Switzerland	2.8	1.5	0.016	0.159
UK	56.2	17.2	31.1	30.0

Annex 4 Observed concentrations of heavy metals in precipitation

Annual mean concentrations of heavy metals in precipitation (µg/l). These are precipitation weighted values; precipitation amounts are given in mm. In this table the following color code is used:

White:	no data reported
Yellow (light grey):	valid data
Red (dark grey):	invalid data (less than 75% data coverage, either by data losses or because concentration were below reported detection limits)

				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
BE0004R	WET ONLY	1996	604	2.436	0.465	7.112	7.419	44.14	483.3	6.246	192.8
		1997	542	4.394	0.264	2.319	3.584	45.5	25.55	2.731	60.74
		1998	984	0.724	0.252	5.87	2.806	33.14	30.17	2.294	49.15
		1999	852		0.303	2.699	17.82	27.11	34.06	4.208	27.17
		2000	810	2.5	1.4	7.08	4.066	28.14	33.09	4.715	22.61
		2001	750		0.567	0.954	6.402	27.91	61.6	1.402	34.48
		2002	823		0.663	1.529	10.59	6.377	37.45	2.381	59.85
BE0004R	BULK	1996	464	0.5	0.28		21.47	6.885			37.96
		1997	914	2.625	7.072		5.277	18.34			38.81
		1998	1386		0.538		9.345	8.922			51.69
		1999	1158		0.808		72.25	4.195			201
		2000	1174	1.035	0.365		4.949	6.873			180.1
		2001	973		0.315			3.34			22
		2002	892		0.635		2.09	2.884			47.67
DE0001R	WET ONLY	1989	478								
		1990	692	0.311	0.149	0.487	1.052	2.429	1.918	0.707	11.47
		1991	571								
		1992	333	0.162	0.062	0.052	1.633	1.343	2.164	0.592	12.68
		1993	440	0.235	0.066	0.056	1.008	3.292	13.73	0.441	6.078
		1994									
		1995	657				2.309	1.653		0.938	14.09
		1996	336	0.428	0.111	0.364	3.13	43.58	30.92	0.458	11.92
		1997	402	0.318	0.057	0.407	1.957	1.861	17.54	0.644	10.47
		1998	703	0.145	0.108	0.156	3.202	1.554	9.314	1.043	17.37
		1999	764	0.108	0.058	0.148	2.495	1.091	12.72	0.844	16.77
		2000	661	0.151	0.066	0.182	0.103	1.093	9.551	0.75	24.68
		2001	657	0.116	0.032	0.15	1.447	0.997	6.344	0.277	6.634
		2002	784	0.153	0.039	0.11	1.402	1.169		0.375	12.93
DK0008R	BULK	1999	760	0.237	0.05	0.237	1.481	1.798		0.364	11.85
		2000	670	0.315	0.056	0.265	1.351	2.332		0.325	11.47
		2001	569	0.312	0.056	0.235	0.949	1.405		0.303	8.568
		2002	718	0.266	0.052	0.199	0.973	1.568		0.268	7.915
DK0031R	BULK	1989	732	0.451	0.117	0.479	1.303	4.157		0.463	15.93
		1990	1063		0.096	0.276	0.995	2.73	11.53	0.499	9.129
		1991	705						14.17		
		1992	905		0.06	0.469	1.84	2.285		0.448	12.15
		1993	819		0.062	0.456	1.216	2.091		0.376	10
		1994									
		1995	754		0.063	0.396	1.818	2.102		0.393	18.22
		1996	660		0.08	0.374	1.304	1.923		0.457	15.84
		1997	690		0.064	0.575	1.781	2.259		0.887	15.39
		1998	941	0.169	0.044	0.316	0.929	1.143		0.314	10.14

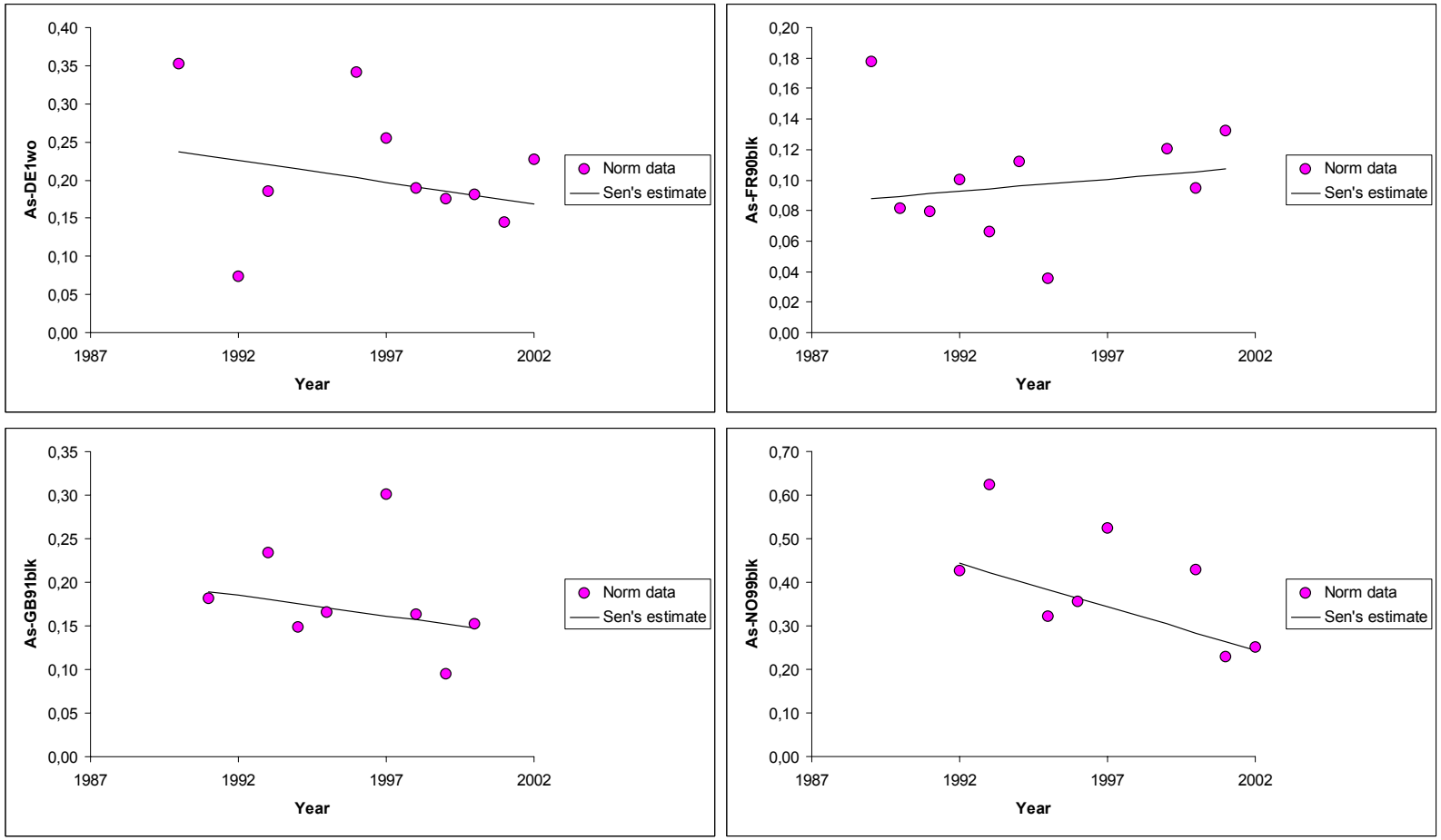
				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
		1999	1067	0.078	0.032	0.102	0.617	0.895		0.219	6.901
		2000	837	0.143	0.041	0.101	0.526	0.863		0.198	7.926
		2001	1017	0.121	0.029	0.096	0.633	0.692		0.225	8.419
		2002	1034	0.113	0.034	0.151	0.706	0.79		0.245	7.33
FR0090R	BULK	1989	489	0.218	0.142	0.201	3.153	3.026		0.574	11.04
		1990	685	0.1	0.037	0.101	0.602	1.532		0.465	4.754
		1991	707	0.095	0.069	0.378	1.437	2.515		1.089	12.22
		1992	650	0.122	0.022	0.114	0.579	1.155		0.43	4.127
		1993	684	0.084	0.029	0.229	0.712	1.775		0.563	3.668
		1994	851	0.111	0.02	0.076	0.487	1.137		0.353	2.401
		1995	790	0.042	0.024	0.069	0.432	1.101		0.492	3.002
		1996									
		1997									
		1998									
		1999	1088	0.093	0.019	0.156	0.921	1.862		0.303	2.468
		2000	1322	0.04	0.024	0.183	0.855	1.815		0.709	2.262
		2001	1168	0.096	0.033	0.063	0.919	1.069		0.541	1.717
		2002	964	0.433	0.029	0.154	0.683	1.015		0.42	2.818
GB0013R	BULK	1996	702								
GB0014R	BULK	1993	907.8								
		1994	723.8	0.431	0.113	0.294	1.874	5.452		0.956	29.56
		1995	609.8	0.318	0.331	0.641	2.881	4.727		1.28	16.58
		1996	466.9								
		1997	791	0.329	0.053	0.163	1.324	3.855		0.184	28.32
		1998	977.2	0.195	0.051	0.389	0.945	2.553		0.389	9.93
		1999	948.6	0.238	0.071	0.207	1.415	2.456		0.314	5.064
		2000	1155	0.143	0.049	0.182	1.128	2.579		0.301	9.607
		2001									
		2002	881.3			0.335	1.529	853		0.282	11.21
GB0090R	BULK	1987	660				2.249				9.09
		1988									
		1989	518				4.747	7.583		1.414	21.03
		1990	529.5	0.48	0.396	0.75	1.998	4.935	60	1.46	16.85
		1991	428.6	0.422	0.165	0.469	5.212	7.67	77.91	1.809	
		1992	812.7	0.417	0.216	0.307	1.948	5.2	40.59	1.604	18.63
		1993	822.2	0.279	0.26	0.169	1.639	4.148		2.343	13.25
		1994	764	0.249	0.204	0.235	1.963	4.294		1.298	23
		1995	663	0.361	0.114	0.378	1.743	4.432		1.613	12.91
		1996	580.3	0.271	0.213	0.165	7.475	3.489		1.107	57.18
		1997	634	0.27	0.064	0.471	3.347	3.85		0.503	9.824
		1998	770.4	0.146	0.068	0.338	1.206	2.084		0.493	8.718
		1999	830.5	0.128	0.045	0.293	1.28	1.887		0.345	5.515
		2000	753.3	0.311	0.088	0.223	1.849	1.665		0.368	26.23
		2001									
		2002	662.7	0.101	0.096	0.107	1.253	1.468		0.521	17.98
GB0091R	BULK	1989	543.1	1.056	0.122	0.687	3.56	3.945	222	0.6	9.605
		1990	693	0.409	0.134	0.265	1.926	3.039	315.1	0.74	8.99
		1991	742	0.258	0.138	0.264	4.282	3.966	65.8	0.688	

				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
		1992	780.6	0.165	0.054	1.324	2.363	2.492	37.88	0.584	5.884
		1993	941.5	0.213	0.108	0.256	1.841	2.464		0.478	11.56
		1994	680.4	0.256	0.073	0.231	1.241	3.147		0.646	22.58
		1995	947.8	0.142	0.074	0.691	4.863	1.988		0.761	9.824
		1996	558	0.03	0.08	1.8	10.5	1		0.06	10.5
		1997	885.1	0.308	0.252	0.295	1.145	2.784		0.2	5.214
		1998	1038	0.095	0.052	0.598	0.857	0.899		0.187	11.35
		1999	895.2	0.096	0.06	0.207	1.177	0.905		0.298	3.229
		2000	1060	0.074	0.043	0.249	1.601	0.896		0.358	9.326
		2001									
		2002	984.2	0.134	0.163	0.065	0.643	1.089		0.209	10.89
GB0092R	BULK	1993	621.9	0.122	0.08	0.118	2.119	5.248		0.412	8.947
		1994	1118	0.277	0.085	0.14	0.74	4.441		0.516	23.09
		1995	808	0.193	0.088	0.28	1.808	4.216		0.647	8.904
GB0093R	BULK	1987	693.2		0.635	1.727	6.196	28.12	86.2	2.746	50.78
		1989	495	1.477	0.371	0.865	7.273	16.15	207.3	3.094	40.86
		1990	708	0.857	0.359	0.838	3.67	11.17	273.2	2.161	30.82
		1991	536.4	1.18	0.462	0.946	7.584	18.98	97.2	5.49	
		1992	342.5	0.632	0.304	0.558	2.565	8.981	61.88	2.151	35.74
GB0094R	BULK	1989	1121	1.479	0.844	0.853	3.762	1.246		0.438	6.761
		1990	1524	1.297	0.19	0.28	1.964	0.875		0.615	7.867
		1991	1172	0.181	0.076	0.231	1.938	1.406		0.424	
		1992	1454	0.194	0.111	0.095	1.21	0.852		0.373	7.389
		1993									
GB0096R	BULK	1994							18.4		
IE0001R	BULK	1992	1224	0.2	0.344	0.503	3.98	1.258	100		22.05
		1993	1471		0.19	0.278	5.973	1.664			15.44
		1994	1808		0.093	0.752	3.033	18.38		0.9	11.47
		1995	1542	0.25	0.26		4.164	1.961		1.954	14.84
		1996	1568		0.23		7.098	1.382			13.82
		1997	1406		0.148		5.15	6.762		1.5	21.74
		1998	1782		0.297		5.099	1.921	100		59.26
		1999	1776	0.6	0.07	0.6	0.852	0.681			15.04
		2000	1769		0.07		5.78	1			32.59
		2001	1261				2.278				37.94
		2002	1919				3	2			43.05
IE0002R	BULK	1992	1127		0.12	0.748	10.19	2.235			10.53
		1993	1443		0.076	0.511	3.812	1.134	60		9.74
		1994	1413		0.365	0.478	6.496	1.623	118.3	1.4	10.67
		1995	1284		0.312		9.892	1.223		3.534	12.82
		1996	1970		0.151		2.845	1.623			19.99
		1997	1683		0.085		1.564	1.766		1.287	16.99
		1998	1922		0.089		0.984	1.156		1	2.628
		1999	1744	0.6	0.083	0.5	0.951	0.951		0.525	4.373
		2000	1907			1.169	9.144	1.161		0.795	10.49
		2001	1434		0.133		13.48	1.806			8.894
		2002	2277			3.458	3.266	2			7.815
IS0002R	WET ONLY	1992	1706		0.269	0.466	6.158	5.071		0.598	15.92

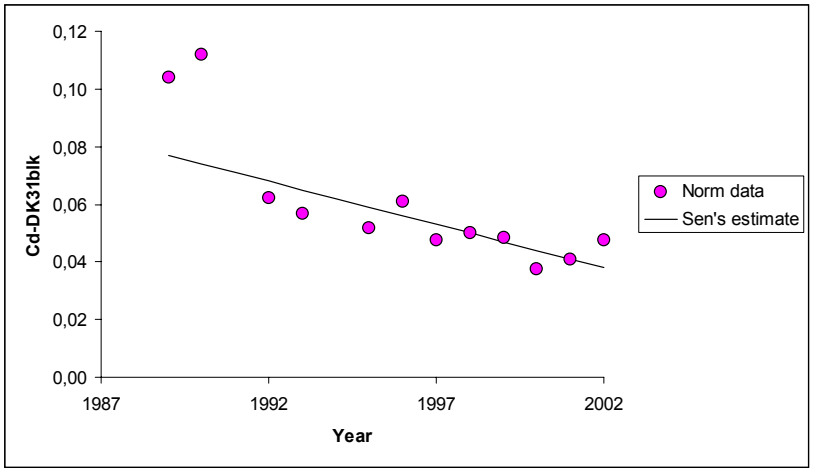
				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
		1993	2199		0.083	0.771	1.899	1.361		0.704	28.89
		1994	1640		0.799	0.182	1.679	2.547		4.402	17.78
		1995	1526		0.245	0.159	4.272	3.514	459	6.232	26.98
		1996	1893		0.199	0.29	1.675	2.045		1.287	17.68
		1997	1784		0.122	0.249	1.495	1.695		1.7	18.94
		1998	1671	0.067	0.057	1.196	3.624	1.154		0.951	159.4
		1999	1218	0.086	0.029	0.787	1.544	1.396		0.413	11.6
		2000	1519	0.061	0.028	1.117	0.998	0.542		0.537	17.41
		2001	681.3	0.162	0.025		0.946	0.465		0.186	10.29
IS0090R	BULK	1992	839.6		0.064	0.374	1.925	2.271		1.018	179.2
		1993	863.8		0.05	0.409	1.675			1.062	155.7
		1994	744		0.105	0.102	2.619	1.187		0.617	163
		1995	610.2		0.063	0.214	2.953	1.049		1.196	183.7
		1996	781.9		0.066	0.344	1.879			0.724	140.6
		1997	838.3		0.19	0.707	1.672			0.781	130.9
		1998	748.7	0.194	0.154	0.665	2.577	0.356		0.485	81.51
		1999	604.5	0.101	0.02	0.515	2.292	0.737		1.44	11.09
		2000	758.2	0.081	0.027	0.489	1.654	0.652		0.781	4.233
		2001	836.3	0.242	0.029	0.388	2.714	0.549		0.789	6.573
		2002	1025	0.25	0.027	1.176	2.025	0.615		1.638	5.549
IS0091R	BULK	2002	1556		0.03	0.984	1.897	0.399		1.566	9.089
NL0009R	WET ONLY	1990	697.5		0.208		2.733	3.895		0.754	15.06
		1991	522.4		0.157		3.449	0.775		1.161	14.99
		1992	729.2		0.126		1.786	2.765		0.916	10.25
		1993	833.3		0.147		1.729	4.199			11.69
		1994	793		0.125		2.873	3.106			14.29
		1995	836.6		0.078		2.651	2.454			9.159
		1996	492.3		0.238		1.835	2.038			15.2
		1997	519		0.177		1.164	2.129			13
		1998	945.7		0.121		1.187	2.026		0.552	10.58
		1999	831.3	0.56	0.134		1.771	1.654		0.572	9.597
		2000	642.7	0.277	0.081	0.536	2.047	2.139		0.519	7.773
		2001	792.7	0.263	0.092		2.911	1.654			8.21
		2002	983.7	0.22	0.063	0.53	1.499	1.282			6.034
NL0091R	WET ONLY	1996	543.4	1.423	0.13	0.676	1.916	3.596	26.79	1.028	9.924
		1997	638.4		0.107		1.731	3.228	17.4	1.407	10.92
		1998	828.1		0.083		1.415	2.653	13.64	0.63	9.752
		1999	993.6		0.067		1.496	2.705	11.1	0.527	7.154
		2000	1026	0.218	0.058		1.7	3.062	9.796	0.652	6.213
		2001	961.9	0.155	0.067		1.991	3.161	9.402	0.538	8.347
		2002	855.5	0.166	0.058		1.598	3.061	9.322	0.465	6.766
NO0001R	BULK	1987	1576		0.121						8.97
		1988									
		1989									
		1990	1792		0.249			3.785			9.629
		1991	1158		0.072			3.606			6.948
		1992	1355		0.054			2.847			5.197
		1993	1207		0.072			3.116			6.474

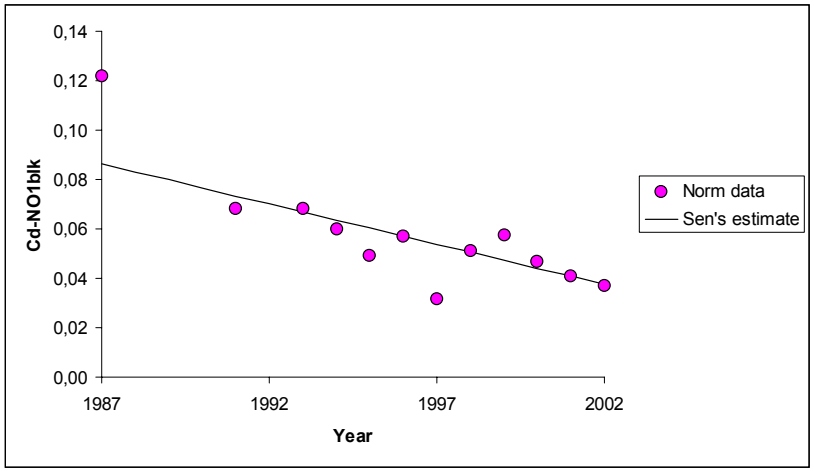
				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
		1994	1406		0.061			2.628			5.011
		1995	1210		0.053			2.339			6.586
		1996	1130		0.062			2.824			4.931
		1997	1187		0.036			1.734			4.158
		1998	1531		0.05			1.592			4.935
		1999	1871		0.052			1.499			4.383
		2000	2253		0.036			1.39			3.225
		2001	1580		0.039			1.255			4.673
		2002	1450		0.037			0.986			3.604
NO0039R	BULK	1987	1463		0.03			1.095			2.407
		1988	1549		0.063			0.897			4.274
		1989	1537		0.054			0.342			1.752
		1990	1520		0.059			0.233			1.202
		1991	1620		0.014			0.319			0.984
		1992	1619		0.01			0.24			0.944
		1993	1448		0.016			0.209			0.695
		1994	1474		0.02			0.399			1.172
		1995	1660		0.016			0.229			1.156
		1996	1170		0.018			0.499			1.353
		1997	1843								2.331
NO0099R	BULK	1990	1565		0.32	1.362	5.75		13.75	2.096	7.624
		1991	1031	2.278	0.061	0.761	2.464		11.83	1.163	14.19
		1992	1376	0.328	0.07	2.966	1.805		10.93	0.999	7.936
		1993	846	0.724	0.114	2.868	1.898		11.27	0.724	13.06
		1994	1202	0.287	0.064	0.482	1.197	2.647	8.092	0.591	7.795
		1995	899.1	0.403	0.057	1.849	1.046	2.338	13.95	0.577	8.545
		1996	886	0.442	0.082	0.428	1.088	3.02	19.67	0.492	8.61
		1997	1219	0.485	0.054	0.48	0.979	2.77	10.63	0.591	6.587
		1998	1093	0.342	0.048	0.649	1.124	2.091	9.03	0.727	8.266
		1999	1273	0.337	0.039	0.49	1.82	1.496	9.71	0.668	7.436
		2000	1494	0.287	0.039	0.305	1.13	1.574	7.318	0.484	6.574
		2001	1210	0.192	0.07	0.418	1.276	1.515	7.297	0.566	7.44
		2002	985	0.299	0.042	0.364	1.304	2.145	12.81	0.62	6.835
PT0003R	BULK	1987	569.2								
		1988									
		1989	1135				1.742	2.734		2.771	51.75
		1990	781.9				2.471	2.047		2.754	37.69
		1991	1146		4.27		2.915	5.032		2.445	19.17
		1992	903.6				1.596	2.92		2.318	15.65
		1993	1151		20.75		2.727	4.174		3.838	26.1
		1994	1495				2.452	2.587		2.296	19.86
		1995	1421				2.124	2.144		2.632	31.46
		1996	1752		2.53		2.66	1.723		2.387	51.06
		1997	1510		1.412		2.323	2.4		2.59	33.31
		1998	887.5		1.277		2.466	1.932		1.911	27.06
		1999	1496		1.88		2.558	2.19		2.348	14.81
		2000	1891		1.28		2.148	2.554		1.44	11.39
		2001	1850				4.53	1.904		2.23	18.57

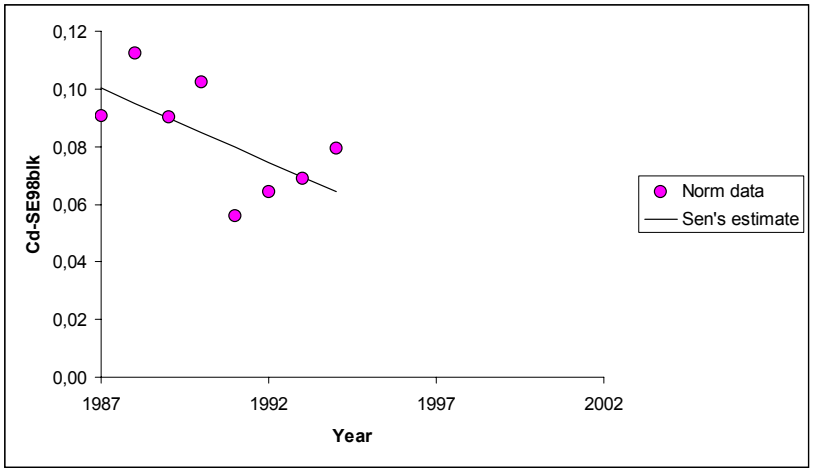
				annual mean concentrations (ug/l)							
Code	sampler	Year	precip	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
		2002	1730				2.165			2.062	15.26
PT0004R	BULK	1997	793.9		1.726		1.148	1.69		1.93	26.46
		1998	206.4		3.03		1.453	1.385		1.843	16.75
		1999	560		1.285		1.416	1.755		2.488	12.68
		2000	574.4				1.366	1.674			8.632
		2001	501.1				1.107			5.48	8.082
		2002	694.5				1.265			2.49	5.745
PT0010R	BULK	1995									
		1996									
		1997									
		1998									
		1999	943.6		2.028		2.041	3.592		4.577	59.91
	WET ONLY	2000	933.5				1.687	2.715			35.96
		2001	1228				2.029	5.253		9.915	22.83
		2002	1065				1.846	1.832		10.84	44.21
SE0002R	BULK	1989	584						30.09		
		1990	853						18.9		
		1991	647						15.33		
		1992	779						11.42		
		1993	653						9.925		
		1994	755						9.748		
		1995	325						21.21		
		1996									
		1997									
		1998	626						6.957		
		1999	692						9.433		
		2000	771.6						8.755		
		2001	579						9.314		
SE0014R	BULK	2002	581.6						12.26		
SE0097R	BULK	1993	842						9.809		
		1994	1117						10.37		
		1995	961.5	0.228	0.07	0.15		2.405	12.04	0.293	11.66
		1996	685	0.188	0.069	0.249		2.521		0.304	9.329
		1997	906	0.219	0.058	0.333	4.558	1.998		0.274	9.483
		1998	807	0.182	0.041	0.125	2.15	1.546		0.215	12.84
		1999	1079	0.102	0.05	0.243		1.645		0.217	11.73
		2000									
		2001	676	0.175	0.043	0.21	1.023	1.362		0.271	11.54
		2002	988	0.164	0.038	0.257	0.801	0.956		0.115	4.602
SE0098R	BULK	1987	782		0.101	0.146	1.922	2.631		0.524	8.861
		1988	857	0.363	0.112	0.2	6.338	3.003		0.403	10.03
		1989	614	0.327	0.125	0.155	1.214	2.861		0.461	11.14
		1990	985	0.243	0.083	0.106	0.853	2.135		0.32	8.632
		1991	736	0.258	0.073	0.302	1.122	2.928		0.347	8.205
		1992	986	0.179	0.045	0.458	0.861	2.138		0.423	5.236
		1993	896	0.244	0.063	0.23	3.245	2.772		0.273	7.112
		1994	982	0.21	0.061	0.252	2.434	2.383		0.331	5.453
		1995	949	0.19	0.052	0.238		2.166		0.285	7.946

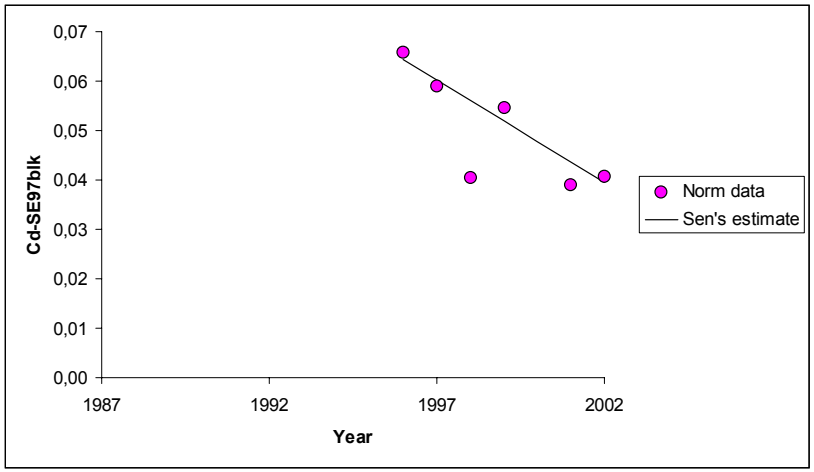


Time series of arsenic in precipitation, concentrations (g/l) normalised against precipitation amount.
DE0001 FR0090
GB0091 NO0099

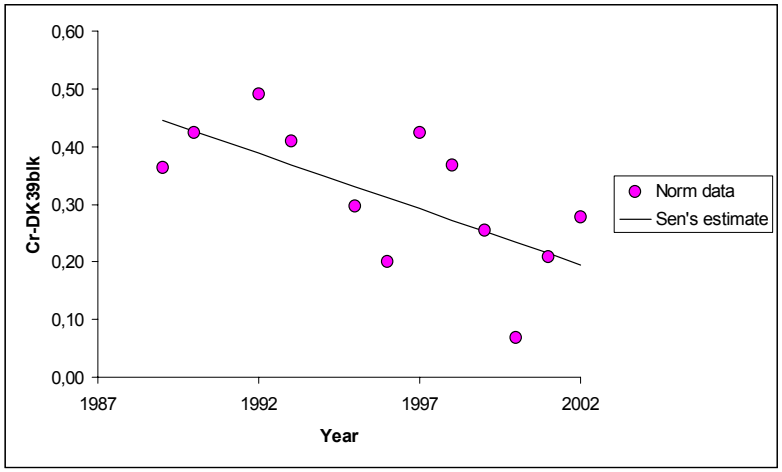


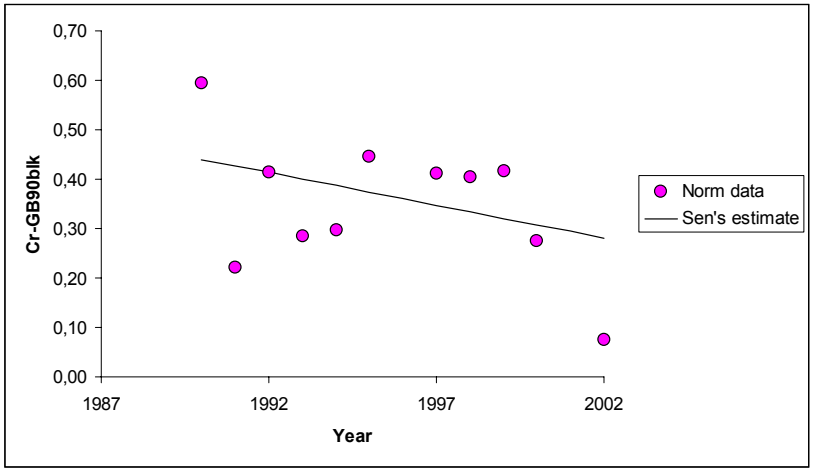


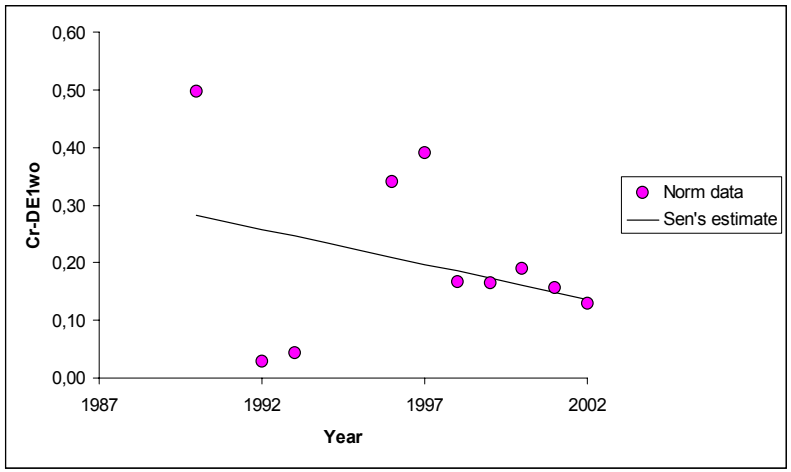


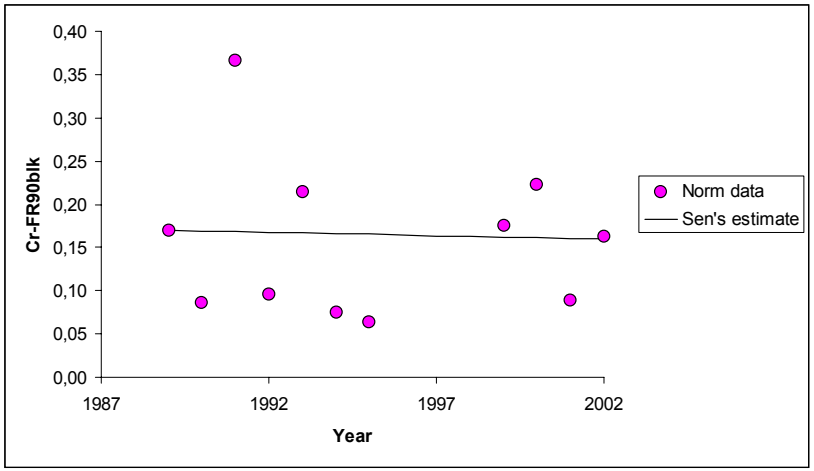


Time series of cadmium in precipitation, concentrations (g/l) normalised against precipitation amount.
DK0031 NO0001
SE0098 SE0097

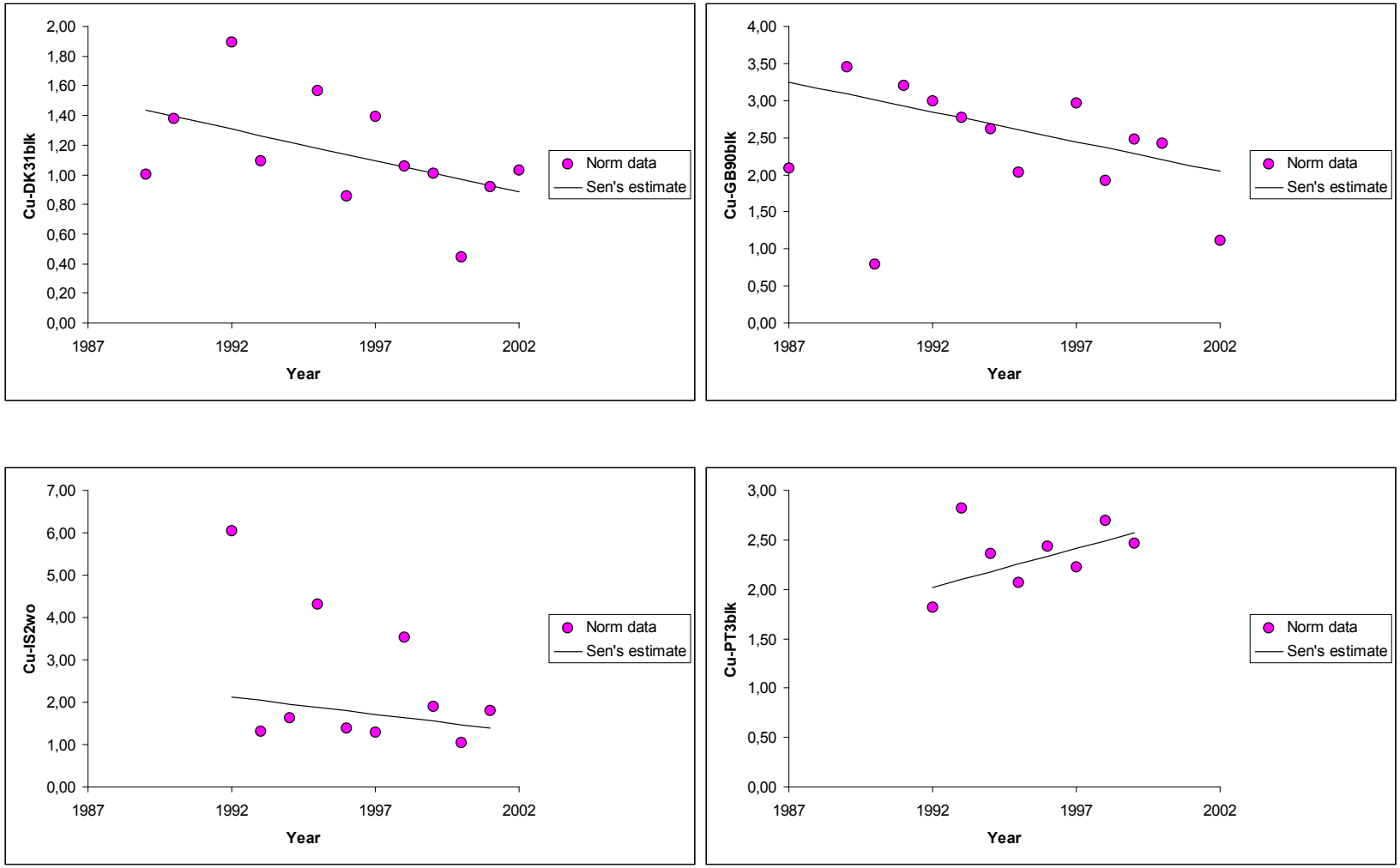




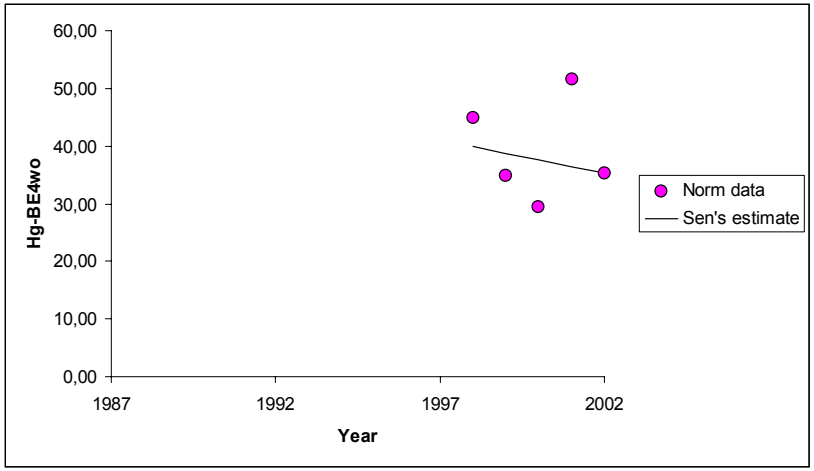


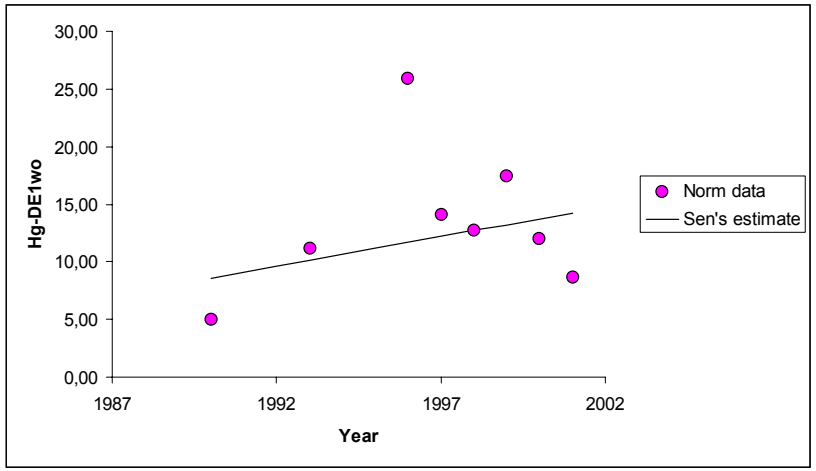


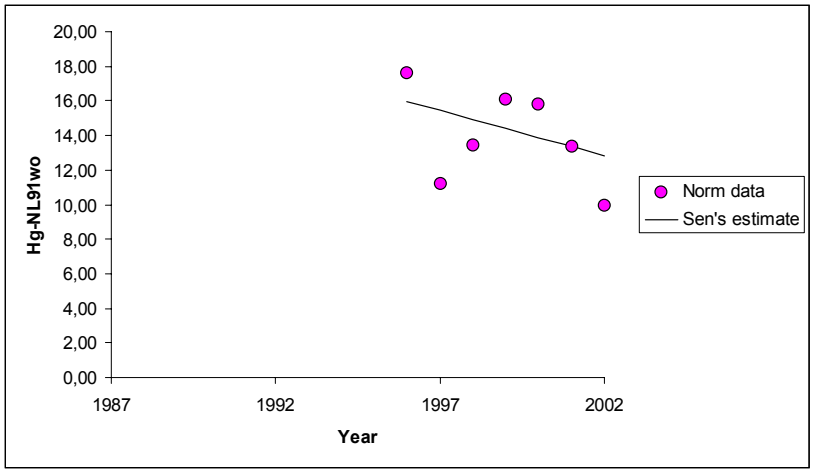
Time series of chromium in precipitation, concentrations (/l) normalised against precipitation amount.
DK0039 GB0090 DE0001 FR0090

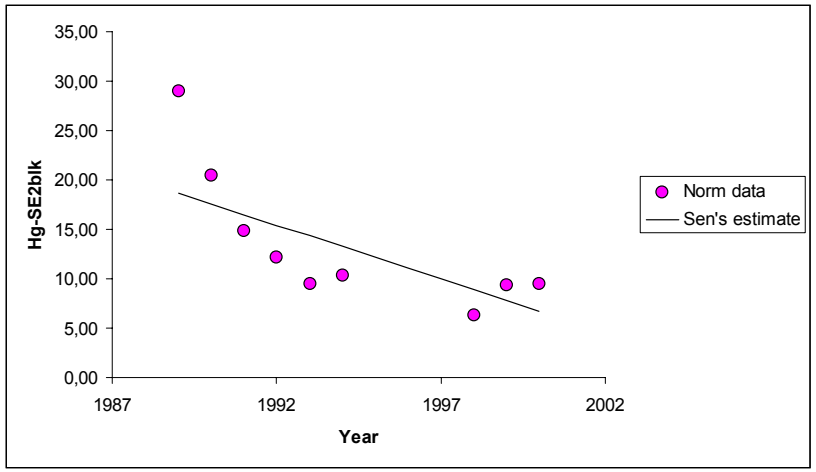


Time series of copper in precipitation, concentrations (g/l) normalised against precipitation amount.
DK0031 GB0090
IS0002 PT0003

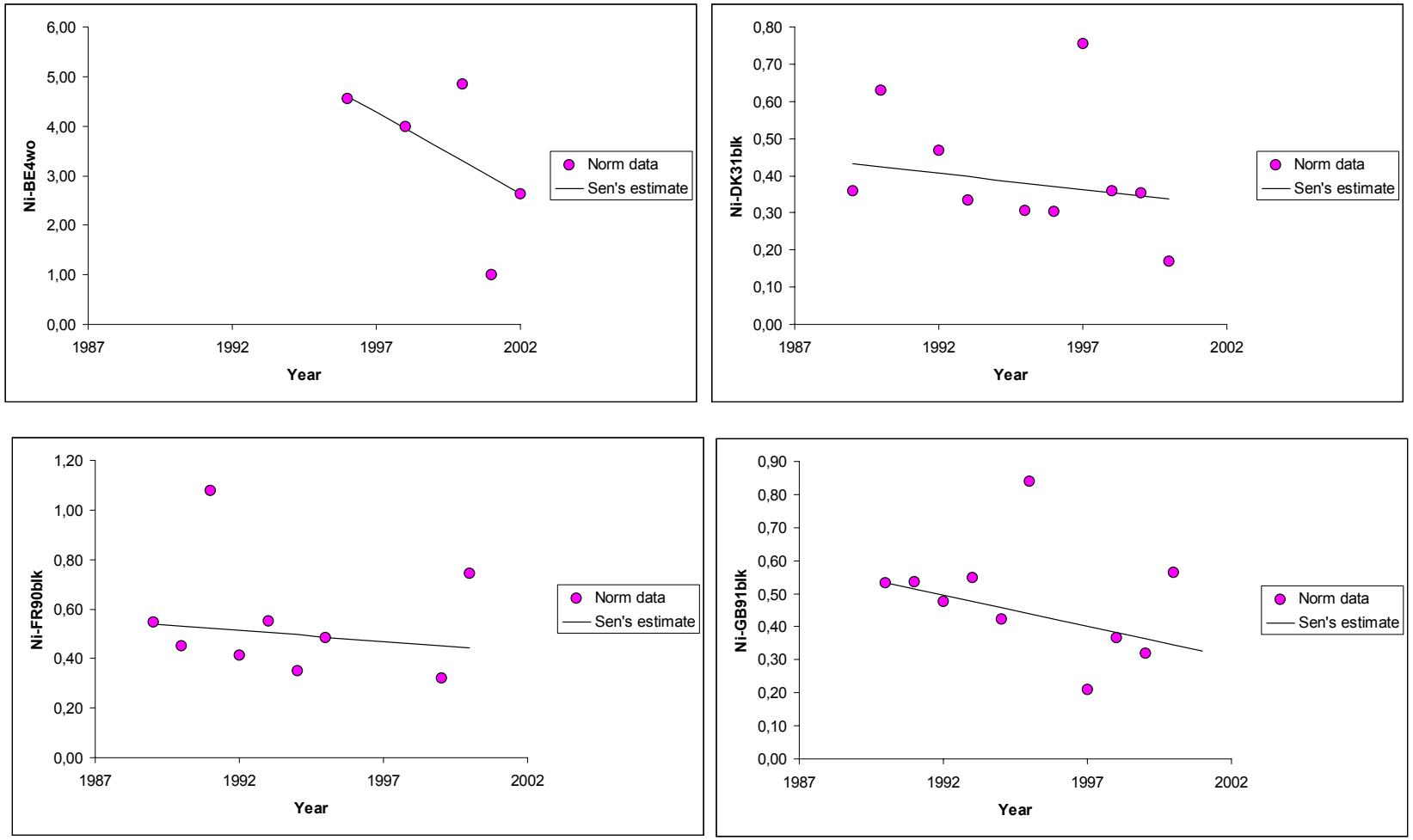




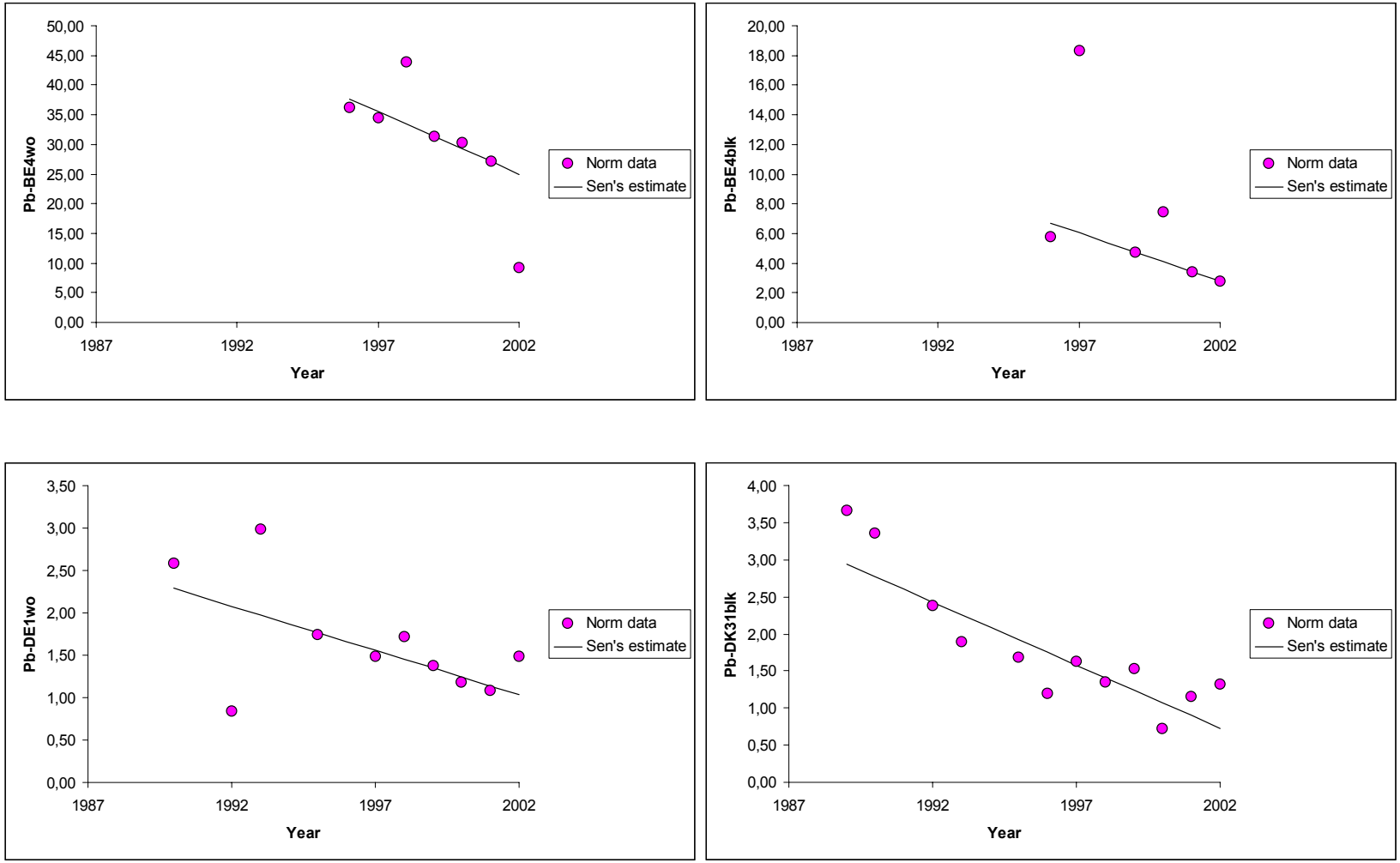




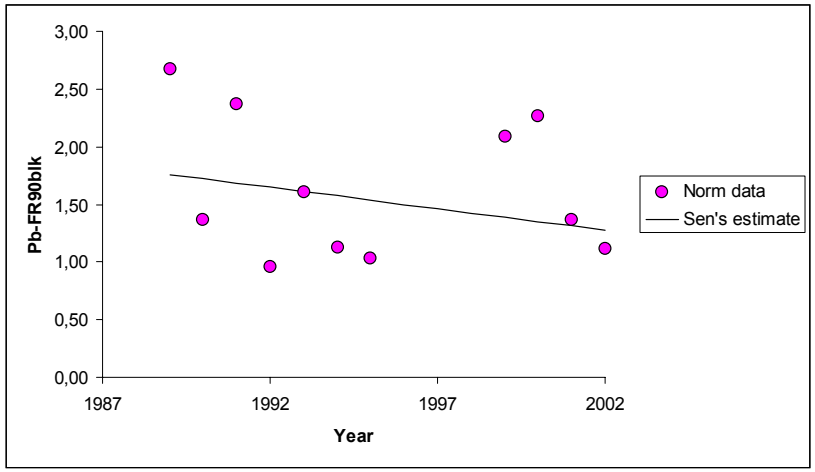
Time series of mercury in precipitation, concentrations (ng/l) normalised against precipitation amount.
BE0004 DE0001 NL0091 SE0002

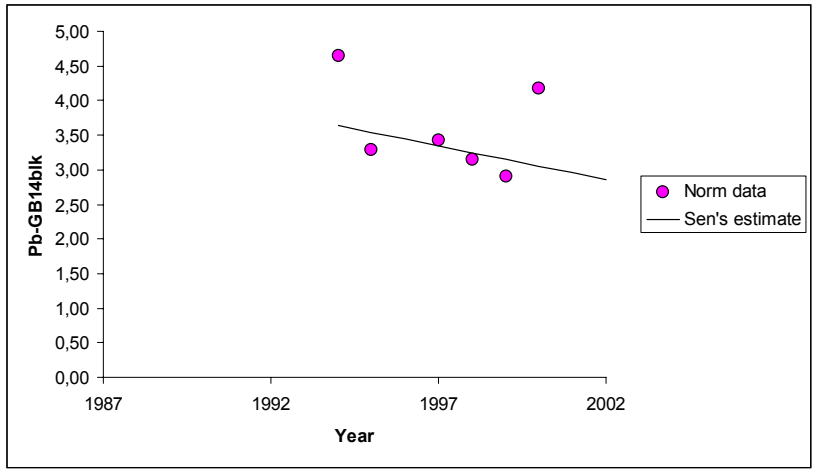


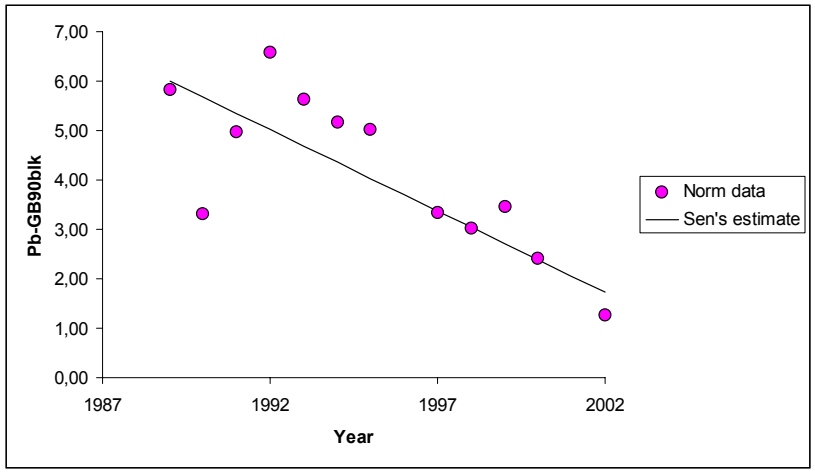
Time series of nickel in precipitation, concentrations (g/l) normalised against precipitation amount.
BE0004 DK0031
FR0090 GB0091

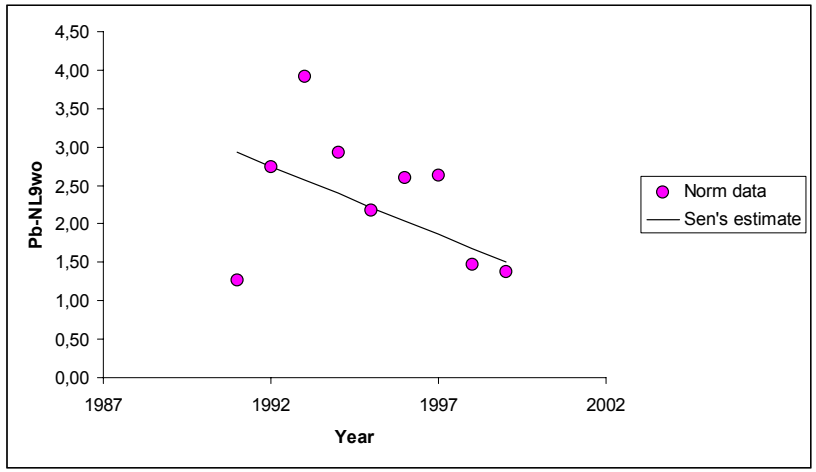


Time series of lead in precipitation, concentrations (g/l) normalised against precipitation amount.
BE0004 wet only sample, BE0004 bulk sample
DE0001, DK0031

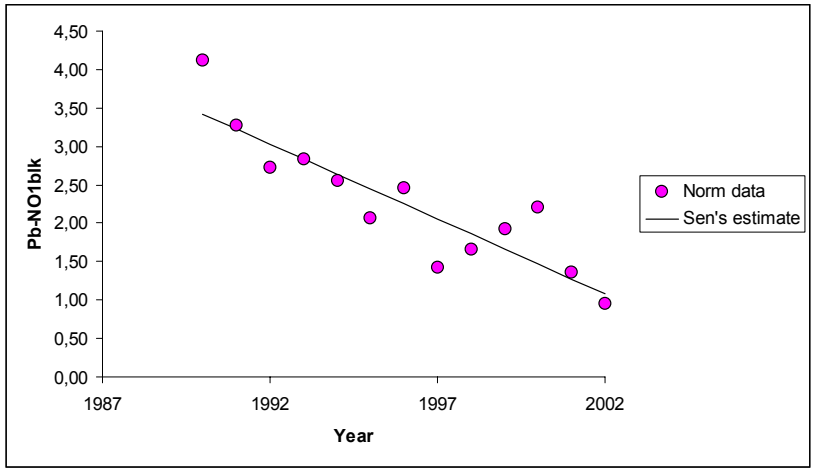


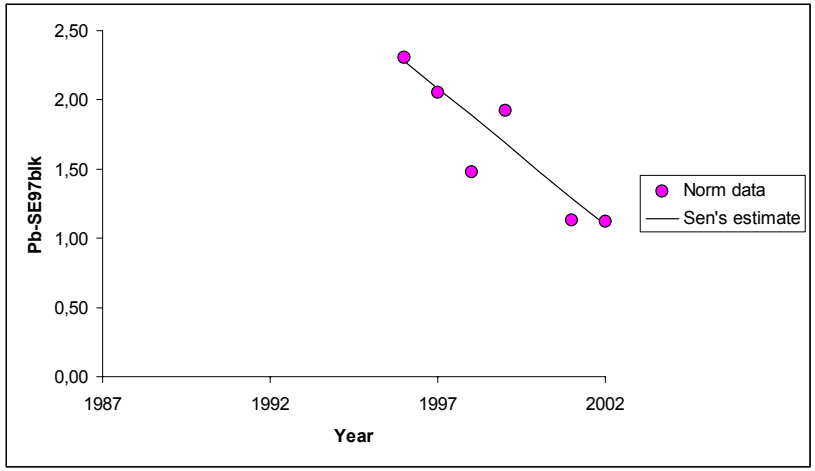


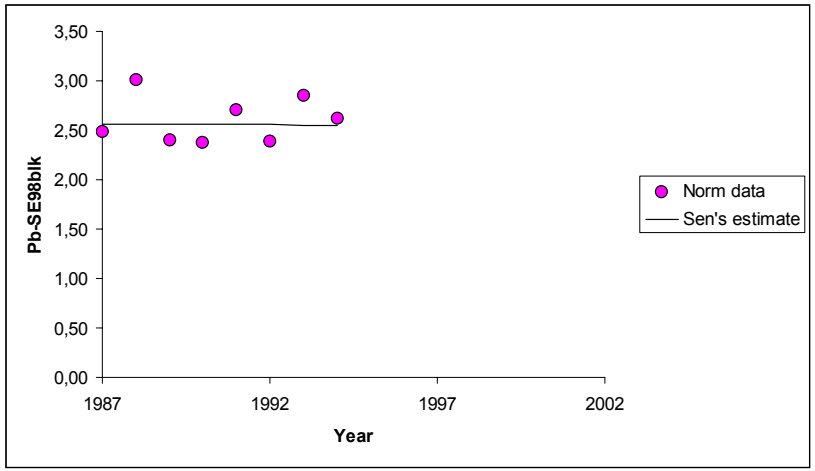




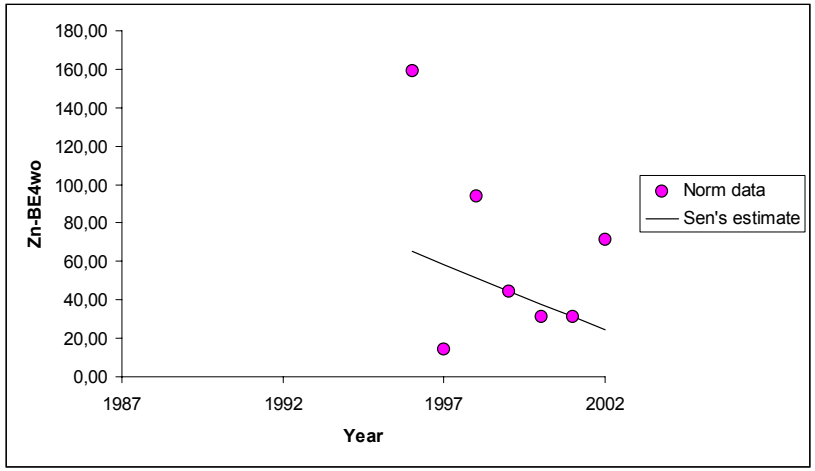
Time series of lead in precipitation, concentrations (g/l) normalised against precipitation amount.
FR0090, GB0014 *GB0090, NL0009*

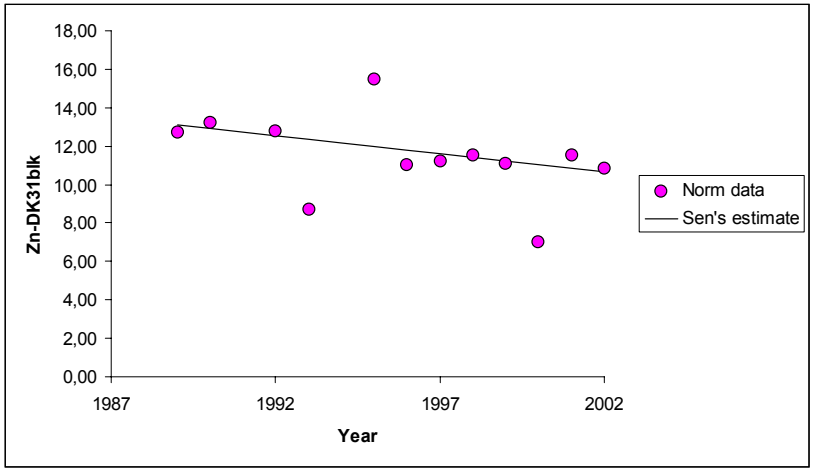


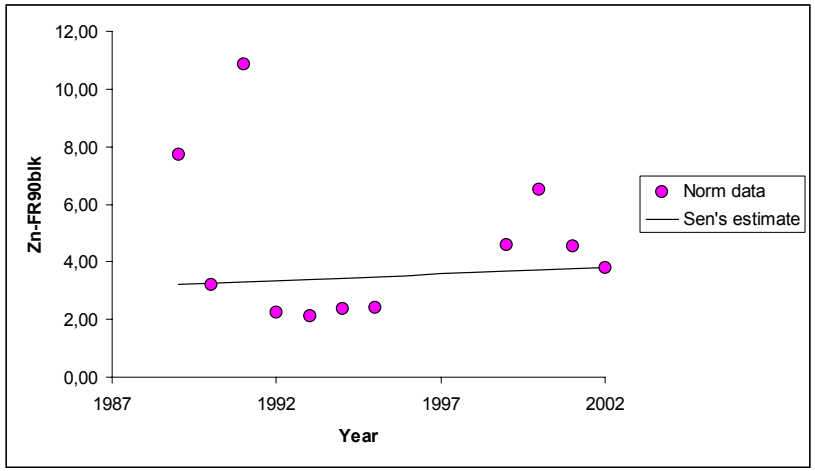


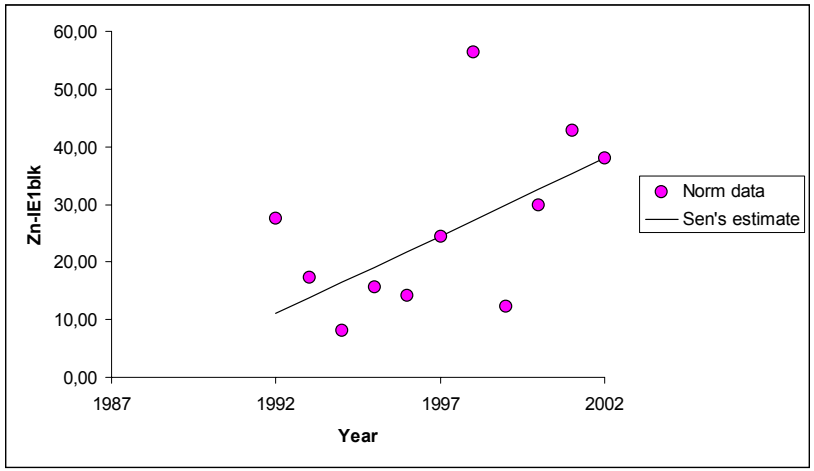


Time series of lead in precipitation, concentrations (g/l) normalised against precipitation amount.
NO0001, SE0097
SE0098



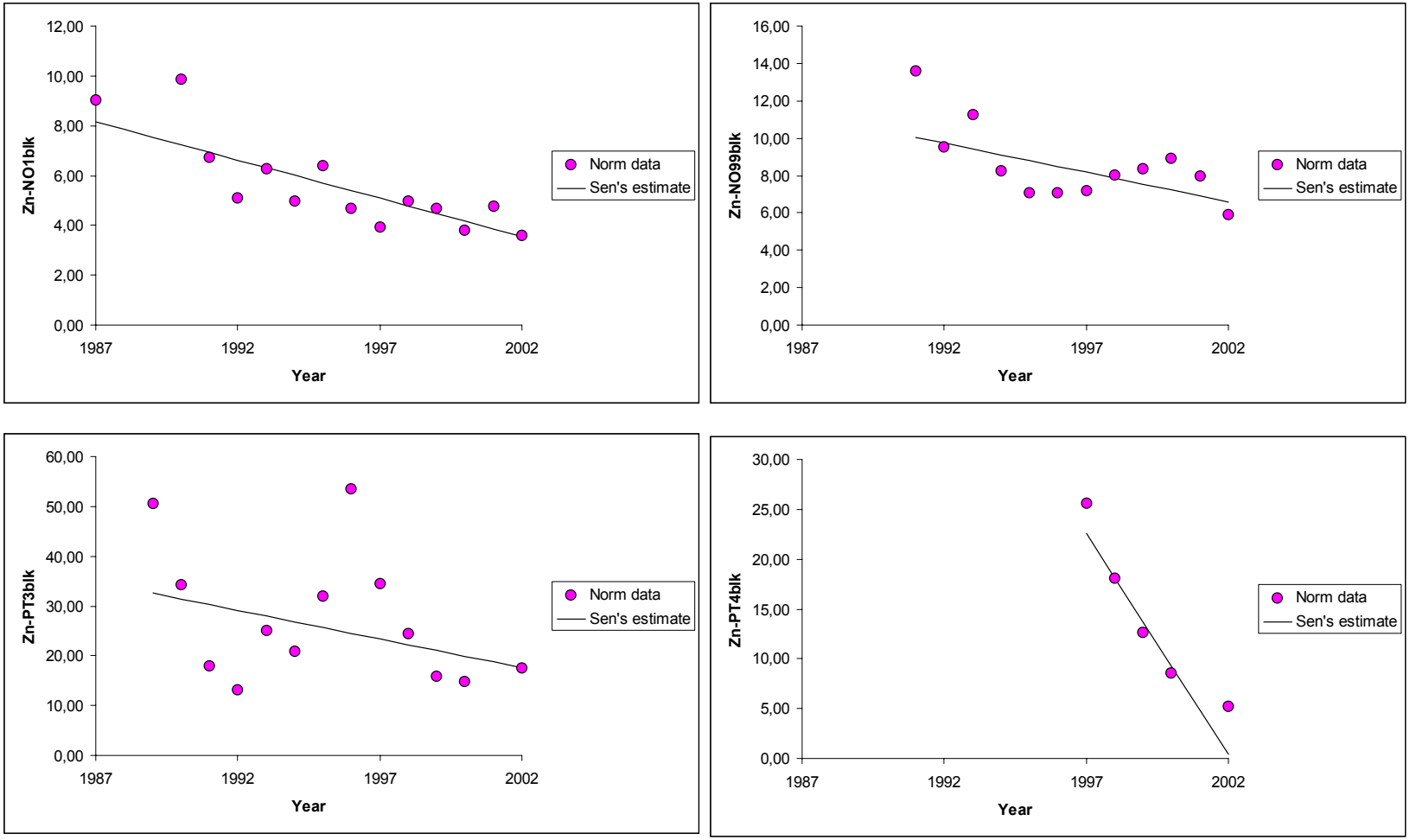






Time series of zinc in precipitation, concentrations (g/l) normalised against precipitation amount.

BE0004, DK0031
FR0090, IE0001



Time series of zinc in precipitation, concentrations (g/l) normalised against precipitation amount.
NO0001, NO0099
PT0003, PT0004

Annex 5 Nitrogen wet deposition data in CAMP

A total of 31 series of wet deposition from 11 countries were available at the CAMP homepage http://www.nilu.no/camp/. Series with a length less then three years were omitted, leaving 27 time series for examination, see Table A5.1.
After a statistical analysis and visual inspection of monthly data and requiring a time series of at least four years, the time series given in Table A5.2 have been selected for further trend analysis.

Table A5.1 Availability of nitrogen wet deposition data in CAMP

Station (CountryCode)	Frequency	Years	Gaps	Comments
Brügge (BE03)	Monthly Daily Weekly	1987; 1989-1995 1996-1997 1998-2000	1988 Okt/91-Apr/92; Nov/93; Sep/94; Aug/95-Dec/95	
Westerland (DE01)	Monthly	1996-2002		
Anholt (DK08)	Weekly	1999-2002 (NO3) 1999-2000(NH4)		
Pedersker (DK20)	Biweekly	1998-2002		
Ulborg (DK31)	Monthly	1987;1989-1991; 1995	1988; 1992-1994	
Noia (ES05)	Monthly Daily	1992 (juni-dec) 1993-May2000	Precipitation Jan-Mar 1993 Nov-Dec 1999	In 1999, missing values and zero precipitation are not distinguished.
Niembro (ES08)	Daily	1999-2002		
La Hague (FR05)	Daily	1987-2002	May/89; Jul-Dec/89; mid-Oct-Dec/90; Sep-Dec/91	Missing values and zero precipitation are not distinguished[a] The NH_4 and NO_3 values for 1st July 1993 probably represent the entire month
Porspoder (FR90)	Monthly	1993-1995; 1999-2002	1996-1998	
Loch Navar (GB06)	Daily	1987-Apr1999		Missing values and no precipitation are not always distingusihed[a]
High Muffles (GB14)	Daily Biweekly	1987-1999 2002	2000-2001	Missing and no precipitation are not always distinguished[a]
Glen Dye (GB16)	Monthly Weekly	1989-1990; 1992-1995; 2000	1991; 1996-1999	-0.03 May 2000

[a] No precipitation is coded as 0 for some years and as missing values (-9999.99) in other years.

Station (CountryCode)	Frequency	Years	Gaps	Comments
Driby(GB95)	Monthly Weekly	1989-1990; 1992-1995; 2000	1991; 1996-1999	
Tourlough Hill (IE02)	Monthly	1992-2002	Nov/00-Jul/01	
Irafoss (IS02)	Monthly Weekly Weekly	1993-1996; May1999-2002 (NO_3) May1999-2002 (NH_4)	1997-Apr1999 (NO3)	
Rejkjavik (IS90)	Monthly Weekly Weekly	1994-1996 (NO_3) 1999-2002 (NO_3) 1999-2002 (NH_4)	1997-1998 (NO_3)	
Kollumerwaard (NL09)	Daily	Jul1993-2002		Missing values and no precipitation are not always distinguished[a]
De Zilk (NL91)	Monthly	1996-2002	A number of missing month under 1996-1998	Two different values for precipitation 23/12/1999-20/01/2000
Birkenes (NO01)	Daily	1987-2002	1998 (NO3)	
Kårvatn (NO39)	Daily	1987-2002		Two series of NH_4 for 1991 - one with values until February, the other one from March to Dec
Ny Ålesund (NO57)	Weekly	1987-2002 (NH_4) 1987-2001 (NO_3)		
Lista (NO99)	Daily	1988-2002 (NH_4) 1999-2001 (NO_3)		
Viana do Costelo (PT03)	Daily	1991-2002 (NH_4) 1995-2001 (NO_3)		Missing values and no precipitation are not always distinguished[a]
Monte Velho (PT04)	Daily	1991-2002 (NH_4) 1995-2002 (NO_3)		Missing values and no precipitation are not always distinguished[a]
Angra do Heroismo (PT10)	Monthly Weekly	Jul1995-dec1995 1999-2002	A number of missing (weekly) values for NH_4 and NO_3	
Svartedalen (SE98)	Monthly	1995; 1999-2002	Sep and Dec 2000	
Rörvik (SE02)	Daily	1987-2002		July+August 1995 only weekly data

[a]Some years, no precipitation is coded as 0, other years as missing values (-9999.99).

Table A5.2 Observed precipitation amount (in mm) annual mean concentration of nitrate and ammonium in precipitation (in mg/l) at selected stations.

station	Year	precipitation	ammonium	nitrate
ES0005R	**1994**	1481.5	0.121	0.152
	1995	1747.1	0.080	0.312
	1996	2510.6	0.131	0.201
	1997	1853.7	0.077	0.119
	1998	3024.1	0.099	0.189
	1999	1219.2	0.132	0.275
FR0005R	**1987**	910.3	0.572	0.432
	1988	888	0.359	0.395
	1989	319.2		0.557
	1990	259.9		0.535
	1991	205.3		0.483
	1992	498.3	0.812	0.360
	1993	829.3	0.993	0.532
	1994	1043.1	0.609	0.367
	1995	826.97	0.655	0.456
	1996	846.7	0.445	0.451
	1997	943.86	0.429	0.346
	1998	1062.89	0.416	0.340
	1999	965.51	0.388	0.329
	2000	1162.23	0.407	0.324
	2001	1003.82	0.441	0.361
GB0014R	**1987**	728.3	0.681	0.563
	1988	718.3	0.787	0.595
	1989	471.8	0.721	0.556
	1990	667.4	0.597	0.479
	1991	539.8	0.710	0.494
	1992	767.2	0.485	0.415
	1993	672.44	0.474	0.509
	1994	581.4	0.547	0.491
	1995	530.097	0.982	0.391
	1996	458.7	1.137	0.612
	1997	680.276	0.553	0.443
	1998	817.561	0.573	0.478
	1999	862.151	0.512	0.422
GB0006R	**1987**	1010.1	0.203	0.117
	1988	1144.5	0.195	0.119
	1989	1000.8	0.171	0.094
	1990	1471.5	0.117	0.088
	1991	1019.4	0.168	0.117
	1992	1464.4	0.174	0.097
	1993	1195.67	0.167	0.150
	1994	1436	0.180	0.166
	1995	1145.866	0.284	0.203
	1996	1107.289	0.223	0.147
	1997	1193.055	0.253	0.182
	1998	1515.52	0.133	0.086

station	Year	precipitation	ammonium	nitrate
NL0009R	**1994**	583.38	1.110	0.569
	1995	589.28	1.086	0.583
	1996			
	1997	458.9	1.189	0.614
	1998	814.62	0.868	0.499
	1999	760.74	0.813	0.453
	2000	698.78	0.940	0.558
	2001	849.68	0.744	0.426
NO0001R	**1987**	1576.2	0.463	0.439
	1988	1986.1	0.612	0.588
	1989	1228.5	0.641	0.780
	1990	1861	0.465	0.480
	1991	1246.5	0.515	0.606
	1992	1343.7	0.478	0.596
	1993	1244.9	0.545	0.617
	1994	1396	0.537	0.609
	1995	1410.8	0.452	0.549
	1996	1192.4	0.528	0.633
	1997	1243.3	0.480	0.553
	1998	1595.9	0.455	
	1999	1844.4	0.386	0.484
	2000	2415.2	0.360	0.481
	2001	1604.7	0.427	0.489
NO0039R	**1987**	1463.8	0.132	0.099
	1988	1549.6	0.102	0.067
	1989	1538.7	0.075	0.075
	1990	1520.1	0.081	0.056
	1991	1618.8	0.114	0.072
	1992	1619.9	0.070	0.079
	1993	1422.8	0.131	0.072
	1994	1474.2	0.091	0.076
	1995	1661.4	0.076	0.059
	1996	1170.9	0.115	0.085
	1997	1841.8	0.120	0.065
	1998	1451.3	0.119	0.064
	1999	1305.3	0.084	0.076
	2000	1243	0.094	0.059
	2001	1522.8	0.080	0.052
PT0004R	**1991**	428.7	0.204	
	1992	294.4	0.305	
	1993	552.6	0.225	
	1993	426.4	0.253	
	1995	536.2	0.126	0.198
	1996	801.8	0.099	0.130
	1997	793.9	0.074	0.129
	1998	206.4	0.234	0.285
	1999	560	0.247	0.164
	2000	574.4	0.173	0.173
	2001	501.1	0.214	0.193

station	Year	precipitation	ammonium	nitrate
PTooo3R	**1991**	1146	0.173	
	1992	903.6	0.190	
	1993	1150.8	0.216	
	1994	1494.5	0.174	
	1995	1455.4	0.219	0.143
	1996	1780.3	0.227	0.168
	1997	1509.8	0.139	0.152
	1998	887.5	0.190	0.188
	1999	1495.8	0.139	0.124
	2000	1890.6	0.206	0.153
	2001	1849.9	0.144	0.142
SE0002R	**1987**	734	0.636	0.593
	1988	834.2	0.730	0.822
	1989	591.3	0.753	0.829
	1990	853.7	0.659	0.662
	1991	647.7	0.634	0.713
	1992	779.8	0.559	0.614
	1993	652.01	0.579	0.605
	1994	755.1	0.427	0.517
	1995			
	1996	463.59	0.636	0.647
	1997	584.1	0.600	0.639
	1998	793.1	0.396	0.475
	1999	867.3	0.493	0.508
	2000	886.1	0.478	0.586
	2001	610.2	0.525	0.540

Annex 6 Observed concentrations of γ HCH in precipitation and air

Observed annual mean concentrations of γ-HCH in precipitation (in ng/l) and in air (in ng/m^3). The precipitation amount is given in mm. See Annex 4 for colour coding. Time series plots of concentrations in precipitation and air are presented in Figure A6.1 and A6.2.

Code	sampler	Year	precip	ng/l	pg/m3
BE0004R	BULK	1996	604	33.38	
		1997	542	104.94	
		1998	984	28.13	
		1999	852	26.78	
		2000	810	18.59	
		2001	750	22.88	
		2002	830	11.49	
DE0001R	WET ONLY	1996	325	10.74	
		1997	402	13.51	
		1998	740	6.04	
		1999			
		2000	641	6.43	
		2001	678	3.24	
DK0031R	BULK	1990	1063	17.17	
		1991	705	11.91	
		1992	905	15.82	
IE0002R	BULK	1992	1127		
		1993	1443		
		1994	1413		
		1995	1284		
		1996	1970		
		1997	1683		
		1998	1922		
		1999	1744		
		2000	1907		
		2001	1434		
		2002	2277	0.77	
IS0091R	BULK	1995	1170	0.62	15.48
		1996	1436	0.27	9.64
		1997	1389	0.42	6.93
		1998	1419	0.25	5.69
		1999	708	0.15	5.12
		2000	622	0.10	3.83
		2001	718	0.21	6.43
		2002	806	0.12	3.51
NL0091R	BULK	1996	598	28.12	
		1997	590	48.19	
		1998	828	48.69	
		1999	994	17.33	

Code	sampler	Year	precip	ng/l	pg/m3
		2000	788	65.90	
		2001	955	14.09	
		2002	885	20.00	
NO0042R		1993			14.41
		1994			16.06
		1995			13.29
		1996			12.85
		1997			14.77
		1998			9.43
		1999			10.74
		2000			5.90
		2001			5.62
		2002			7.62
NO00099R	BULK	1991	1031	4.05	84.58
		1992	1376	5.02	84.17
		1993	846	8.45	57.50
		1994	1180	10.16	120.83
		1995	895	5.24	66.38
		1996	911	8.01	60.72
		1997	1220	4.89	61.99
		1998	1239	4.75	55.88
		1999	793	4.01	33.75
		2000	1572	3.09	24.48
		2001	1172	2.47	15.55
		2002	1129	1.68	12.41
SE0002R	BULK	1995		3.31	51.20
		1996		8.87	25.08
		1997		4.59	25.33
		1998		3.53	28.17
		1999		1.78	17.73
		2000		3.41	23.92
SE0012R (a)		1995			49.83
		1996			74.00
		1997			45.25
		1998			59.27
		1999			
		2000			74.08
		2001			
		2002			12.42
SE0014R		2002		0.07	11.60

(a) The station SE0012R, Aspvreten (located on the Baltic coastline) is included in the CAMP database although it is not part of the CAMP network.

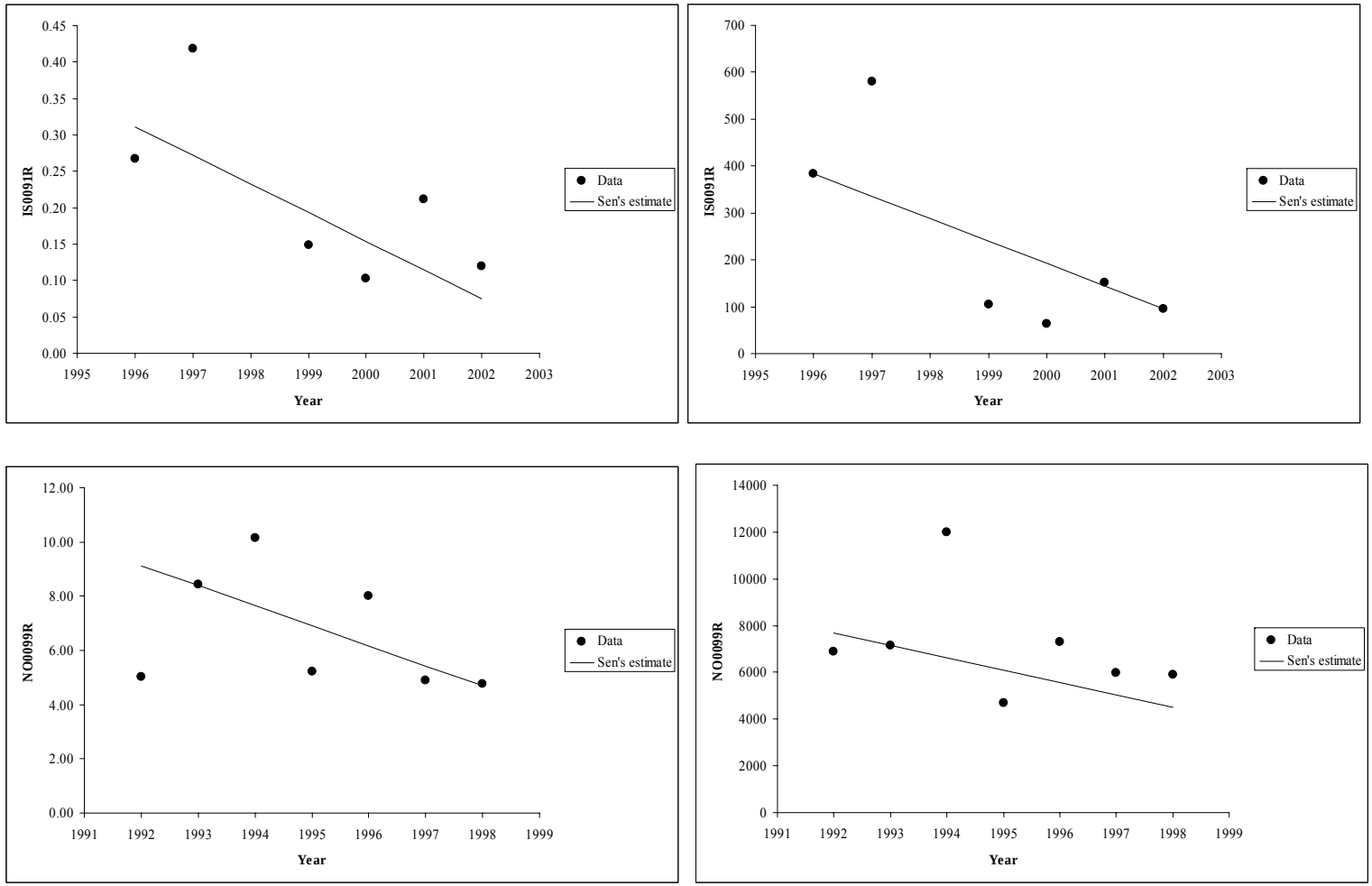


Figure A6.1. γ-HCH (lindane) concentrations in precipitation (left panels, in ng/l) and annual deposition (right panels, in ng/m2 per year) at the stations IS0091R, and NO0099R. The trends at NO0099R are significant.

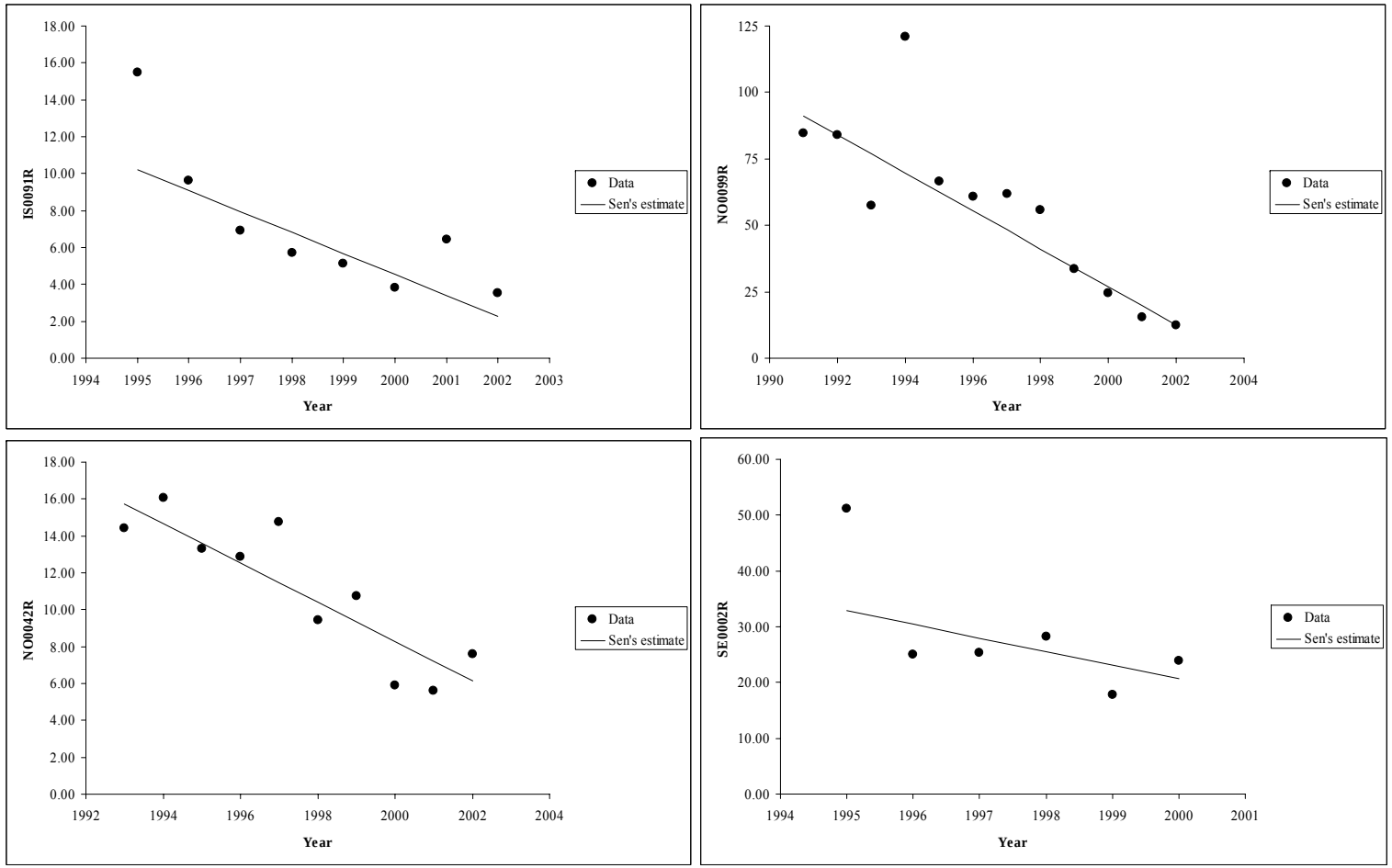


Figure A6.2. γ-HCH (lindane) concentrations in air (in pg/m3) at the stations IS0091R, NO0042R, NO0099R and SE0002R. Except at SE0002R a significant downward trend is observed.

Annex 7 Ambient concentrations of nitrogen components in CAMP

A total of 17 series of air concentrations of nitrogen components from 9 countries were available at the CAMP homepage http://www.nilu.no/camp/. Series with a length less than three years were not considered. see Table A7.1.
After a statistical analysis and visual inspection of monthly data and requiring a time series of at least four years, the time series given in Table A7.2 have been selected for further trend analysis.
In Table A7.1 and A7.2 information on the following nitrogen components is given:

- NO2 nitrogen dioxide (gaseous)
- NO nitrogen monoxide (gaseous; voluntary component)
- HNO3 nitric acid (gaseous)
- NO3 nitrate (aerosol)
- NH4 ammonium (aerosol)
- sNH4 sum of ammonia (NH3, gaseous) and ammonium
- sNO3 sum of nitric acid and nitrate.

Table A7.1 Availability of data on air concentrations of nitrogen components in CAMP

Station (CountryCode)	Frequency	Years	Gaps	Comments
Moerkerke (BE11)	Monthly	1990-2002 (NO_2)		
		1990-1998; 2000 (NO)	1999 (NO)	
Houtem (BE13)	Monthly	1990-2002 (NO_2)	Feb-Mar 1991 (NO_2, NO)	
		1990-1998; 2000 (NO)	1999 (NO)	
Westerland (De01)	Monthly	1996 -1999 (NH_4, NO_3)		
		1996-1998; 2000-2002 (NO_2)	1999 (NO_2)	
Anholt (DK09)	Daily	1991-2000 (sNH_4)		
		1991-2002 (sNO_3)		
		1997-2000 (NH_3)		
Ulborg (DK31)	Monthly	1990-1995 (NH_4)		
	Daily	1999-2002 (sNH_4, sNO3)		
	Monthly	1990-1991; 1995 (NH_3)	1992-1994 (NH_3)	
	Daily	1996 (NH_3)		
	Monthly	1990-1992; 1995 (NO_2)	1993-1994 (NO_2)	
Noia (ES05)	Monthly	1992		
	Daily	1993-2000 (NH_4, sNH_4, sNO_3) 1993-1997 (NO_2)		
Niembro (ES08)	Daily	1999-2000 (NH_4)		
		1999-2002 (sNH_4, sNO_3)		
High Muffles	Daily	1989-1994; 1997-	1995-1996 (sNH4)	High concentration

Station (CountryCode)	Frequency	Years	Gaps	Comments
		Apr1999 (sNH_4) 1989-1995;1997-Apr1999 (sNO_3)	 1996 (sNO3)	values for both variables just before the end of measurement period (April 1999)
Kolummerwaard (NL09)	Hourly	1990-2002 (NO_2) 1996-2002 (NO)	Aug 1994 (NO_2)	Two series for NO (different analytical procedures?)
De Zilk (NL91)	Daily	1996-2002 (NH_4, NO_3)		
Birkenes (NO01)	Daily	1987-2002 (NH_4, NH_3, sNH_4, sNO_3) 1993-2002 (NO_3) 1993-2001 (HNO_3) 1987-2001 (NO_2)	Oct-Dec2001 (NH4, sNH_4) 1994; Oct-Dec 2001 (NO_3) 1994	
Kårvatn (NO39)	Daily	1993-2002 (NH_4, NH_3, NO_3, HNO_3) Jul1987-2002 (sNH_4, sNO_3, NO_2)	Oct-Dec2001 (NH_4, NO_3) Oct-Dec2001 (sNH_4)	
Spitzbergen (NO42)	Daily	1993-2002 (NH_4, NH_3, HNO_3, NO_3) Nov1989-2002 (sNH_4, sNO_3) 1991-1994 (NO_2)	Oct-Dec 2001 (NH_4, NO_3) Oct-Dec 2001 (sNH_4, sNO_3)	High concentration values for Sep 2001 (sNH_4, sNO_3) (just before the gap in measurements)
Ny Ålesund (NO57)	Monthly	1993-1994 (NO_2,) 1993-1997 (sNH_4, sNO3)		
Lista (NO99)	Monthly	1993-1997 (NO_2, sNH_4, sNO_3)		
Rörvik (SE02)	Daily	1991-2002 (NO_2) 1987-2002 (sNH_4) 1992-2002 (sNO_3)		

Table A7.2 Observed ambient annual mean concentrations of nitrogen components (in µg/m^3).

station		sNH_4	sNO_3	NO_2
DK0008	**1991**	2.071	1.079	
	1992	1.703	0.944	
	1993	1.787	0.995	
	1994	1.556	0.879	
	1995	1.622	0.958	
	1996	1.654	0.922	
	1997	1.409	0.792	
	1998	1.141	0.678	
	1999	1.390	0.858	
	2000	1.281	0.870	
	2001		0.739	
ES0005R	**1993**	0.981	0.173	6.550
	1994	0.927	0.172	5.278
	1995	0.997	0.234	4.028
	1996	0.435	0.329	2.383
	1997	0.486	0.098	4.225
	1998	0.723	0.217	
GB0014R	**1989**	1.445	0.644	
	1990	2.054	0.762	
	1991		1.043	
	1992		0.872	
	1993	1.579	0.855	
	1994	1.702	0.639	
	1995		0.657	
	1996			
	1997	1.854	0.799	
	1998	1.563	0.674	
NL0009R	**1992**			6.358
	1993			5.963
	1994			6.096
	1995			5.220
	1996			
	1997			6.166
	1998			5.330
	1999			5.169
	2000			5.196
	2001			4.461
NO0001R	**1987**	0.654	0.299	1.119
	1988	0.632	0.284	1.274
	1989	0.621	0.257	1.109
	1990	0.761	0.270	1.005
	1991	0.763	0.264	0.928
	1992	0.538	0.243	0.689
	1993	0.543	0.231	0.595
	1994	0.611	0.274	0.648
	1995	0.532	0.293	0.677

station		sNH_4	sNO_3	NO_2
	1996	0.575	0.290	0.679
	1997	0.534	0.234	0.692
	1998	0.407	0.188	0.616
	1999	0.511	0.201	0.524
	2000	0.433	0.204	0.570
	2001			0.464
NO0039R	**1988**	0.435	0.070	0.559
	1989	0.421	0.080	0.331
	1990	0.350	0.062	0.413
	1991	0.356	0.063	0.262
	1992	0.383	0.062	0.190
	1993	0.381	0.066	0.160
	1994	0.477	0.102	0.225
	1995	0.372	0.096	0.262
	1996	0.471	0.085	0.241
	1997	0.500	0.073	0.253
	1998	0.370	0.047	0.264
	1999	0.438	0.048	0.231
	2000	0.556	0.053	0.319
	2001			0.187
NO0042R	**1990**	0.089	0.043	
	1991	0.094	0.045	
	1992	0.079	0.036	
	1993	0.093	0.056	
	1993	0.092	0.057	
	1995	0.099	0.082	
	1996	0.108	0.081	
	1997	0.131	0.069	
	1998	0.122	0.041	
	1999	0.192	0.032	
	2000	0.107	0.028	
SE0002R	**1989**	1.209		
	1990	1.052		
	1991	1.037		2.471
	1992	1.016	0.597	2.573
	1993	1.379	0.636	2.187
	1994	1.116	0.629	1.752
	1995	1.217	0.646	2.166
	1996	1.256	0.647	2.160
	1997	0.938	0.527	2.013
	1998	0.961	0.518	1.667
	1999	0.995	0.711	1.642
	2000	0.890	0.645	1.511
	2001	0.988	0.757	1.478

Annex 8 Observed concentrations of heavy metals in air

Annual mean concentrations of heavy metals on aerosol (ng/m^3). In this table the following colour code is used: White: no data reported
Yellow (light grey): valid data
Red (dark grey): invalid data (less than 75% data coverage).

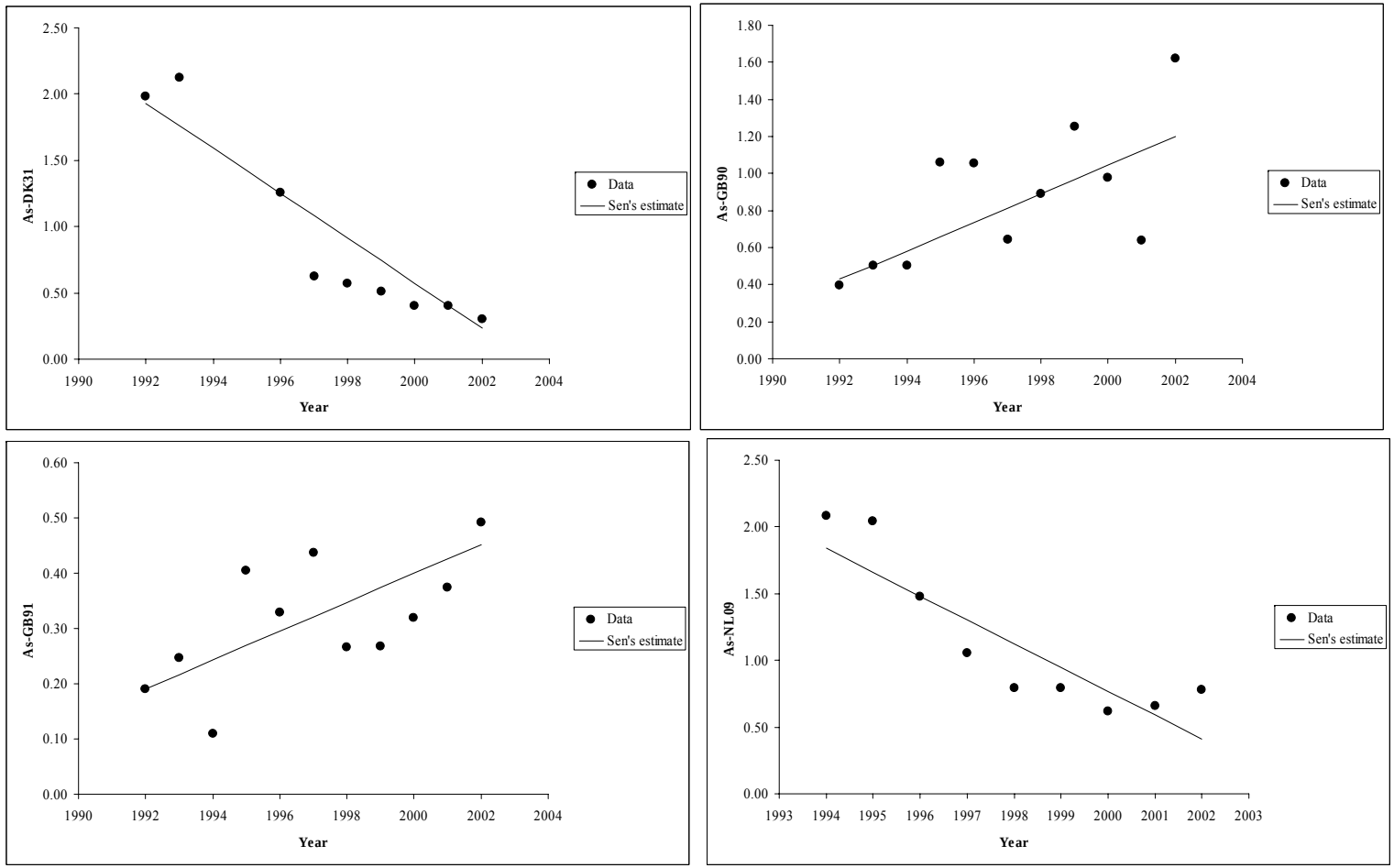
A trend analysis has been made for all time series of at least five years. The results for station/pollutant combinations showing a significant (at an α = 0.1 level or better) change over time are shown as time series plots, see section 6.2 for further discussion.

Station code	year	As	Cd	Cr	Cu	Pb	Ni	Zn	Hg
BE0002R	1994					52.56			
	1995								
	1996		10.00		10.00	74.20	10.00	121.70	
BE0004R	1997		-		-	-	-	120.00	
	1998		-		-	-	-	132.50	
	1999		-		24.60	26.50	13.50	36.80	
	2000				-	42.00	22.80	74.50	
	2001		-		36.10	25.00	12.90	42.00	
	2002				-	17.00	17.90	42.90	
BE0090R	1990			-	43.30	104.20	28.60	350.80	
	1991				31.70	115.00	23.30	221.70	
	1992								
	1993					66.40			
DE0001R	1987		0.37		2.55	23.33			
	1988		0.39		3.35	23.25			
	1989		0.41		2.64	19.55			
	1990		0.30		3.13	17.33			
	1991		0.36		1.53	18.03			
	1992		0.25		2.16	11.88			
	1993		0.20		2.26	12.72			
	1994		0.16		1.89	9.92			
	1995		0.18		1.92	8.79	1.42		
	1996		0.25		1.61	9.63	1.35		
	1997		0.20		1.53	8.04	1.24		
	1998	0.63	0.15		1.31	6.68	1.06		
	1999	0.55	0.18		1.84	5.25	1.06		
	2000					6.47			
	2001		0.16						
DK0008R	1996				1.53	8.28	1.91	12.60	
	1997				1.27	5.58	1.86	10.44	
	1998				1.58	5.04	1.54	10.72	

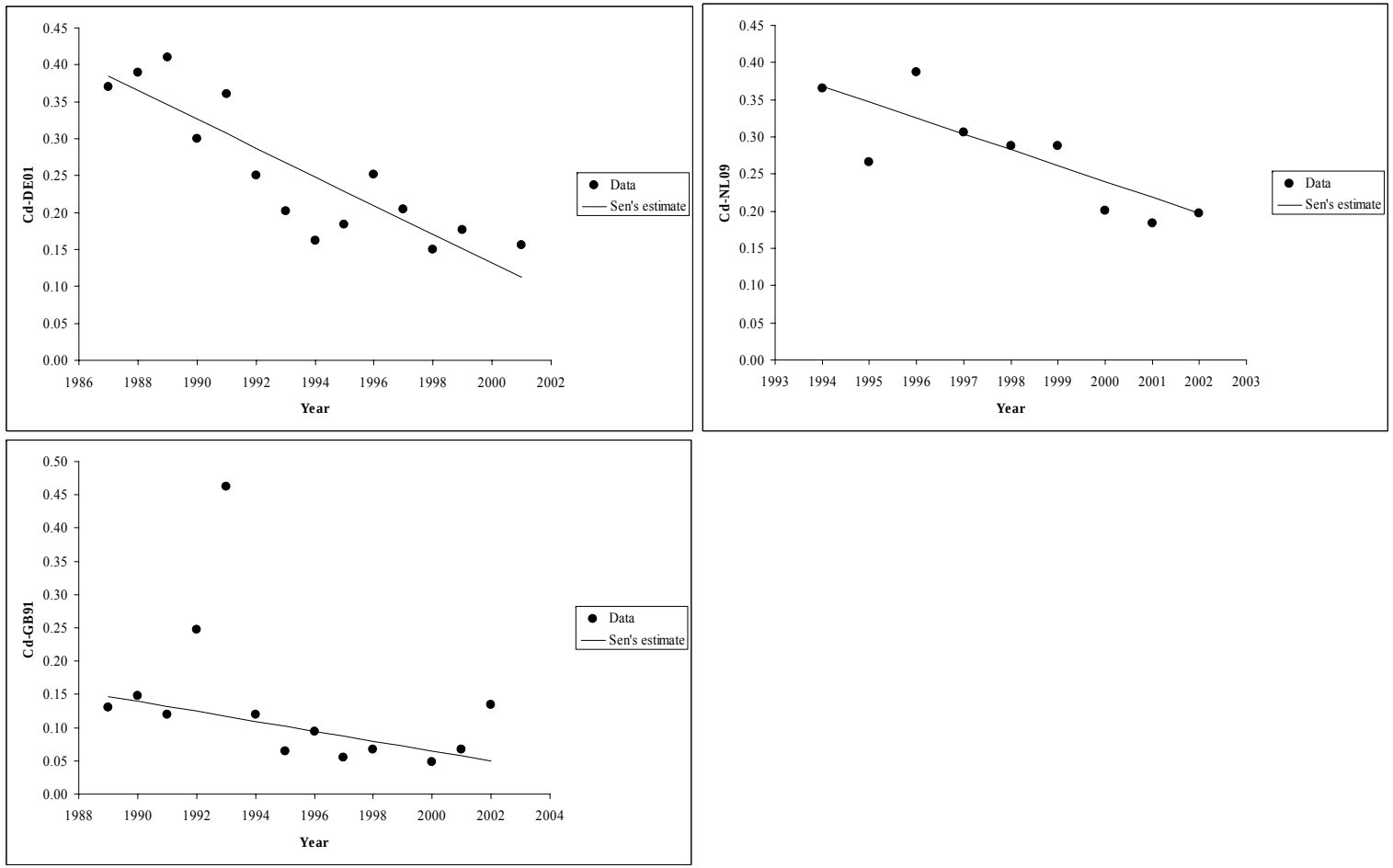
Station code	year	As	Cd	Cr	Cu	Pb	Ni	Zn	Hg
	1999	0.45	0.62	1.23	1.49	5.29	1.59	10.67	
	2000	0.40	0.59	0.92	1.49	5.06	1.54	10.67	
	2001	0.46	0.65	0.85	1.10	4.56	1.68	10.23	
	2002	0.36	0.62	1.01	1.06	3.66	1.63	9.84	
DK0031R	1990			1.48	1.62	17.40	1.68	23.96	
	1991			2.63	2.18	17.33	1.98	23.25	
	1992	1.99		1.45	1.91	13.42	1.81	22.08	
	1993	2.12		1.45	1.56	11.89	1.61	19.80	
	1994			1.90	1.65		1.54	18.08	
	1995	2.03			1.90	9.33	1.72	17.61	
	1996	1.26	1.83	1.24	1.42	7.47	1.49	15.00	
	1997	0.62	0.74	1.21	1.28	6.14	1.37	12.37	
	1998	0.57	0.67	1.28	1.52	6.75	1.24	13.17	
	1999	0.51	0.58	1.36	1.39	5.28	1.17	12.02	
	2000	0.40	0.62	1.05	1.41	4.70	1.02	10.58	
	2001	0.40	0.76	1.01	1.08	4.40	1.20	9.74	
	2002	0.30	0.61	0.92	0.92	2.99	0.94	8.82	
ES0008R	2001		0.15		37.84	8.45			
	2002		0.18		27.90	12.93			
GB0014R	1993	0.51	0.51	1.15	5.06	13.67	2.38	28.84	
	1994	0.44	0.15	0.69	2.94	11.03	1.86	20.06	
	1995	0.77	0.16	1.18	2.72	12.16	1.81	20.39	
	1996	0.75	0.15	1.02	2.99	7.68	1.56	25.63	
	1997	0.40	0.13	0.87	6.33	8.15	2.18	96.00	
	1998	0.40	0.10	0.55	4.00	5.43	1.10	34.10	
	1999	0.63	0.19	0.41	4.86	9.09	1.15	39.90	
	2000	0.54	0.23	1.30	3.27	6.31	2.47	57.57	
	2001	0.34	0.21	1.11	2.04	5.50	1.16	34.98	
	2002	0.67	0.27	1.71	1.45	5.33	1.17	48.75	
GB0090R	1989		0.40	0.80	3.72	28.30	2.21	26.10	
	1990		0.36	0.48	2.77	21.00	1.75	22.75	
	1991		0.66	0.72	3.36	26.14	3.27	73.94	
	1992	0.40	0.67	0.69	3.86	21.94	3.75	101.11	
	1993	0.51	0.30	1.91	4.16	11.36	4.17	50.67	
	1994	0.50	0.41	0.51	4.32	13.89	2.05	32.47	
	1995	1.06	0.23	0.79	4.18	15.01	2.00	38.08	
	1996	1.05	0.22	0.85	3.80	13.29	1.68	18.76	
	1997	0.64	0.17	0.51	2.43	11.70	1.58	11.00	
	1998	0.89	0.19	0.43	2.48	11.42	1.35	28.07	
	1999	1.25	0.31	1.16	3.48	16.92		18.13	
	2000	0.98	0.28	1.09	2.46	12.80	0.98	30.21	
	2001	0.64	0.22	0.92	2.24	8.81	2.19	38.38	
	2002	1.62	0.45	1.36	2.27	8.02	1.92	42.36	

Station code	year	As	Cd	Cr	Cu	Pb	Ni	Zn	Hg
GB0091R	1989		0.13	0.33	1.29	7.64	0.74	7.50	
	1990		0.15	1.99	1.97	6.77	2.85	9.40	
	1991		0.12	0.28	1.58	7.03	0.80	10.55	
	1992	0.19	0.25	0.28	1.32	5.68	0.82	22.83	
	1993	0.25	0.46	1.23	2.24	4.60	2.77	15.84	
	1994	0.11	0.12	0.41	2.06	5.19	1.41	20.84	
	1995	0.41	0.06	0.33	1.90	4.36	0.71	16.23	
	1996	0.33	0.09	0.54	2.64	4.35	0.61	11.63	
	1997	0.44	0.06	0.40	2.73	3.45	0.95	26.95	
	1998	0.27	0.07	0.10	1.93	2.17	0.43	11.63	
	1999	0.27	0.09	0.83	1.20	2.24	0.28	15.18	
	2000	0.32	0.05	0.67		2.13	0.58	20.80	
	2001	0.38	0.07	0.48	0.76	2.35	0.47	13.43	
	2002	0.49	0.13	0.68	0.71	1.92	0.50	27.23	
GB0092R	1993	0.97	0.38	0.60	4.28	21.72	2.02	21.33	
	1994	0.60	0.24	0.60	3.20	14.47	2.55	25.08	
	1995	0.75	0.18	0.57	2.78	12.93	2.20	32.25	
GB0093R	1989		0.59	1.22	4.10	22.72	2.43	56.00	
	1990								
	1991		1.90	0.79	3.06	17.04	2.36	50.67	
	1992		0.99	1.05	4.32	22.26	2.90	48.92	
IS0091R	1995	0.27	0.03	3.67	1.43	0.96	3.41	6.29	
	1996	0.17	0.04	3.68	1.13	0.91	8.55	7.20	
	1997	0.25	0.04	6.15	1.70	0.96	14.09	8.66	
	1998	0.05	0.10	8.75	0.70	0.74	6.75	6.69	1.37
	1999	0.08	0.21	7.18	1.15	1.03	11.56	17.00	0.53
	2000	0.10	0.09	10.63	1.45	0.54	7.24	9.39	1.70
	2001	0.05	0.04	6.41	0.51	0.36	3.67	2.73	4.81
	2002	0.24	0.05	7.54	1.31	0.71	4.72	4.77	21.63
NL0009R	1990	1.59				32.60			
	1991	3.40				39.76			
	1992	3.72				29.31			
	1993	4.60				27.52			
	1994	2.09	0.37			17.16		50.56	
	1995	2.05	0.27			14.95		39.42	
	1996	1.48	0.39			14.31		49.02	
	1997	1.06	0.31			11.83		33.44	
	1998	0.79	0.29			9.76		29.77	
	1999	0.79	0.29			9.76		29.77	
	2000	0.62	0.20			8.98		32.45	
	2001	0.66	0.18			7.15		30.57	

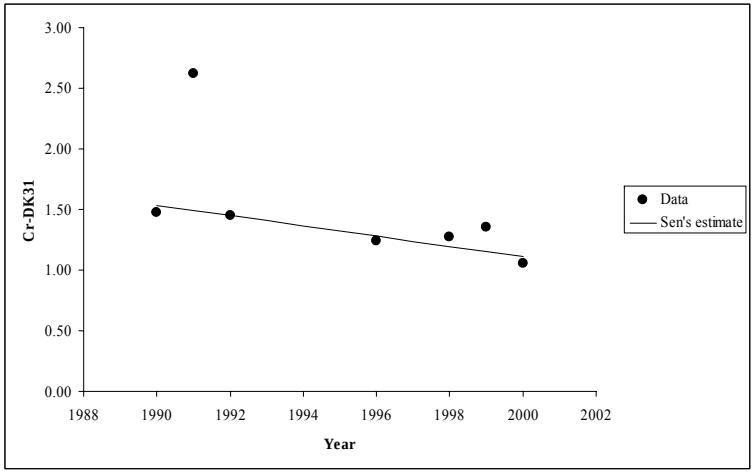
Station code	year	As	Cd	Cr	Cu	Pb	Ni	Zn	Hg
	2002	0.78	0.20			7.70		31.04	
NO0042G	1994	0.58	0.10	0.68	0.41	1.25	0.52	2.28	1.79
	1995	0.17	0.03	0.31	0.50	0.83	0.25	1.55	1.62
	1996	0.08	0.01	0.91	0.38	0.52	0.30	2.03	1.59
	1997	0.24	0.03		0.47	0.70	0.30	2.25	1.19
	1998	0.16	0.03	0.40	0.37	0.72	0.20	1.46	1.55
	1999	0.10	0.03	0.34	0.42	0.62	0.30	1.88	1.76
		0.08	0.02	0.13	0.28		0.14	1.39	
	2000	0.32	0.03	0.12	0.52	0.73	0.16	1.73	1.50
	2001	0.40	0.02	0.06	0.33	0.50	0.08	1.39	1.56
	2002	0.39	0.04	0.08	0.34	0.77	0.11	1.88	1.60
NO0099R	1991	0.77	0.06	1.86	0.80	2.69	0.59	4.40	
	1992	0.19	0.05	1.79	0.47	2.35	1.33	3.93	2.06
	1993	0.41	0.07	3.70	0.85	3.67	0.81	6.98	1.84
	1994	0.36	0.07	2.80	0.90	3.68	0.88	4.53	1.84
	1995					1.02	0.25	2.76	
				1.25	0.41	2.74	1.36	4.85	
	1996			1.00	0.48	0.79	0.26	1.54	
				0.37	0.42	2.96	0.58	4.40	
	1997			0.82	0.73	0.73	0.23	2.17	
				1.33	0.53	2.58	0.67	5.08	
	1998			2.83	0.42	0.69	0.44	3.39	
				1.46	0.42	2.02	0.43	4.02	
	1999			1.37	0.47	0.47	0.26		1.86
				0.40	0.27	1.76	0.30	3.89	
	2000			0.72	0.36	0.52	0.11	1.78	1.67
	2001	0.61	0.01	1.73	0.34	0.63	0.14	1.62	1.65
			0.06	1.25	0.40	2.81	0.61	5.69	
	2002	0.06	0.01	1.63	0.44	0.58	0.15	1.78	1.64
		0.24	0.05	0.38	0.51	2.62	0.55	4.38	
SE0002R	1997								1.35
	1998								1.55
	1999								1.41
	2000								1.36
	2001								1.66
	2002								1.67
SE0014R	2002								14.07
									1.67

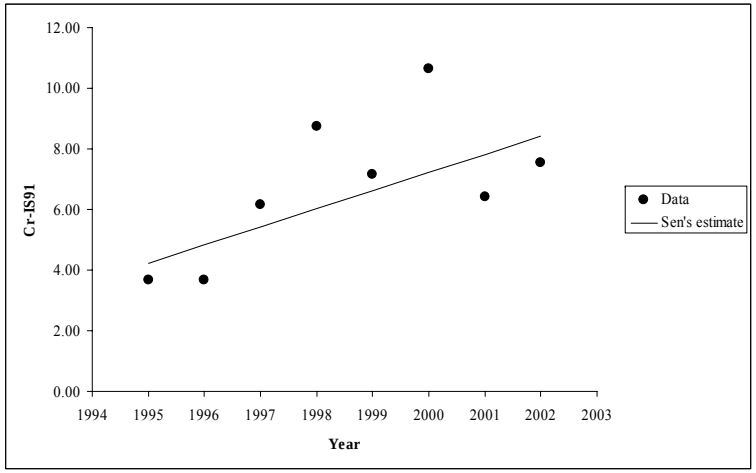


Time series of arsenic on aerosol, stations DK0031R (top left), GB0090R (top right), GB0091R (bottom left) and NL0009R (bottom right).

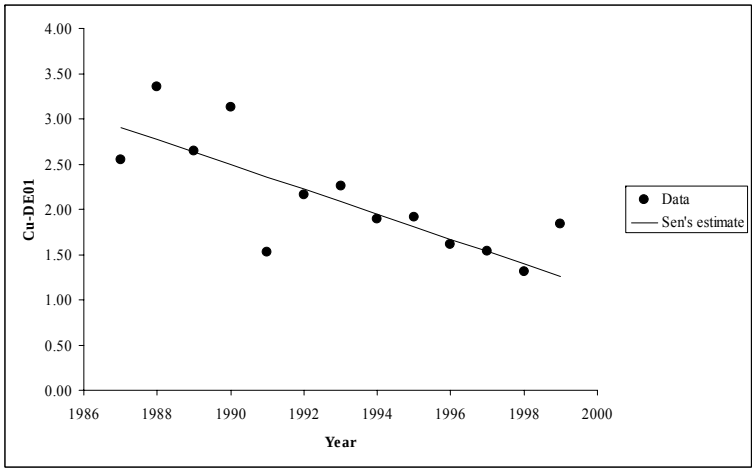


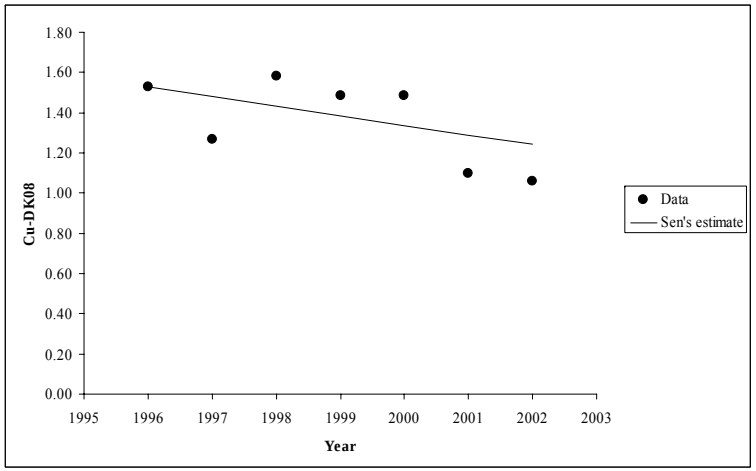
Time series of cadmium on aerosol, stations DE0001R (top left), NL0009R (top right), GB0091R (bottom left) .



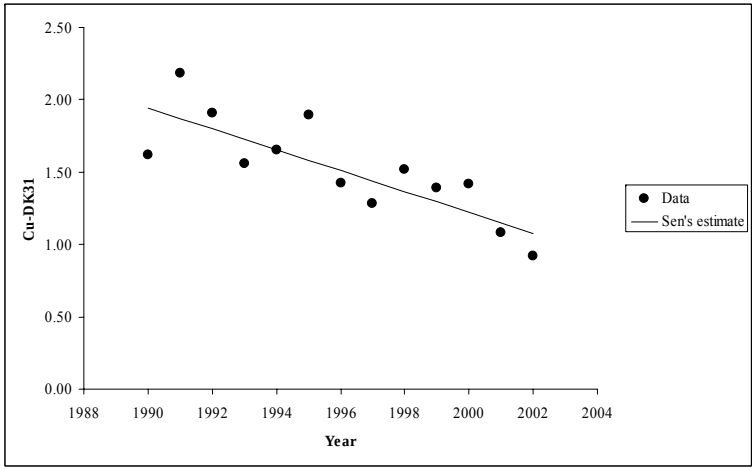


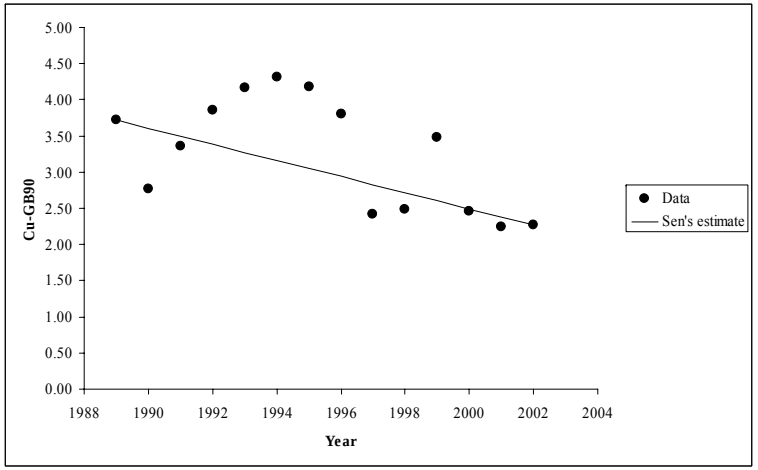
Time series of chromium on aerosol, stations DK0031R (left), IS0091R (right).



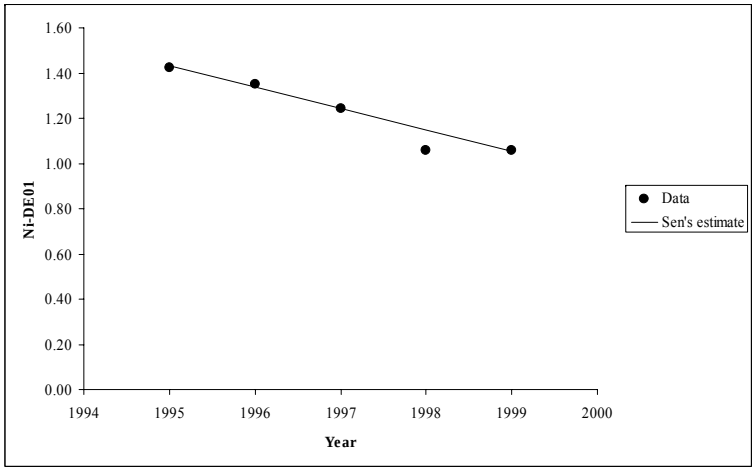


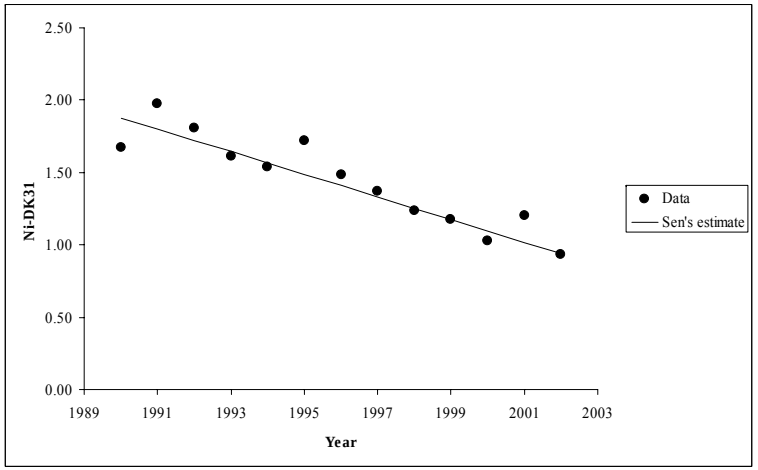
Time series of copper on aerosol, stations DE0001R (left), DK0008R (right).



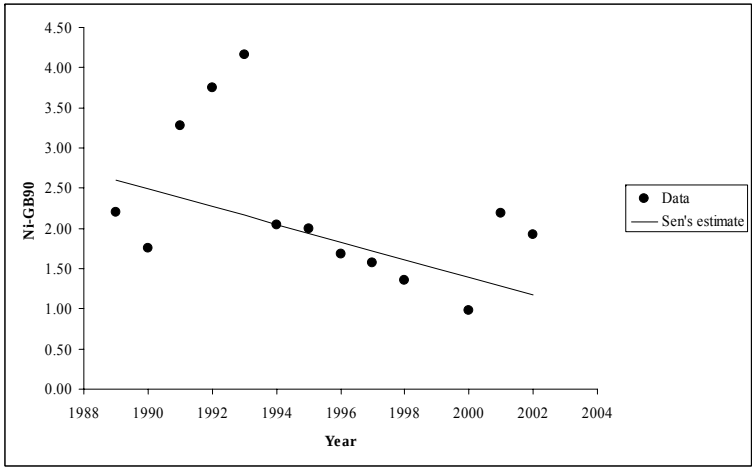


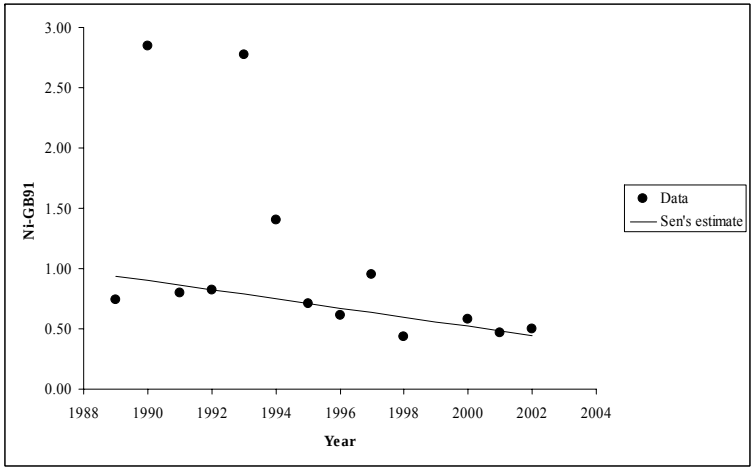
Time series of copper on aerosol, stations DK0031R (left), GB0090R (right).



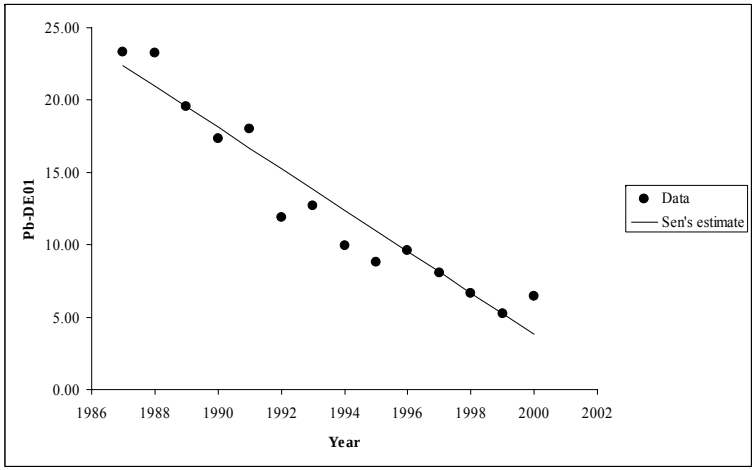


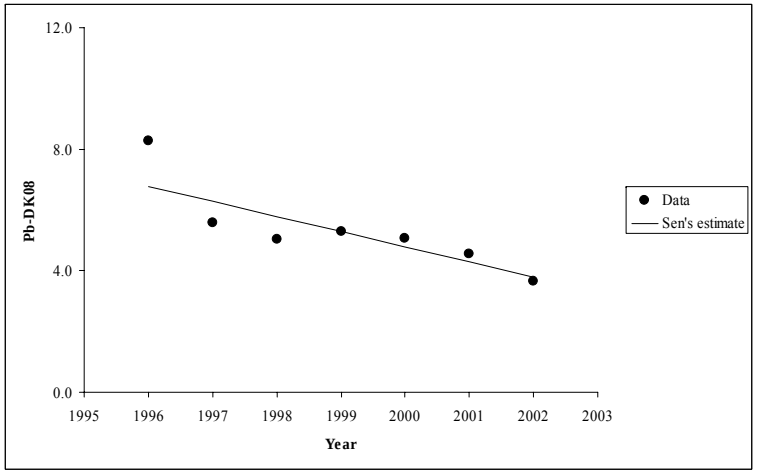
Time series of nickel on aerosol, stations DE0001R (left), DK0031R (right).



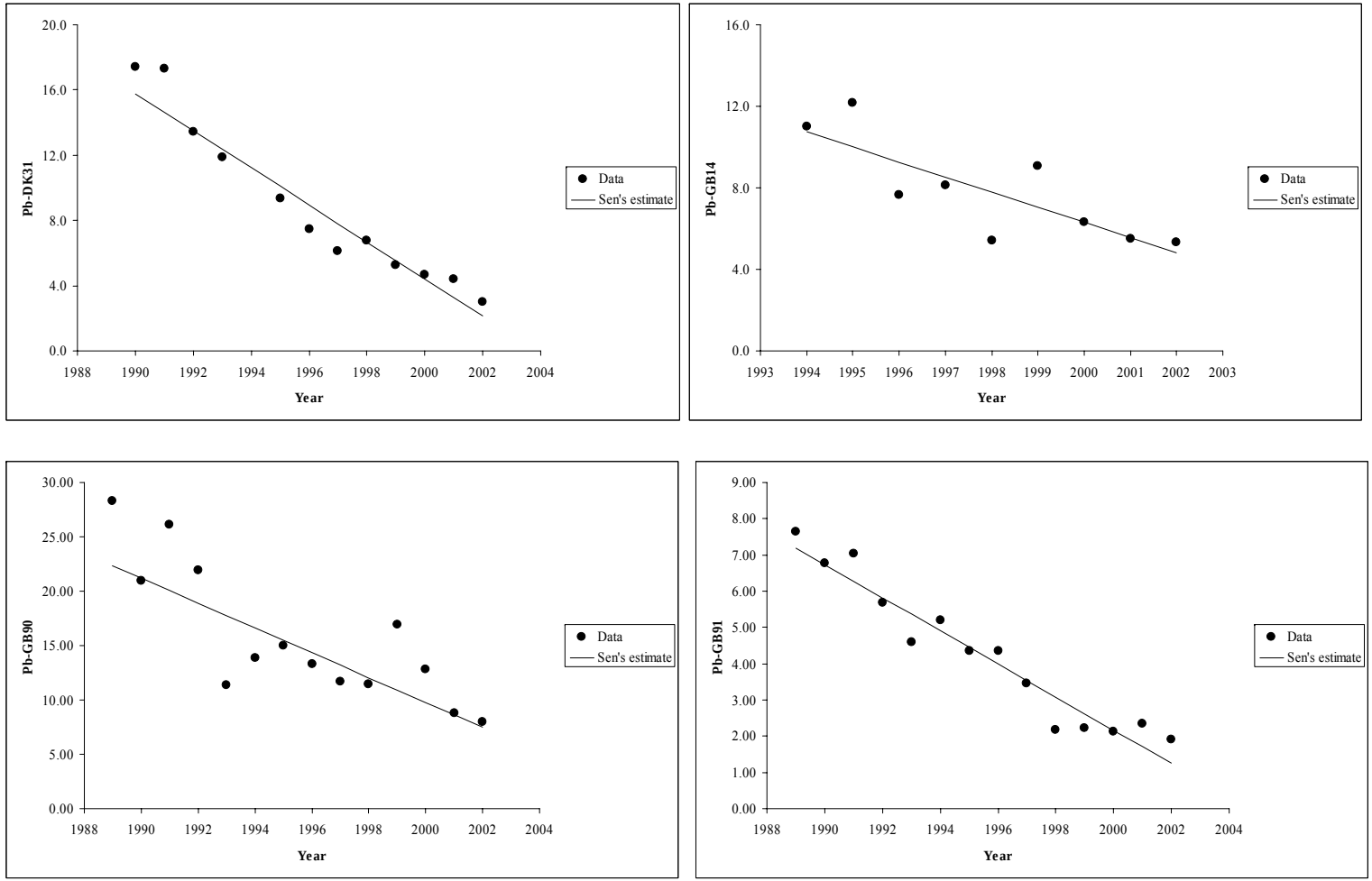


Time series of nickel on aerosol, stations GB0090R (left), GB0091R (right).

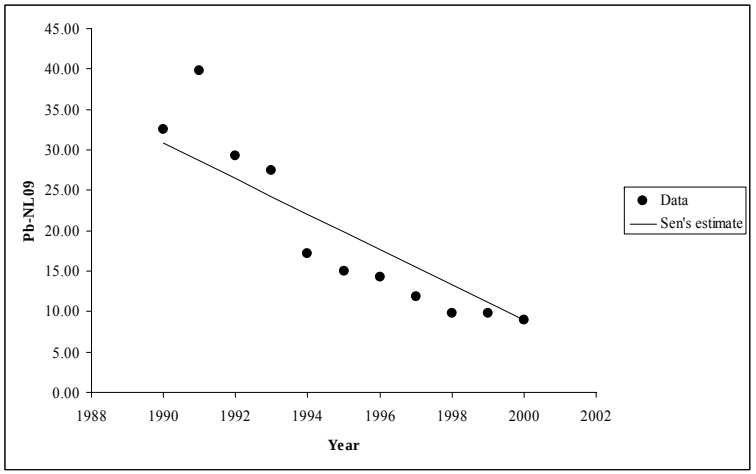




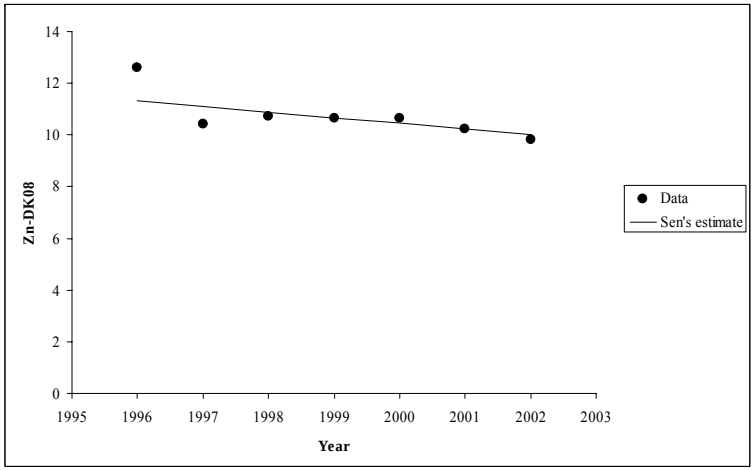
Time series of lead on aerosol, stations DE0001R (left), DK0008R (right).

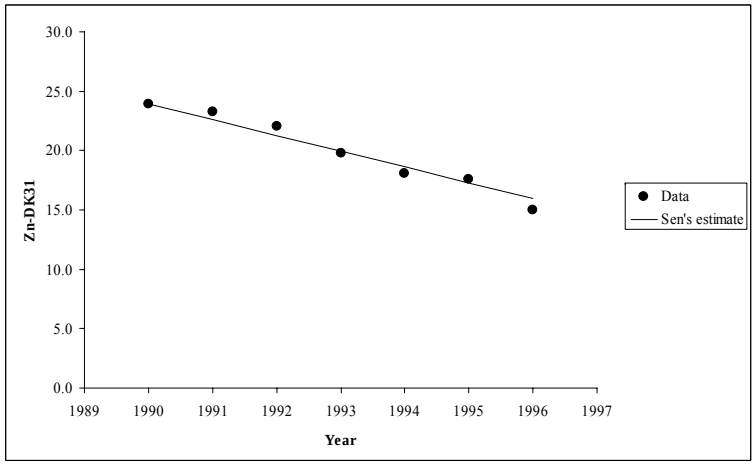


Time series of lead on aerosol, stations DK0031R (top left), GB0014R (top right), GB0090R (bottom left) and GB0091R (bottom right).

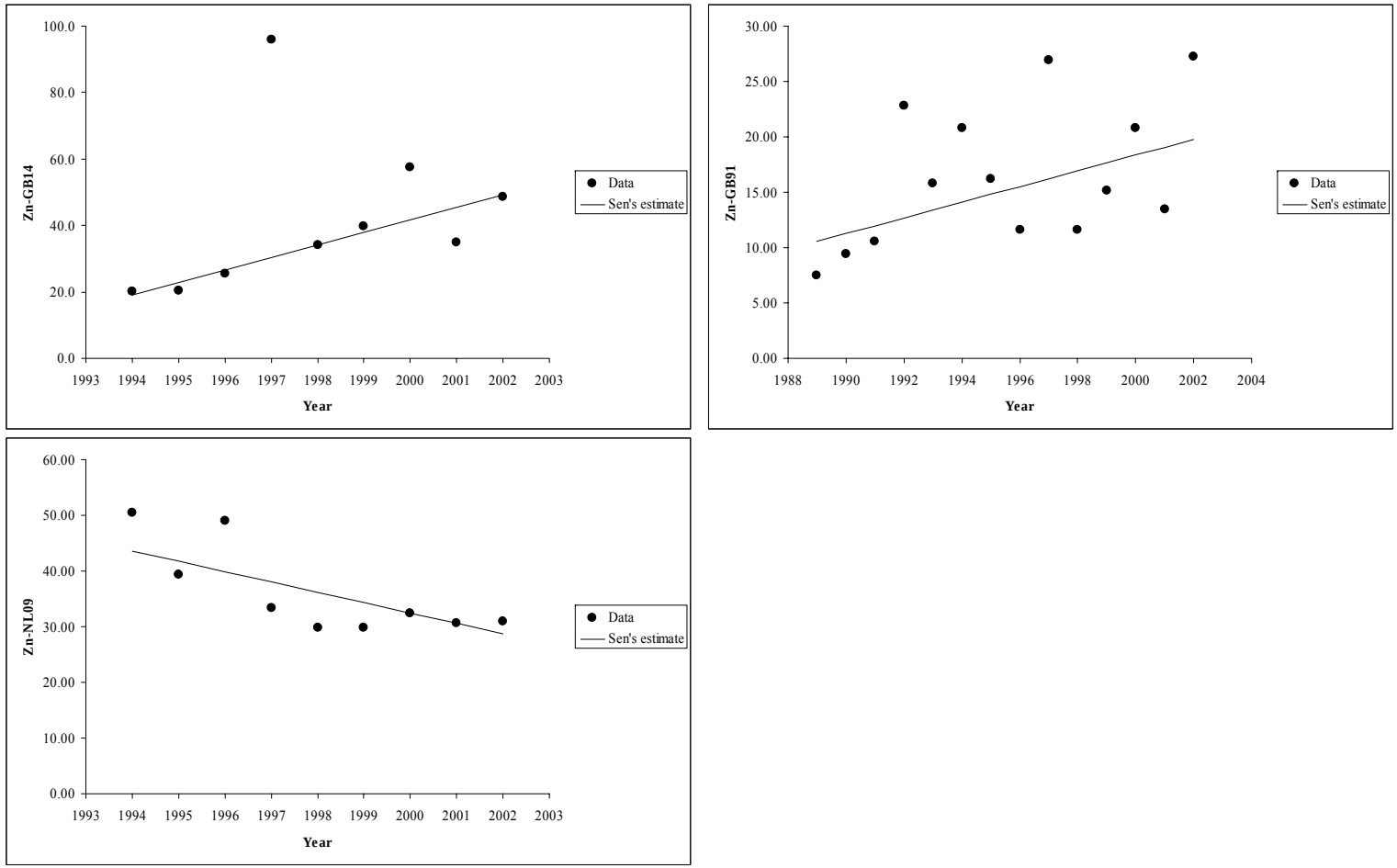


Time series of lead on aerosol, stations NL0009R





Time series of zinc on aerosol, stations DK0008R (left), DK0031R (right).



Time series of zinc on aerosol, stations GB0014R (top left), GB0091R (top right), NL0009R (bottom).

Annex 9. Comparison of RTrend and EMEP trend-tool

The statistical package Rtrend Version 3.2.0.1 which was designed for the assessment of trends in riverine loads was used to investigate trends in the precipitation concentrations of a number of data sets with and without the outliers identified in the quality assurance checks removed.

RTrend is not designed for the analysis of data arising from atmospheric measurements. In consequence its use for this purpose is not straightforward and a number of dummy parameters are required to be entered into the database in order for the statistical analysis to be possible.

RTrend produces an output which contains a number of useful measures of trend. The attached table is a summary of the results. These include the estimated trend using both the Mann-Kendall and LOESS approaches, the probability of these trends being due to random variation in the data and the power to detect a trend of 50% and the minimum detectable trend.

Those data sets identified as having zero outliers are the ones which are quality assured. The investigations with the complete datasets show that the removal of outliers can change not just the size of a trend but the direction as well depending on the timing and scale of the incorrect data point(s). Hence the two Westerland datasets when an early high outlier is removed the trend reverses from a significant downward trend to a upward, but not significant trend.

The size of the trend which is required in order that RTrend can identify it as significant is generally large. Few of the dataset would allow detection of a trend of 50%. This does not change when outliers are removed as the statistical analysis can be biased by the presence of a small number of data points into detecting a incorrect trend. Possibly as a result of the variability in the datasets even after the removal of outliers. Hence policy measures which are not large in impact may not be detectable through atmospheric monitoring.

Conclusions

RTrend is not applied easily to the analysis of CAMP data. It could be developed for this purpose but other tools are available with similar functionality.

The detection and sign of the trends in the CAMP datasets used varied on whether or not outliers were included. Hence thorough quality assurance of the CAMP dataset is to be encouraged.

It will take around a decade for policy measures which produce changes in atmospheric concentrations of around 50% to produce statistically significant trends in atmospheric concentrations.

					LOESS Analysis for the Level				LOESS Analysis for the Level			MK and Thiel slopes			Power Analysis	
	Site	Pollutant	No of Outliers	Years	Estimated Level LOESS at start	Estimated Level LOESS at end	relative change in level %	p value for change	Estimated annual linear trend %	Estimated change of the level of the linear trend %	p value for linear trend	Estimated annual trend %	Estimated change of the level of the Theil slope %	P value for Mann Kendal test	Power to detect a change of 50%	Detectable change %
1	Westerland	Cu	1	1990-2002	1.233	0.965	-21.8	0.743	0.1	1.7	0.966	2.2	30.3	0.76	0.2	182.7
2	**Westerland**	**Cu**	**0**	**1990-1999, 2001-2002**	1.233	1.397	13.3	0.759	3	35.9	0.21	2.9	40.9	0.428	0.342	118.7
3	Westerland	Pb	1	1990-2002	3.987	-0.071	-101.8	0.751	-4.8	-57.9	0.66	-6.2	-53.4	0.024	0.069	892.5
4	**Westerland**	**Pb**	**0**	**1990-1995, 1997-2002**	2.261	0.985	-56.4	0.042	-4.9	-58.7	0.014	-5	-46.3	0.004138	0.607	76.4
5	**Westerland**	**Zn**	**0**	**1990-2002**	11.173	12.815	14.7	0.747	3.7	44.7	0.295	4.4	68.2	0.246	0.271	141.9
6	**Ulborg**	**Cr**	**0**	**1989-2002**	0.397	0.065	-83.6	0.012	-5.2	-67.7	0.004224	-5	-48.3	0.009	0.541	84
7	**Ulborg**	**Pb**	**0**	**1989-2002**	3.601	0.593	-83.5	0.00000865	-6.4	-83.6	0.00000422	-5.8	-54.1	0.0000317	0.994	35.1
8	**Ulborg**	**Zn**	**0**	**1989-2002**	13.37	6.767	-49.4	0.032	-3	-39.4	0.027	-3.8	-39.9	0.063	0.735	64.6
9	**Porspoder**	**Cu**	**0**	**1989-2002**	2.225	0.807	-63.7	0.021	-4.9	-63.7	0.072	1	14	0.913	0.479	92.3
10	**Porspoder**	**Cu**	**0**	**1990-2002**	0.942	0.807	-14.3	0.626	0.3	3.3	0.917	3.8	57.2	0.36	0.435	99.1
11	**Porspoder**	**Ni**	**0**	**1989-2002**	0.651	0.507	-22.2	0.51	-1.9	-25.1	0.408	-0.7	-9.1	0.381	0.405	104.8
12	East Ruston	As	3	1990-2002	0.472	0.163	-65.5	0.00265	-5.8	-69.3	0.001339	-6.1	-52.7	0.001862	0.833	56
13	East Ruston	As	2	1991-2002	0.406	0.163	-60	0.015	-5.8	-63.9	0.008	-6.2	-50.8	0.007	0.698	67.8
14	East Ruston	As	1	1991-1995, 1997-2002	0.3664	1.417	-61.3	0.049	-5.4	-59.7	0.067	-3.4	-31.3	0.193	0.429	100.2
15	East Ruston	As	1	1991-2000	0.408	0.207	-49.4	0.043	-5.8	-52.4	0.038	-6.6	-45.8	0.032	0.667	70.6
16	**East Ruston**	**As**	**0**	**1991-1995, 1997-2000**	0.3664	1.478	-59.7	0.079	-6.2	-56.2	0.127	-3.8	-29.3	0.474	0.376	110.9
17	East Ruston	As	2	1990-1995, 1997-2002	0.472	0.163	-65.5	0.00265	-5.8	-69.3	0.001339	-6.1	-52.7	0.001862	0.833	56
18	East Ruston	As	1	1990-1995, 1997-2000	0.472	0.205	-56.5	0.009	-5.9	-59.2	0.008	-6.5	-49	0.013	0.811	58
19	East Ruston	As	2	1990-2000	0.472	0.207	-56.3	0.009	-6	-59.6	0.008	-7	-51.7	0.008	0.817	57.4
20	East Ruston	Cu	1	1990-2002	3.074	1.622	-47.2	0.474	-3.2	-38	0.482	-2.6	-27.4	3.6	0.167	216.4
21	**East Ruston**	**Cu**	**0**	**1990-1995, 1997-2002**	3.205	1.417	-55.8	0.144	-4.3	-52	0.151	-2.7	-28.2	0.161	0.33	122
22	East Ruston	Pb	1	1990-2002	6.088	1.589	-73.9	0.001665	-6.6	-79.1	0.0003686	-6.4	-55	0.0007924	0.829	56.4
23	**East Ruston**	**Pb**	**0**	**1990-1995, 1997-2002**	6.075	1.61	-73.5	0.00169	-6.5	-78.3	0.0003705	-5.9	-52	0.0005061	0.832	56.1
24	**Valentia Obs**	**Zn**	**0**	**1992-2002**	16.901	39.459	133.5	0.146	22.5	225.2	0.069	-237.3	2286	0.087	0.134	273.5
25	Kollumerwaard	Pb	4	1990-2002	2.694	1.461	-45.8	0.232	-4.2	-50.8	0.091	-5.4	-48.5	0.02	0.379	109.8

					LOESS Analysis for the Level				LOESS Analysis for the Level			MK and Thiel slopes			Power Analysis	
	Site	Pollutant	No of Outliers	Years	Estimated Level LOESS at start	Estimated Level LOESS at end	relative change in level %	p value for change	Estimated annual linear trend %	Estimated change of the level of the linear trend %	p value for linear trend	Estimated annual trend %	Estimated change of the level of the Theil slope %	P value for Mann Kendal test	Power to detect a change of 50%	Detectable change %
26	Kollumerwaard	Pb	3	1991-2002	2.026	1.461	-27.9	0.489	-3.5	-38.7	0.171	-4.7	-40.8	0.055	0.398	106.1
27	**Kollumerwaard**	**Pb**	**0**	**1991-1999**	2.026	1.659	-18.1	0.698	-2.5	-20	0.584	-5.5	-36.5	0.175	0.312	128
28	Kollumerwaard	Pb	1	1990-1999	2.694	1.659	-38.4	0.372	-4.3	-38.3	0.3	-6.6	-46.1	0.074	0.312	127.7
29	**de Zilk**	**Hg**	**0**	**1996-2002**	23.273	8.061	-65.4	0.004293	-11.9	-71.5	0.003907	-12.7	-55.7	0.002667	0.888	51.1
30	**Birkenes**	**Pb**	**0**	**1990-2002**	3.742	1.031	-72.5	0.000004878	-6.3	-75.5	0.000001256	-7.3	-60	0.0000148	1	25.6
31	**Birkenes**	**Zn**	**0**	**1990-2002**	8.341	3.784	-54.6	0.001499	-4.7	-56.6	0.0009386	-4.6	-42.9	0.0007922	0.963	42.6
32	Lista	As	4	1991-2002	1.497	0.234	-84.4	0.016	-8.8	-96.6	0.029	-6.7	-53.2	0.04	0.35	116.7
33	Lista	As	3	1992-2002	0.454	0.234	-48.4	0.155	-4.7	-46.6	0.12	-4.6	-37.8	0.139	0.428	100.5
34	Lista	As	2	1992-1993 1995-2002	0.483	0.234	-51.6	0.079	-5.1	-51.4	0.05	-5.2	-41.2	0.043	0.549	83
35	Lista	As	1	1992-1997 2000-2002	0.452	0.231	-49	0.149	-4.5	-45.1	0.127	-4.9	-39.7	0.139	0.431	99.9
36	**Lista**	**As**	**0**	**1992-1993 1995-1997 2000-2002**	0.489	0.23	-53.1	0.067	-5	-49.7	0.049	-4.8	-38.7	0.043	0.569	80.7
37	Lista	As	3	1991-1993 1995-2002	1.507	0.234	-84.5	0.017	-8.8	-97.3	0.022	-8.1	-60.7	0.004923	0.362	113.9
38	Lista	As	1	1991-1993 1995-1997 2000-2002	1.51	0.228	-84.9	0.017	-8.6	-94.8	0.025	-7.3	-56.8	0.024	0.358	114.9
39	Lista	As	2	1991-1997 2000-2002	1.497	0.231	-84.6	0.016	-8.7	-95.8	0.03	-6.7	-53.5	0.04	0.351	116.6
40	V D Casterlo	Cu	6	1989-2002	2.111	3.042	44.1	0.251	2.8	36.4	0.26	0.4	4.7	0.743	0.381	109.7
41	V D Casterlo	Cu	3	1992-2002	2.056	3.042	48	0.252	4.3	43	0.266	0.4	3.6	0.64	0.346	117.8
42	**V D Casterlo**	**Cu**	**0**	**1992-1999**	2.056	2.516	22.4	0.295	3.1	21.8	0.289	4.5	36.2	0.536	0.769	61.6
43	V D Casterlo	Cu	3	1989-1999	2.111	2.516	19.2	0.405	1.6	15.8	0.446	0.9	9.3	0.755	0.7	67.6
44	Rorvik	Hg	1	1989-2001	25.729	8.467	-67.1	0.002185	-5.7	-68.1	0.003599	-5.1	-46.9	0.001862	0.787	60
45	**Rorvik**	**Hg**	**0**	**1989-1994 1998-2001**	25.981	8.901	-65.7	0.00003668	-6	-72.2	0.00005611	-5.2	-47.2	0.000198	0.997	33.3
46	Gardsjon	Pb	1	1995-2002	2.482	1.125	-54.7	0.002871	-8	-55.8	0.002677	-8.6	-46.7	0.019	0.983	38.7
47	**Gardsjon**	**Pb**	**0**	**1996-2002**	2.3	1.105	-52	0.008	-9.1	-54.5	0.006	-9.7	-46	0.035	0.954	43.7
48	Svartdalen	Ni	1	1987-1995	0.487	0.292	-40	0.018	-5.1	-41	0.019	-6	-38.9	0.048	0.949	44.6
49	**Svartdalen**	**Ni**	**0**	**1987-1994**	0.487	0.316	-35.2	0.049	-5.3	-36.8	0.046	-6	-35.4	0.108	0.895	50.5

Data sets shown in bold are those containing no outliers

Annex 10. Comparison of pollution loads to Region I - II by riverine input and direct discharges and atmospheric deposition

Table 10.1. Nitrogen load to OSPAR Region I by direct discharges, riverine inputs and atmospheric deposition

year	Direct discharges of Total-N in kt/a	Riverine inputs of Total-N in kt/a	Direct discharges and riverine inputs of Total-N in kt/a	Atmospheric deposition Total-N in kt/a
1990	8	32	41	409
1991	9	26	35	
1992	9	31	40	
1993	10	29	40	
1994	12	27	39	
1995	14	31	45	364
1996	16	26	41	505
1997	18	30	49	378
1998	18	29	47	283
1999	18	31	49	323
2000	20	25	45	319
2001	18	32	50	301
2002	22	29	51	

Table 10.2. Nitrogen load to OSPAR Region II, Greater North Sea by direct discharges, riverine inputs and atmospheric deposition

year	Direct discharges of Total-N in kt/a	Adjusted riverine inputs of Total-N in kt/a	Direct discharges and adjusted riverine inputs of Total-N in kt/a	Atmospheric deposition Total-N in kt/a
1990	112	1054	1128	534
1991	103	1077	1182	
1992	101	1098	1276	
1993	99	1119	1194	
1994	90	1126	1233	
1995	91	1125	1207	483
1996	85	1113	1194	562
1997	85	1090	1256	513
1998	81	1066	1129	523
1999	81	1034	1055	469
2000	78	1003	1133	550
2001	79	974	1012	486
2002	79	947	1049	

Table 10.3. Nitrogen load to OSPAR Region II, Celtic Seas by direct discharges, riverine inputs and atmospheric deposition[(a)]

year	Direct discharges of Total-N in kt/a	Riverine inputs of Total-N in kt/a	Direct discharges and riverine inputs of Total-N in kt/a	Atmospheric deposition Total-N in kt/a
1990	45	243	288	128
1991	41	269	310	
1992	53	243	295	
1993	45	267	312	
1994	48	281	329	
1995	43	254	298	146
1996	38	286	324	180
1997	40	281	321	155
1998	46	298	343	129
1999	38	126	163	115
2000	38	132	171	116
2001	35	105	140	126
2002	32	220	252	

(a) For the year 1999-2001 riverine loads from Ireland (estimated to be around 150 ktonne N/a) are missing.