Convention on Long-range Transboundary Air Pollution

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Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe

TECHNICAL REPORT
MSC-W 2/2014

EMEP Centres Joint Report for HELCOM EMEP/MSC

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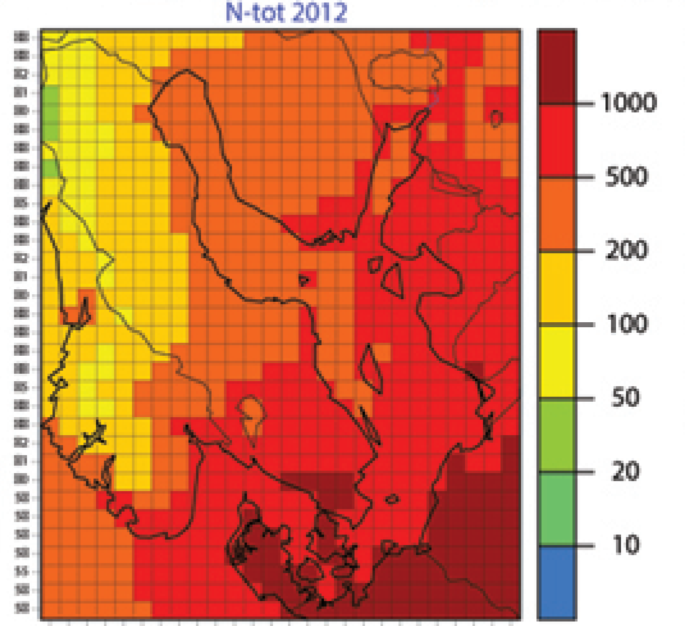

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EMEP Centres Joint Report for HELCOM
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(DRAFT)

Atmospheric Supply of Nitrogen, Lead, Cadmium, Mercury and Dioxins/Furans to the Baltic Sea in 2014

Semeena Valiyaveetil Shamsudheen[1], Jerzy Bartnicki[1],
Alexey Gusev[2], Wenche Aas[3]

[1]Meteorological Synthesizing Centre-West (MSC-W)
[2]Meteorological Synthesizing Centre-East (MSC-E)
[3]Chemical Coordinating Centre (CCC)

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Summary

The results presented in this EMEP Centres Joint Report for HELCOM are based on the modelling and monitoring data presented to the 38th Session of the Steering Body of EMEP in Geneva in September 2014. It includes measurements, as well as emissions and depositions calculated by the EMEP models of nitrogen compounds, heavy metals and PCDD/F for the year 2012.

Until 2012, the Baltic Sea basin was sub-divided into six sub-basins. In 2013, new nine sub-basins were introduced for the first time. The new sub-basins of the Baltic Sea have been used for computing all atmospheric nitrogen depositions presented in this report. The are listed below in alphabetical order, together with their abbreviations and surface areas.

Sub-basin	Abbreviation	Area in km^2
Archipelago Sea	ARC	13405
Baltic Proper	BAP	209258
Bothnian Bay	BOB	36249
Bothnian Sea	BOS	65397
Gulf of Finland	GUF	29998
Gulf of Riga	GUR	18646
Kattegat	KAT	23659
The Sound	SOU	2328
Western Baltic	WEB	18647
Baltic Sea basin	**BAS**	**417587**

The measured monthly and annual 2012 concentrations in air and precipitation for nitrogen species and heavy metals are presented in the report. For most components a significant south-east gradient can be noticed in the measured concentrations in 2012. Further the concentration levels seem to be higher in southwest than southeast for the nitrogen components, maybe due to influence of the extensive traffic (ship as well as cars) and agricultural activities in this region.

The temporal patterns of monthly Cd and Pb concentrations in air show a winter maximum, similar tendency for elemental Hg. Also nitrogen concentration in air show elevated levels in the spring and generally higher concentrations in winter than summer. These elevated concentrations in winter occur probably due to longer atmospheric residence time and reduced vertical mixing. The seasonal patterns in precipitation are not as strong as for airborne components. This is due to the presence of the precipitation

effect, but there is a maxima of reduced nitrogen wet deposition in summer due to enhanced agricultural activities.

Annual emissions from the HELCOM Contractig Parties in 2012 are shown below for all pollutants considered in the report.

Country/ship	POLLUTANT					
	NO_2 kt N	NH_3 kt N	Cd tonnes	Pb tonnes	Hg tonnes	PCDD/F g TEQ
Denmark	35	63	0.2	11.8	0.3	23
Estonia	10	9	0.6	33.6	0.6	4
Finland	45	30	1.3	18.6	0.8	14
Germany	387	449	5.6	185	10.4	67
Latvia	11	16	0.6	3.7	0.1	32
Lithuania	18	31	0.6	4.7	0.4	24
Poland	249	217	38.7	553.6	10	278
Russia	901	1115	22.6	32	1	NA
Sweden	40	42	0.5	11	0.5	38
HELCOM	**1695**	**1972**	**71**	**855**	**24**	**1379**
Ship-Baltic	105					

Annual depositions of all considered pollutants in 2012 are shown in the Table below for the new, nine sub-basins of the Baltic Sea and for the entire Baltic Sea.

Basin	POLLUTANT					
	Ox-N kt N	Red-N kt N	Cd tonnes	Pb tonnes	Hg tonnes	PCDD/F g TEQ
ARC	3,9	2,7	0.2	5	0.09	11
BOB	5,7	4,0	0.2	6	0.18	20
BOS	12,6	7,7	0.6	14	0.34	29
BAP	72,1	59,5	3.9	90	1.42	124
GUF	9,0	6,2	0.5	11	0.2	39
GUR	6,4	4,9	0.3	7	0.12	28
KAT	9,3	10,5	0.4	11	0.17	16
SOU	1,1	1,2	0.04	1	0.02	7
WEB	7,6	11,0	0.3	9	0.14	27
BAS	**119,0**	**97,2**	**6.4**	**153**	**2.68**	**300**

Compared to 2011, nitrogen oxides emissions in 2012 are lower (1-13%) in six out of nine HELCOM Contracting Parties. These are Denmark, Estonia, Finland, Germany, Poland and Sweden. Other three countries, Latvia, Lithuania and Russia reported increased (2-15%) nitrogen oxides emissions in the 2012 compared to 2011. Ship emissions from the Baltic Sea were also 1.6% higher.

Annual 2012 ammonia emissions are higher than that of 2011 ammonia emissions in five out of nine HELCOM countries: Denmark, Estonia, Latvia, Lithuania and Russia. The 2012 emissions are lower than 2011 emissions in the rest four CPs: Finland (2%), Germany (3%), Poland (3%) and Sweden (2%).

Among the HELCOM Contracting Parties, the largest per cent of 2012 nitrogen emissions deposited to the Baltic Sea basin can be noticed for Denmark (16.3%) and the lowest for Russia (0.6%).

Calculated annual deposition of total nitrogen to the Baltic Sea basin in 2012 is 233 kt, approximately 8% higher than in 2011. Deposition of oxidised nitrogen was 6% higher and deposition of reduced nitrogen is 10.5% higher in 2012 compared to 2011. Deposition of oxidized nitrogen accounts for 53% of total nitrogen deposition in 2012.

Normalised nitrogen depositions to the Baltic Sea have been calculated for the first time in 2013. Normalised depositions of oxidized, reduced and total nitrogen to the Baltic Sea show clear decreasing pattern in the period 1995-2012.

Germany, Poland, ship traffic on the North Sea and on the Baltic Sea are the main emission sources contributing to oxidized nitrogen deposition into the Baltic Sea basin in 2012.

As in previous three years Germany, Poland and Denmark are top three sources contributing to reduced nitrogen deposition into the Baltic Sea basin in 2012.

As in previous years, also in 2012 some distant sources like United Kingdom, France and ship traffic on the North Sea contribute significantly to nitrogen deposition into the Baltic Sea basin.

The main sources contributing to total nitrogen deposition to the Baltic Sea basin are: Germany, Poland, Denmark, and Sweden. Compared to 2011, contribution from the United Kingdom is lower by 23% and contribution from Russia is higher by 47% in 2012. Contribution of other distant sources like ship traffic on the North Sea, France and the Netherlands is also significant.

The results of the EMEP/MSC-W model are routinely compared with available measurements at EMEP and HELCOM stations. The comparison of calculated versus measured data indicates that the model predicts the observed air concentrations and depositions of nitrogen compounds within the accuracy of approximately 30%.

Annual 2012 emissions of cadmium, lead, mercury, and dioxins and furans have slightly increased comparing to 2011 (by 1.6%, 0.4%, 2%, and 0.2%).

Levels of cadmium and lead deposition to the entire Baltic Sea have declined in 2012 comparing to 2011 by 0.6% and 12%, respectively. At the same time deposition of mercury and PCDD/Fs to the Baltic Sea has increased by 11% and 9% respectively from 2011 to 2012

Anthropogenic emission sources of HELCOM countries contributed to annual deposition over the Baltic Sea in 2012 about 30% for lead (30% higher than that of 2011) and about 16% for mercury, respectively. For cadmium and PCDD/Fs this contribution is accounted for 51% (27% higher than 2011) and 36%. Among the HELCOM countries the most significant contribution to deposition of HMs and PCDD/Fs to the Baltic Sea in 2012 was made by Poland, Russia and Germany.

Along with anthropogenic emission sources of HELCOM countries essential contribution to total annual deposition was made by other sources, in particular, natural emissions, re-suspension with dust, distant emissions, and re-emission (about 40-80%).

Modelling results in comparison with available measurements for 2012 made around the Baltic Sea are within an accuracy of a factor of two for Pb and Cd, and 25% for Hg.

Preface

The Co-operative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) and the Baltic Marine Environment Protection Commission (HELCOM) are both conducting work on air monitoring. modelling and compilation of emission inventories. In 1995, HELCOM decided to rationalize its current programs by avoiding duplication of efforts with specialised international organizations. At the request of HELCOM, the steering Body of EMEP at its nineteenth session agreed to assume the management of atmospheric monitoring data. the preparation of air emission inventories and the modelling of air pollution in the Baltic region.

Following the coordination meeting held in Potsdam in Germany and the Pollution Load Input meeting held in Klaipeda-Joudkrante in Lithuania, both 1996. it was agreed that EMEP Centres should be responsible for regular evaluation of the state of the atmosphere in the Baltic Sea region and should produce an annual joint summary report which includes updated emissions of selected air pollution, modelled deposition fields, allocation budgets and measurement data.

This report was prepared for HELCOM. Based on model estimates and monitoring results presented to the 38th session of the Steering Body of EMEP. Following decision of the HELCOM /MONAS-10 Meeting, it presents the results for the year 2012.

Acknowledgements

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Contents

1. Introduction

The first EMEP Centres Joint Report for HELCOM was delivered in 1997 (Tarrason *et al*. 1997) and was followed by fourteen annual reports (Bartnicki *et al*. 1998, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010, 2011 and 2012). The present EMEP Centres Joint Report for HELCOM is focused on the year 2012. It is based on the modelling and monitoring data presented to the 38th Session of the Steering Body of EMEP in Geneva in September 2014.

Following decisions of the 9th HELCOM MONAS Meeting held in Silkeborg in 2007, the main deliverables expected from the EMEP Centres are the Indicator Fact Sheets for nitrogen, heavy metals and PCDD/Fs. These Indicator Fact Sheets include time series of emissions and depositions of selected pollutants, and can be found on the HELCOM web pages (links shown in Appendix C). In this report we present additional important information about emissions, depositions and source allocation budgets for nitrogen, heavy metals and PCDD/Fs in the year 2012.

Eight countries have submitted data from all together Nineteen HELCOM stations for 2012 (Fig. 2.1), which is the same number as in 2011. The stations are distributed in eight of the nine sub-basins (Fig. 2.1). Not all sites measure all HELCOM relevant parameters. Sixteen sites measure oxidized nitrogen and fourteen sites measure reduced nitrogen in air, and fifteen sites measure both oxidized and reduced nitrogen in precipitation. For heavy metals there were eleven stations with cadmium and/or lead in air, and twelve stations in precipitation, though these sites are not necessarily co-located. There were six sites with mercury measurements in precipitation and four in air. All the data can be downloaded from ebas.nilu.no.

The EMEP model has been used for all nitrogen computations presented here (Simpson et al., 2012). In 2011, the model name has been changed from EMEP Unified to EMEP/MSC-W model. The earlier model versions have been documented in detail in EMEP Status Report 1/2003 Part I (Simpson *et al.* 2003) and in EMEP Status Report 1/2004 (Tarrasón *et al*., 2004). In EMEP Status Report 1/2003 Part II (Fagerli *et al.* 2003) we presented an extensive evaluation of the acidifying and eutrophying components for the years 1980, 1985, 1990 and 1995 to 2000. In EMEP Status Report 1/2003 Part III (Fagerli *et al.* 2003), a comparison of observations and modelled results for 2001 was conducted, and in EMEP Status Report 1/2004 (Fagerli, 2004) we presented results for 2002 with an updated EMEP Unified model, version 2.0. This version differed slightly from the 2003 version, as described in EMEP Status Report 1/2004 (Fagerli, 2004), however the main conclusions on the model performance was the same. In 2005, we presented results for the year 2003 in EMEP Status Report 1/2005 (Fagerli, 2005) and in 2006 we presented results for 2004 in EMEP Status Report 1/2006 (Fagerli *et al.* 2006). It has been shown that the EMEP model performance is rather homogeneous over the years (Fagerli *et al.* 2003), but depend on geographical coverage and quality of the measurement data. The EMEP model has also been validated for nitrogen compounds in Simpson *et al.*, 2006, and for dry and wet deposition of sulphur, and wet depositions for nitrogen in Simpson *et al*., 2006b with measurements outside the EMEP network.

The version rv4.5 of the EMEP/MSC-W Eulerian model has been used for all nitrogen computations presented in this Chapter (EMEP, 2014).

In 2008, the Steering Body adopted an extension of the official EMEP domain to facilitate the inclusion of countries in Eastern Europe, Caucasus and Central Asia (EECCA) in the EMEP calculations (ref. ECE/EB.AIR/GE.1/2007/9, Item 3 of the provisional agenda of thirty-first session of the EMEP Steering Body, available from http://www.unece.org/env/lrtap/emep/emep31_docs.htm). Thus from 2008, the official 50 x 50 km^2 polar stereographic EMEP grid has been extended from 132 x 111 to 132 x 159 grid cells, following Stage 1 in ECE/EB.AIR/GE.1/2007/9. In geographical projection it leads to an extension eastward as well as northward. Both the old and new extended EMEP domains are presented in Figure 1.1.

The present extension of the EMEP modelling area has many advantages, but also recognized drawbacks. Main advantage is a possibility of taking into account much larger part of the Russian emissions in the extended model domain. The main drawback is that the current extended EMEP domain still only partly covers the Russian Federation. It is also recognized that results on air pollution in central Asian countries are highly dependent on sources outside the calculation domain. Countries in Central Asia are contiguous with other Asian countries like China, India, Pakistan and Iran that significantly affect pollution levels over the EECCA territories but are not included directly in the calculations. Consequently, the current EMEP modelling capacity for EECCA countries and the related grid domain is an interim solution until 2013. In 2014, a new EMEP official domain covering adequately transport of pollution to all 12 EECCA countries is expected to be adopted.

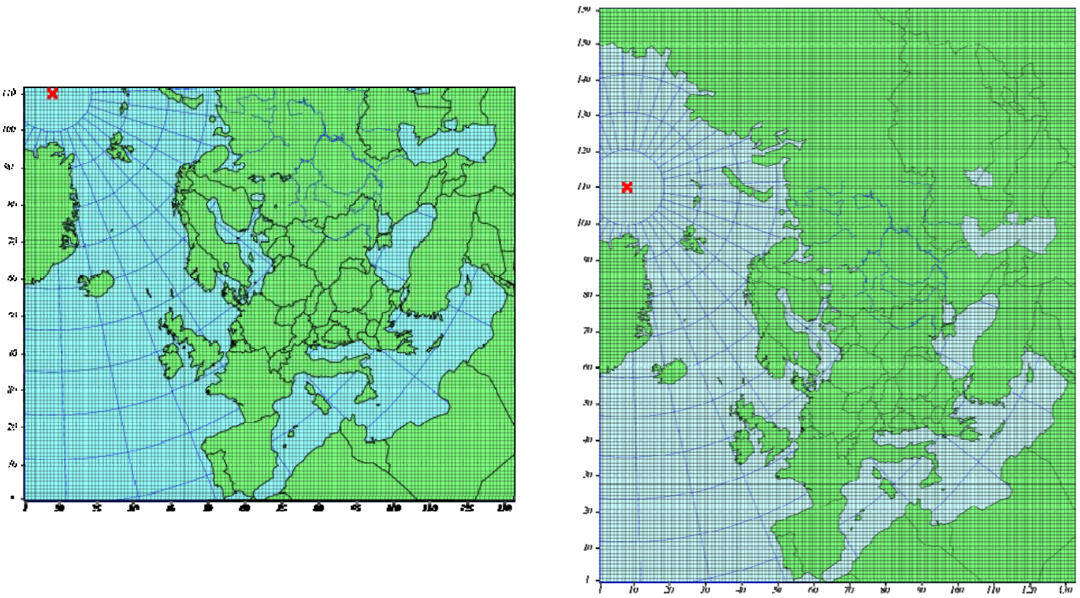


Figure 1.1. Comparison of old (used before 2007) official EMEP domain on the left side and new official EMEP domain on the right side. The new model domain was used for all computations for 2012 presented in this report.

Atmospheric input and source allocation budgets of heavy metals (cadmium, lead, and mercury) to the Baltic Sea were computed using the latest version of MSCE-HM model. MSCE-HM is the regional-scale model operating within the EMEP region. This is a three-dimensional Eulerian-type chemistry transport model driven by off-line meteorological data. The model considers HM emissions from anthropogenic and natural sources, transport in the atmosphere, chemical transformations (of mercury only) both in gaseous and aqueous phases, and deposition to the surface. The model domain is defined on polar stereographic projection and covers the standard EMEP region by a regular grid with 50x50 km spatial resolution at 60º latitude. For national scale applications finer resolution is applied (e.g. 5x5, 10x10 km). Vertical structure of the model is formulated in the sigma-pressure coordinate system, particularly, 15 irregular sigma-layers are used in the model covering the whole troposphere. Detailed description of the model is available in (Travnikov and Ilyin, 2005) and in the Internet on EMEP web page http://www.emep.int under the link to information on Heavy Metals.

It is assumed in the model that such HMs as lead and cadmium and their compounds are transported in the atmosphere in composition of aerosol particles. It is believed that possible chemical transformations of lead and cadmium do not change properties of carrying particles with regard to removal processes. On the contrary, for mercury the model considers its transformations in the atmosphere including transitions between the gaseous, aqueous and solid phases, and chemical reactions in the gaseous and aqueous environment. Model description of removal processes includes dry deposition and wet scavenging. The dry deposition scheme is based on the resistance analogy and allows taking into account deposition to different land cover types. The model distinguishes in-cloud and sub-cloud wet scavenging of particulate species and highly soluble reactive gaseous mercury. Wind re-suspension of particle-bound lead and cadmium from soil and seawater is an important process which affects essentially ambient pollution levels, particularly, in areas with low direct anthropogenic emissions. The model includes parameterization of HM re-suspension with dust aerosol particles from soil and generation of sea-salt and wind suspension of HMs from sea surface.

Evaluation of PCDD/F atmospheric input to the Baltic Sea was carried out using the latest version of MSCE-POP model. Similar to MSCE-HM model the MSCE-POP model is a three-dimensional Eulerian multimedia POP transport model operating within the geographical scope of EMEP region with spatial resolution 50 km at 60^o latitude. Both models share the same description of atmospheric transport and structure of the atmospheric compartment. The MSCE-POP model considers the following environmental compartments: air, soil, sea, vegetation and forest litter fall. The following basic processes are included in the model to describe POP fate: emission, advective transport, turbulent diffusion, dry and wet deposition, gas/particle partitioning, degradation, and gaseous exchange between the atmosphere and the underlying surface (soil, seawater, vegetation). Detailed description of MSCE-POP model is given in EMEP report (Gusev et al., 2005) and in the Internet on EMEP web page http://www.emep.int under the link to information on Persistent Organic Pollutants.

The formulation of MSCE-HM and MSCE-POP models and their performance were thoroughly evaluated within the framework of activity of EMEP/TFMM on the EMEP Models Review (ECE/EB.AIR/GE.1/2006/4). One of the main conclusions of the TFMM Workshop held in Moscow in 2005 was that MSCE-HM and MSCE-POP models represent the state of the science and fit for the purpose of evaluating the contribution of long-range transport to the environmental impacts caused by HMs and POPs.

Along with the regional-scale models there is ongoing development of the global multiscale modelling approach for HMs and POPs at the MSC-E. The Global EMEP Multi-media Modelling System (GLEMOS) is being elaborated to evaluate HM and POP pollution at different scales (global, regional, and local) and substitute MSCE-HM and MSCE-POP in future.

In 2013, an important change has been made by HELCOM concerning sub-basins of the Baltic Sea. Until 2012, all depositions, as well as, source allocation budgets have been calculated for the six sub-basins of the Baltic Sea. The names and acronyms of these old sub-basins are given below:

1. Gulf of Bothnia (GUB)
2. Gulf of Finland (GUF)
3. Gulf of Riga (GUR)

4. Baltic Proper (BAP)
5. Belt Sea (BES)
6. The Kattegat (KAT)

New, nine sub-basins of the Baltic Sea are listed in Table 1.2 in alphabetical order including their abbreviations and surface area in km^2.

Table 1.2. The "new" sub-basins of the Baltic Sea used for computing atmospheric nitrogen deposition from 2012 listed in alphabetical order. The abbreviations and areas of sub-basins of the Baltic Sea and the entire Baltic Sea basin are also shown.

Sub-basin	**Abbreviation**	**Area in km^2**
Archipelago Sea	ARC	13405
Baltic Proper	BAP	209258
Bothnian Bay	BOB	36249
Bothnian Sea	BOS	65397
Gulf of Finland	GUF	29998
Gulf of Riga	GUR	18646
Kattegat	KAT	23659
The Sound	SOU	2328
Western Baltic	WEB	18647
Baltic Sea basin	**BAS**	**417587**

The area of the entire Baltic Sea, calculated as a sum of sub-basins 417 587 km^2. The locations of the new basins are presented in Fig. 1.2. These new sub-basins have been used for all computations presented and discussed in the present report.

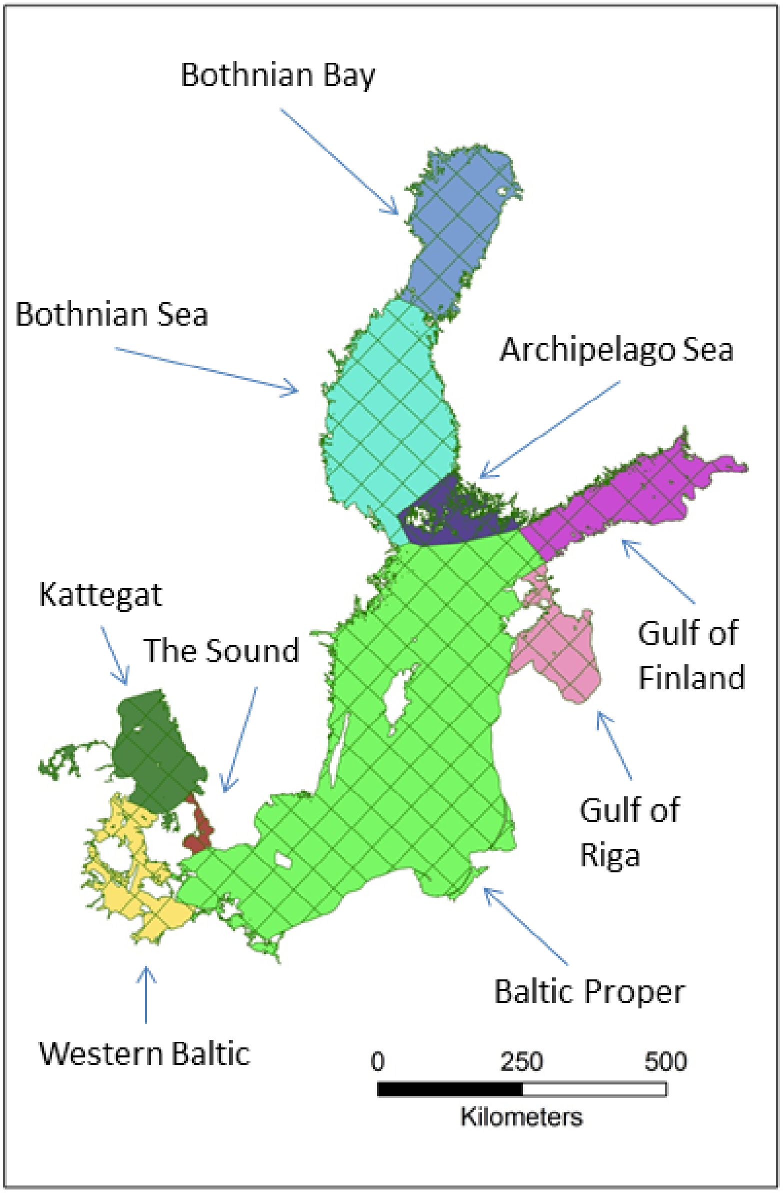


Fig 1.2. Locations of the new sub-basins of the Baltic Sea listed in Table 1.2 and used for all nitrogen deposition calculations presented in this report. The original figure with the sub-basins was provided by the Baltic Nest Institute (BNI).

In the results presented in the present report country, source and receptor names are often abbreviated. The list of these abbreviations is given below together with the EMEP identification number.

CODE	**EMEP ID**	**NAME**
AL	1	Albania
AT	2	Austria
BE	3	Belgium
BG	4	Bulgaria
FCS	5	Former Czechoslovakia
DK	6	Denmark
FI	7	Finland
FR	8	France
FGD	9	Former German Democratic Republic
FFR	10	Former Federal Republic of Germany
GR	11	Greece
HU	12	Hungary
IS	13	Iceland
IE	14	Ireland
IT	15	Italy
LU	16	Luxembourg
NL	17	Netherlands
NO	18	Norway
PL	19	Poland
PT	20	Portugal
RO	21	Romania
ES	22	Spain
SE	23	Sweden
CH	24	Switzerland
TR	25	Turkey
FSU	26	Former USSR
GB	27	United Kingdom
VOL	28	Volcanic emissions
REM	29	Remaining land Areas
BAS	30	Baltic Sea
NOS	31	North Sea
ATL	32	Remaining North-East Atlantic Ocean
MED	33	Mediterranean Sea
BLS	34	Black Sea
NAT	35	Natural marine emissions
RUO	36	Kola & Karelia
RUP	37	St.Petersburg & Novgorod-Pskov
RUA	38	Kaliningrad
BY	39	Belarus

UA	40	Ukraine
MD	41	Republic of Moldova
RUR	42	Rest of the Russian Federation
EE	43	Estonia
LV	44	Latvia
LT	45	Lithuania
CZ	46	Czech Republic
SK	47	Slovakia
SI	48	Slovenia
HR	49	Croatia
BA	50	Bosnia and Herzegovina
CS	51	Serbia and Montenegro
MK	52	The former Yugoslav Republic of Macedonia
KZ	53	Kazakhstan in the former official EMEP domain
GE	54	Georgia
CY	55	Cyprus
AM	56	Armenia
MT	57	Malta
ASI	58	Remaining Asian areas
LI	59	Liechtenstein
DE	60	Germany
RU	61	Russian Federation in the former official EMEP domain
MC	62	Monaco
NOA	63	North Africa
EU	64	European Community
US	65	United States
CA	66	Canada
BIC	67	Boundary and Initial Conditions
KG	68	Kyrgyzstan
AZ	69	Azerbaijan
ATX	70	EMEP-external Remaining North-East Atlantic Ocean
RUX	71	EMEP-external part of Russian Federation
RS	72	Serbia
ME	73	Montenegro
RFE	74	Rest of Russian Federation in the extended EMEP domain
KZE	75	Rest of Kazakhstan in the extended EMEP domain
UZO	76	Uzbekistan in the former official EMEP domain
TMO	77	Turkmenistan in the former official EMEP domain
UZE	78	Rest of Uzbekistan in the extended EMEP domain
TME	79	Rest of Turkmenistan in the extended EMEP domain
CAS	80	Caspian Sea

TJ	81	Tajikistan
ARO	82	Aral Lake in the former official EMEP domain
ARE	83	Rest of Aral Lake in the extended EMEP domain
ASM	84	Modified Remaining Asian Areas in the former official EMEP domain
ASE	85	Remaining Asian Areas in the extended EMEP domain
AOE	86	Arctic Ocean in the extended EMEP domain
KZT	92	Kazakhstan
RUE	93	Russian Federation in the extended EMEP domain (RU + RFE + RUX)
UZ	94	Uzbekistan
TM	95	Turkmenistan
AST	96	Asian areas in the extended EMEP domain (ASM + ASE + ARO + ARE + CAS)
FYU	99	Former Yugoslavia
BEF	301	Belgium (Flanders)
BA2	302	Baltic Sea EU Cargo o12m
BA3	303	Baltic Sea ROW Cargo o12m
BA4	304	Baltic Sea EU Cargo i12m
BA5	305	Baltic Sea ROW Cargo i12m
BA6	306	Baltic Sea EU Ferry o12m
BA7	307	Baltic Sea ROW Ferry o12m
BA8	308	Baltic Sea EU Ferry i12m
BA9	309	Baltic Sea ROW Ferry i12m
NO2	312	North Sea EU Cargo o12m
NO3	313	North Sea ROW Cargo o12m
NO4	314	North Sea EU Cargo i12m
NO5	315	North Sea ROW Cargo i12m
NO6	316	North Sea EU Ferry o12m
NO7	317	North Sea ROW Ferry o12m
NO8	318	North Sea EU Ferry i12m
NO9	319	North Sea ROW Ferry i12m
AT2	322	Remaining North-East Atlantic Ocean EU Cargo o12m
AT3	323	Remaining North-East Atlantic Ocean ROW Cargo o12m
AT4	324	Remaining North-East Atlantic Ocean EU Cargo i12m
AT5	325	Remaining North-East Atlantic Ocean ROW Cargo i12m
AT6	326	Remaining North-East Atlantic Ocean EU Ferry o12m
AT7	327	Remaining North-East Atlantic Ocean ROW Ferry o12m
AT8	328	Remaining North-East Atlantic Ocean EU Ferry i12m
AT9	329	Remaining North-East Atlantic Ocean ROW Ferry i12m
ME2,	332,	Mediterranean Sea EU Cargo o12m

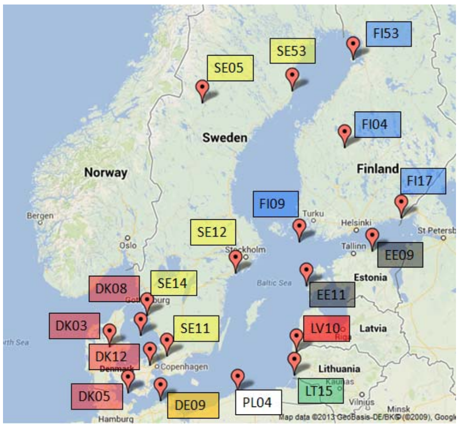
ME3	333	Mediterranean Sea ROW Cargo o12m
ME4	334	Mediterranean Sea EU Cargo i12m
ME5	335	Mediterranean Sea ROW Cargo i12m
ME6	336	Mediterranean Sea EU Ferry o12m
ME7	337	Mediterranean Sea ROW Ferry o12m
ME8	338	Mediterranean Sea EU Ferry i12m
ME9	339	Mediterranean Sea ROW Ferry i12m
BL2	342	Black Sea EU Cargo o12m
BL3	343	Black Sea ROW Cargo o12m
BL4	344	Black Sea EU Cargo i12m
BL5	345	Black Sea ROW Cargo i12m
BL6	346	Black Sea EU Ferry o12m
BL7	347	Black Sea ROW Ferry o12m
BL8	348	Black Sea EU Ferry i12m
BL9	349	Black Sea ROW Ferry i12m
GL	601	Greenland

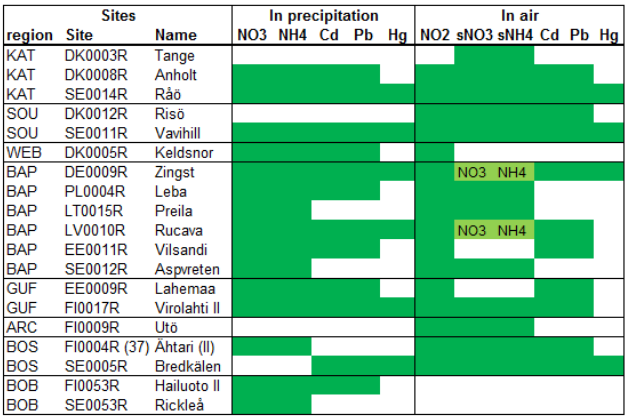
2. Observed Concentrations of Nitrogen, Cadmium, Lead and Mercury at HELCOM Stations in 2012

2.1 HELCOM measurement stations

Eight countries have submitted data from all together nineteen HELCOM stations for 2012 (Fig. 2.1), which is the same number as in 2011. The stations are distributed in eight of the nine sub-basins (Fig. 2.1) as following: Three in Kattegat (KAT), two in The Sound (SOU), one in Western Baltic (WEB), six in the Baltic proper (BAP), two in Gulf of Finland (GUF), one in Archipelago Sea (ARC), two in Bothnian Sea (BOS) and two in Bothnian Bay (BOB). There is one station from Germany, Lithuania, Latvia, Poland; two stations from Estonia; four in Denmark and Finland; and five stations from Sweden. Råö and Vavihill in Sweden are the only ones with data for all the components in air and precipitation for 2012. In addition, the German site Zingst almost fulfil the requirements, but lack measurements of ammonia and nitric acid in air.

Fig 2.1. HELCOM sites with measurements of nitrogen, lead, cadmium and mercury in 2012





Sites			In precipitation					In air					
region	Site	Name	NO3	NH4	Cd	Pb	Hg	NO2	sNO3	sNH4	Cd	Pb	Hg
KAT	DK0003R	Tange							🟩	🟩			
KAT	DK0008R	Anholt	🟩	🟩	🟩	🟩		🟩	🟩	🟩	🟩	🟩	
KAT	SE0014R	Råö	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
SOU	DK0012R	Risö						🟩	🟩	🟩	🟩	🟩	
SOU	SE0011R	Vavihill	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
WEB	DK0005R	Keldsnor	🟩	🟩	🟩	🟩		🟩					
BAP	DE0009R	Zingst	🟩	🟩	🟩	🟩	🟩	🟩	NO3	NH4	🟩	🟩	🟩
BAP	PL0004R	Leba	🟩	🟩	🟩	🟩		🟩	🟩	🟩			
BAP	LT0015R	Preila	🟩	🟩				🟩	🟩	🟩			
BAP	LV0010R	Rucava	🟩	🟩	🟩	🟩	🟩	🟩	NO3	NH4	🟩	🟩	
BAP	EE0011R	Vilsandi	🟩	🟩	🟩	🟩		🟩			🟩	🟩	
BAP	SE0012R	Aspvreten	🟩	🟩				🟩	🟩	🟩			
GUF	EE0009R	Lahemaa	🟩	🟩	🟩	🟩		🟩			🟩	🟩	
GUF	FI0017R	Virolahti II	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	
ARC	FI0009R	Utö						🟩	🟩	🟩			
BOS	FI0004R (37)	Ähtari (II)	🟩	🟩				🟩	🟩	🟩	🟩	🟩	
BOS	SE0005R	Bredkälen			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
BOB	FI0053R	Hailuoto II	🟩	🟩	🟩	🟩							
BOB	SE0053R	Rickleå	🟩	🟩									

In this section, we provide a broad view of the patterns and levels evident in monitoring data from 2012. Where possible regional average values are provided for the principal regions within the Baltic Sea. For actual monthly values on a component-by-component basis, the reader is referred to Appendix A. A description of sampling and analytical methods is given in Appendix B. Further statistical details are also found in the EMEP reports for 2012 data (Hjellbrekke, 2014; Aas and Nizzetto, 2014), and all the data are available from the web database at http://ebas.nilu.no/. The HELCOM laboratories have participated in the EMEP and laboratory intercomparison and the laboratories generally have a satisfactory quality.

2.2 Nitrogen concentrations in air

Altogether thirteen stations have delivered data for total reduced nitrogen (NH_3+NH_4^+) and total nitrate (HNO_3+NO_3^-), and thirteen for nitrogen dioxide (NO_2). Annual averages of the different nitrogen species are presented in Figure 2.2. The lowest concentrations for all the three nitrogen species were reported at the northernmost Swedish site (SE05) in 2012: The concentrations were 0.14, 0.03 and 0.12 µg N/m^3 for respectively NH_3+NH_4^+, HNO_3+NO_3^- and NO_2 at this site. Highest concentrations of nitrogen in air were found in Danish site Risö (DK12) with annual concentration means of 2.8 µgN/m^3 for NO_2), 1.86 µgN/m^3 for sum ammonium and 0.86 µgN/m^3 for sum nitrate. Data for particulate nitrate and ammonium from the German site (DE09) is not included in the figures below, because it is not comparable with the sums shown there, though the data are given in the annex. Details for monthly and annual concentrations for all the sites are found in the annex, table A.1.

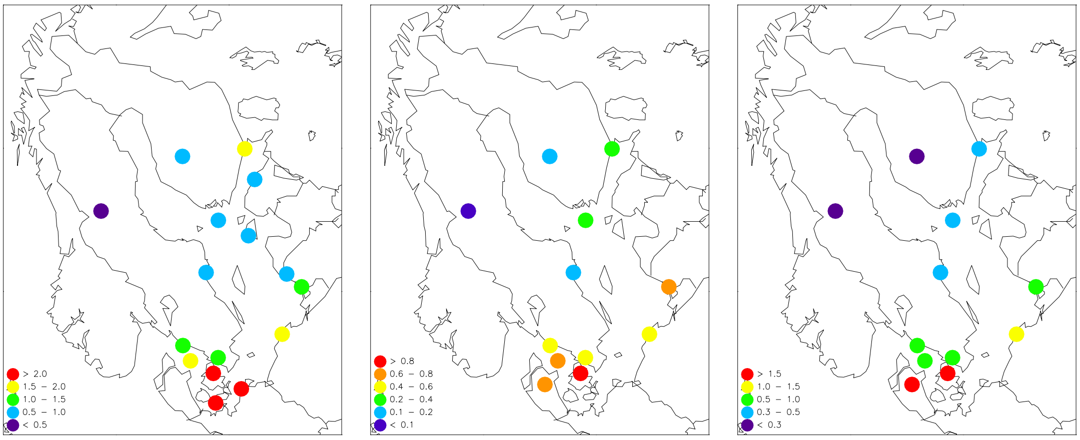


Figure 2.2. Concentrations of left: NO_2 in air, middle: total nitrate (HNO_3+NO_3^-) and right: total reduced nitrogen (NH_3+NH_4^+) in 2012 Unit: µg N/m^3.

There is a tendency of increasing concentrations from north to south and towards west. This concentration gradients reflect the varied influence of traffic (ship as well as cars) and agricultural activities. A similar gradient can also be noticed in Figure 2.3-2.5 displaying the station averages of NH_3+NH_4^+, HNO_3+NO_3^- and NO_2 observations across six sub-basins

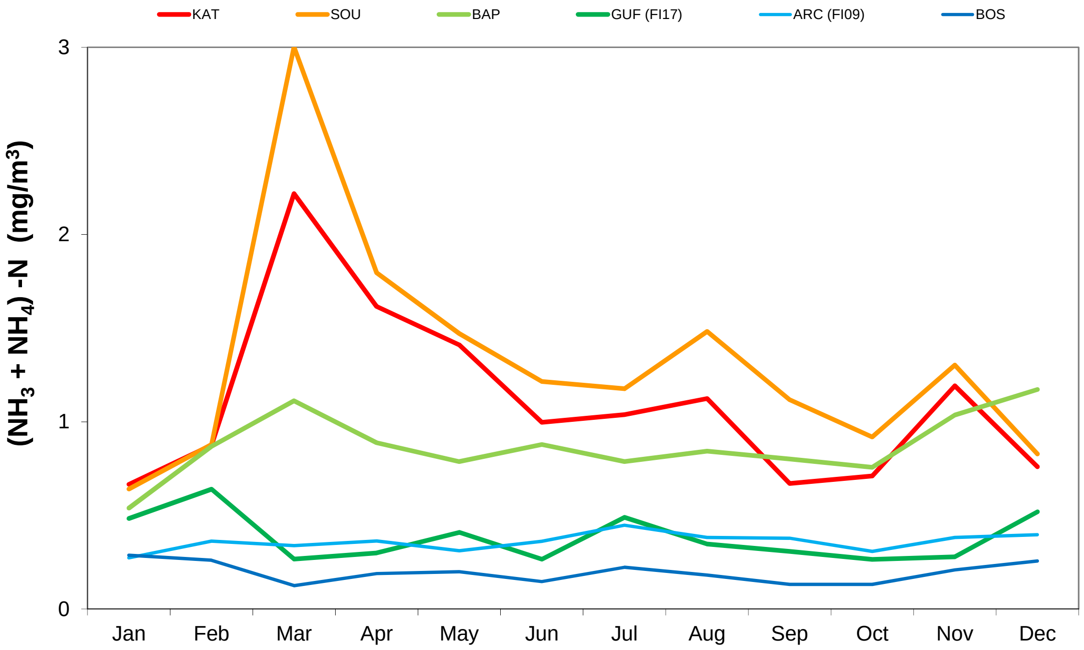


Figure 2.3. Monthly total reduced nitrogen (NH_3+NH_4) concentrations in the air in 2012

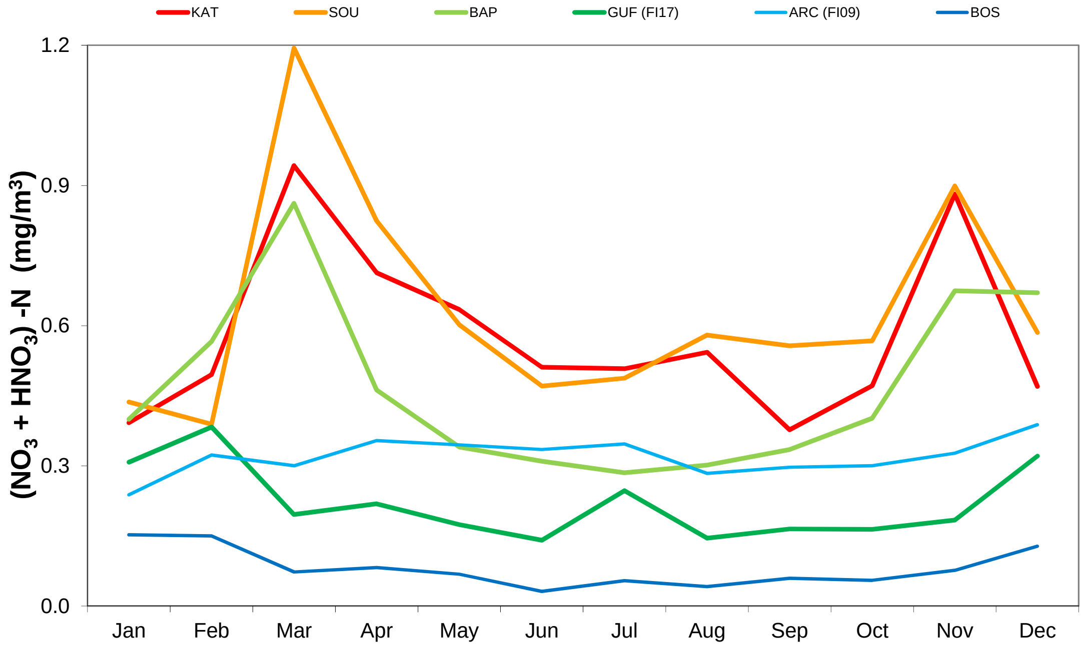


Figure 2.4. Monthly total oxidized nitrate (HNO_3+NO_3^-) concentrations in the air in 2012.

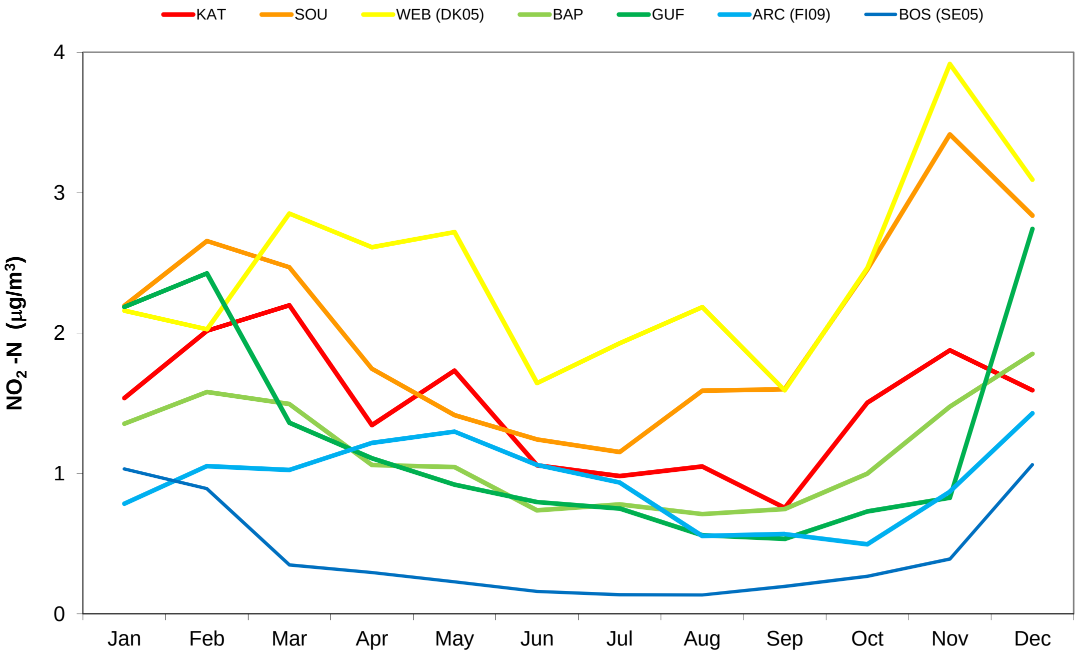


Figure 2.5. Monthly NO_2 concentrations in the air in 2012

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Observations of the total reduced nitrogen (NH_3+NH_4^+), show a seasonal pattern similar for most the sub-basins with highest concentrations during April when the fertilizing is most important. Agricultural activities (natural fertilizer) are the main source for NH_3+NH_4^+. But also high concentrations are seen in late autumn, maybe due to some fall fertilization, but more likely due to longer resistance time of NH_4NO_3 in the atmosphere (see below).

Total nitrate (HNO_3+NO_3^-) concentration show elevated levels in the spring late autumn; and generally somewhat higher concentrations in winter than summer. NO_2 is reacting photo-chemically and the reaction product is total nitrate. This reaction is mostly dominating during spring and summer. However, total nitrate is dominated by particulate nitrate in the cold season, which has a higher residence time in the atmosphere than nitric acid. In the summer, more of total nitrate consists of nitric acid, which is dry deposited very fast. This effect of fast removal of sum nitrate seems to be relatively more important than higher production in summer, causing summer minima. Concentrations of NO_2 also show, not unexpectedly, a temporal pattern with a winter maxima/summer minima. During winter the atmospheric residence time is longer due to low photo-chemically activity and reduced vertical mixing.

2.3 Nitrogen in precipitation

Altogether fifteen stations have delivered data for ammonium and nitrate in precipitation. Stations from eight sub-basins have delivered data for ammonium and nitrate in precipitation. Annual averages of the two nitrogen species are presented in Figure 2.6.

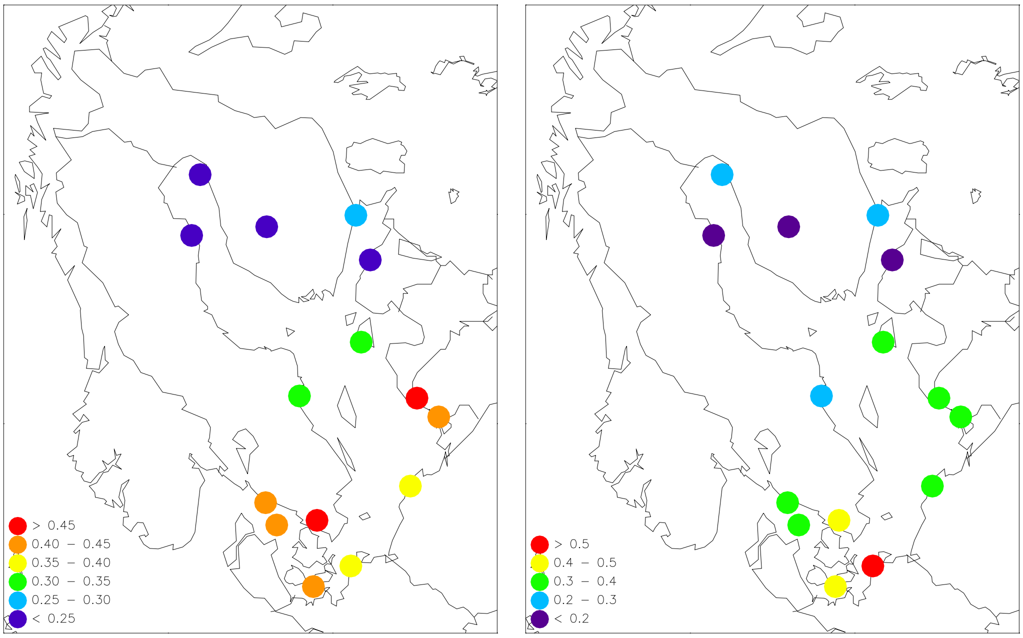


Figure 2.6. Concentrations of left: nitrate (NO_3^-), and right: ammonium (NH_4^+) in precipitation in 2012. Units: mg N/l.

The yearly mean concentrations in precipitation have been calculated from daily or weekly reported values as precipitation-weighted averages. A south-north gradient similar to air can also be seen for nitrogen in precipitation with higher concentrations in the south, and also a west-east gradient is seen, Lowest concentrations for both nitrogen species were seen at FI04 and SE53 with (0.11 and 0.16 mg N/L) and (0.20 – 0.21 mg N/L) for nitrate and ammonium respectively. The highest levels are found at DE09 and DK05,SE11 for ammonium (0.61 and 0.49 mg N/L) and SE11 and LV10 for nitrate (0.50 and 0.48 mg N/L). Figure 2.7 displays the station average monthly depositions of oxidized and reduced nitrogen across the regions given.

There is no clear seasonal patterns for the nitrogen wet deposition as for airborne components. The spatial pattern persists, however, with clearly decreasing depositions with progression northwards. For example, the northern regions typically receive half the deposition of reduced nitrogen supplied to southern areas.

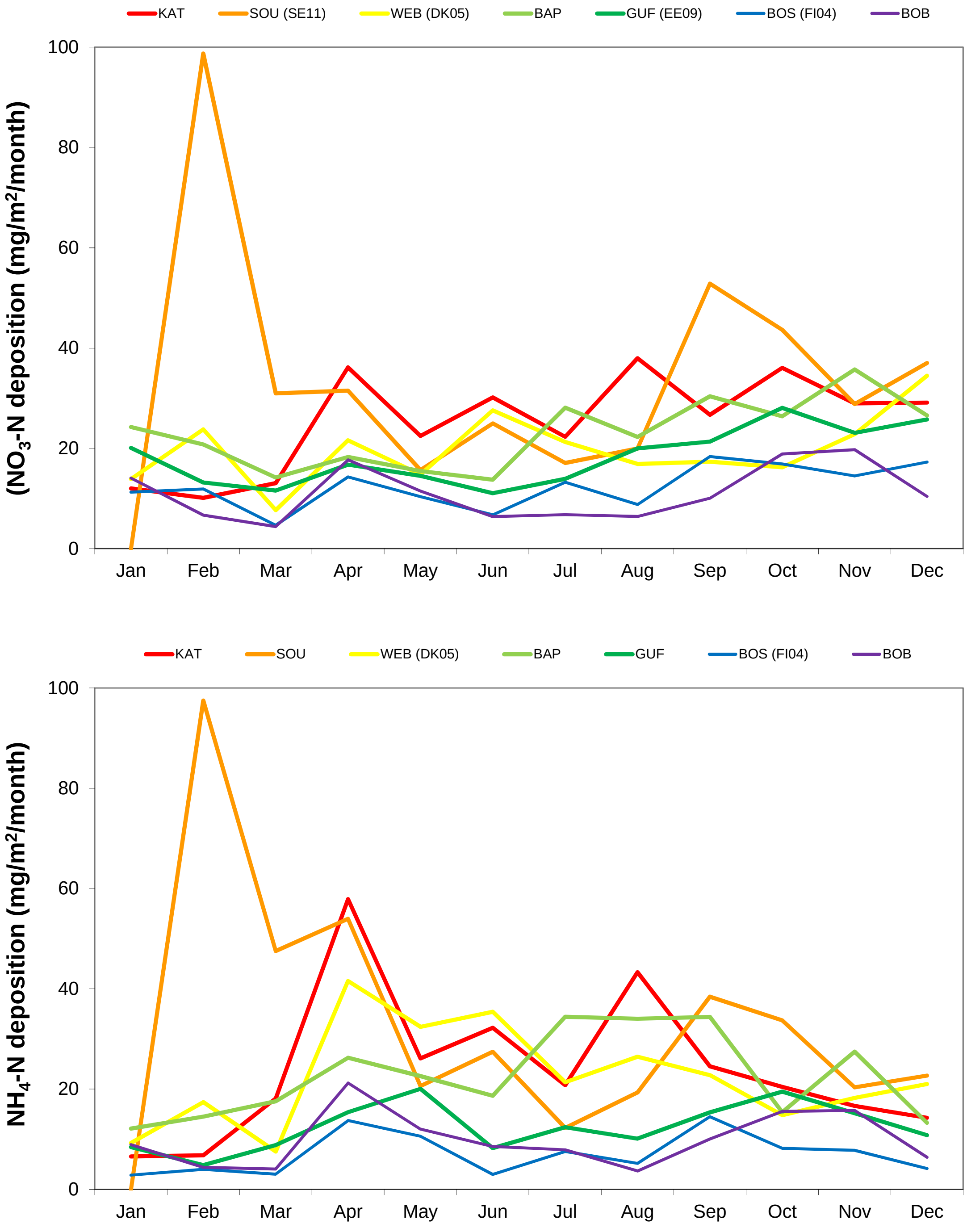


Figure 2.7. Monthly nitrogen depositions in 2012 averaged for the sub-basins. Top: nitrate (NO_3^-), and bottom: reduced nitrogen (NH_4^+).

2.4 Heavy metals in the air

Altogether twelve stations have delivered heavy metal data. Only four sites have reported data for elemental Hg in air, and three of these sites were Swedish. Annual averages of Cd and Pb are presented in Figure 2.8. The lowest concentrations of Cd is seen at the Swedish site SE05 (0.009 ng/m^3) while the highest concentration was seen at FI17 with 0.11 ng/m^3. For lead, the highest concentrations were observed at DE09 and FI17 with 3.3 ng/m^3; while the lowest level was at SE05 with 0.28 ng/m^3. At the Latvian site LV10 there were only measurements seven of the months in 2012, so these data are not included in the maps below. However the pollution level are one of the the highest in this regions with 0.14 and 4.4 ng/m^3 respectively for cadmium and lead for the seven month with measurements. For elemental mercury the concentrations ranged from 1.25 (SE05) to 1.66 (DE09).

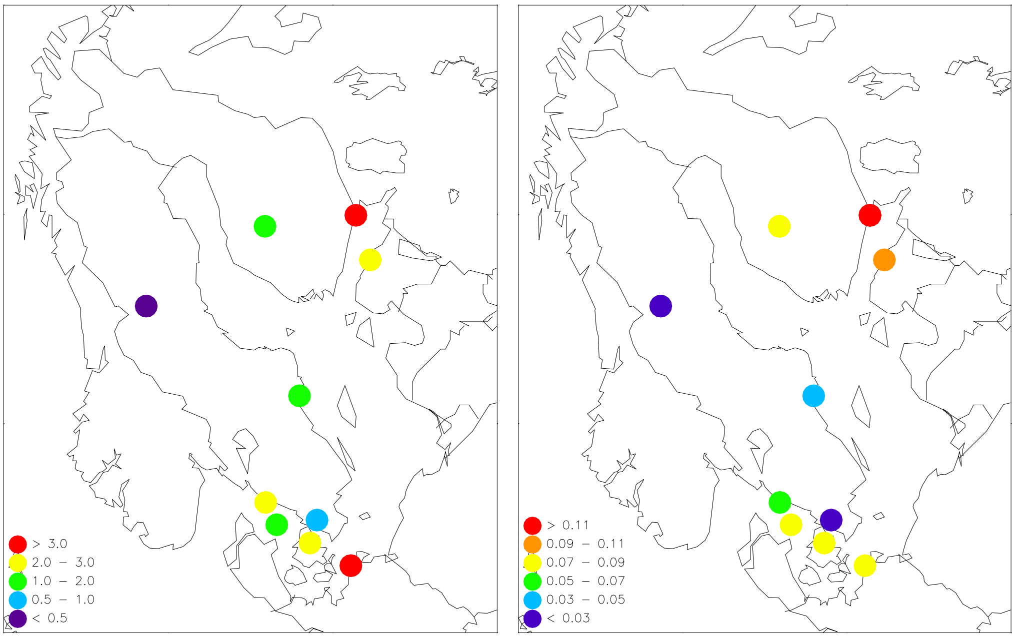
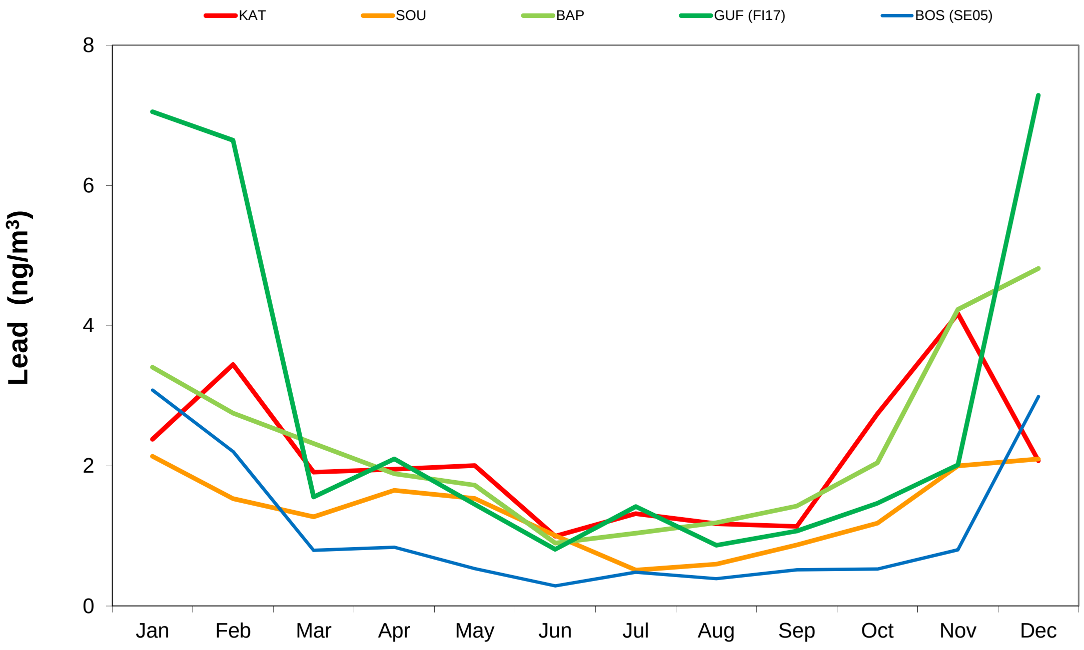


Figure 2.8. Concentrations of left: lead (Pb) and right: cadmium (Cd) in aerosol in air in 2012. Units: ng/m^3.

There are insufficient stations to reasonably represent regional patterns; hence, the station data itself is presented here for some of the sites (Fig. 2.9). From this, it is to be observed that the temporal patterns for Cd and Pb show a winter maximum. This is probably due longer atmospheric residence time in winter and reduced vertical mixing.



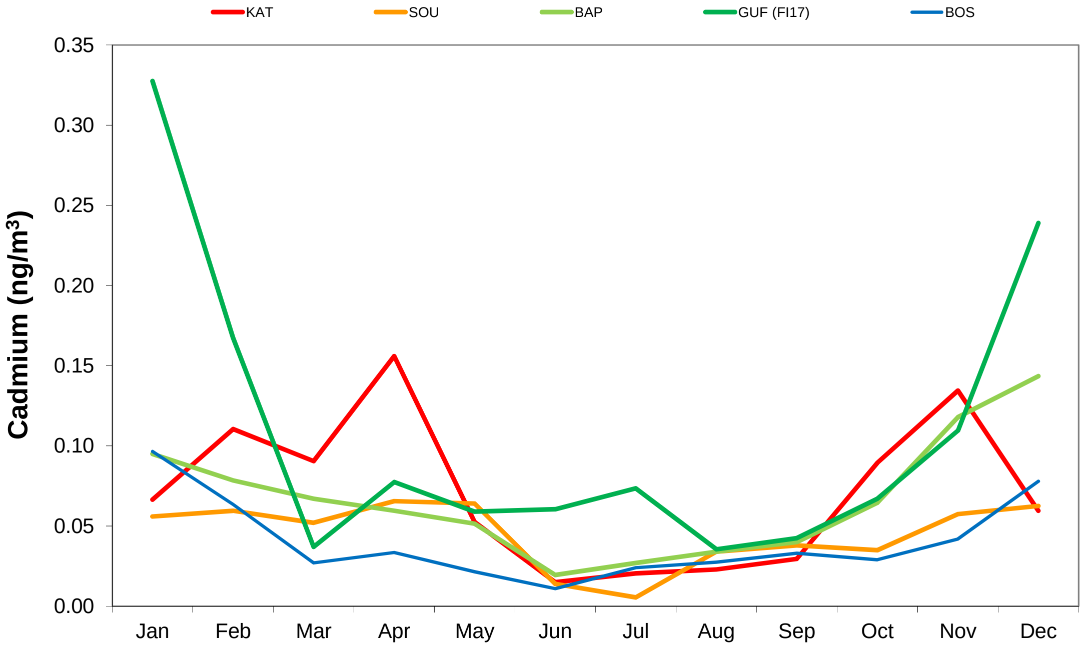


Figure 2.9. Monthly concentrations in air in 2012 averaged for the sub-basins: Top: cadmium, bottom: lead.

Hg concentrations at the four sites also show a weak winter maxima. September-October seems to be relatively low for several of the sites. Figure 2.10.

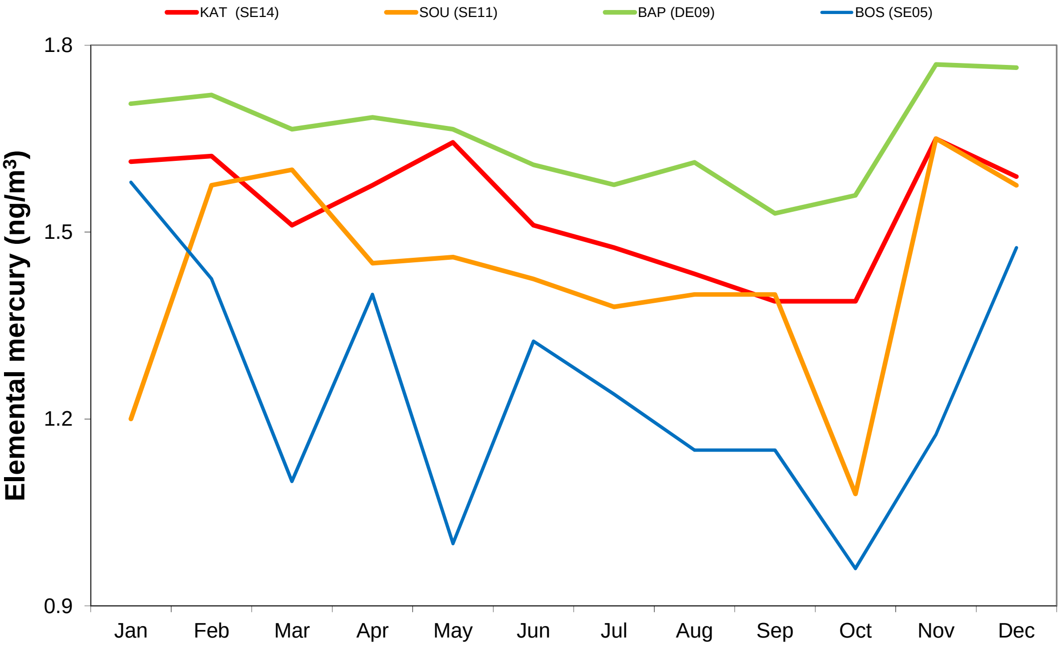


Figure 2.10. Monthly concentrations of Hg in air in 2012 for the four sites representing different the sub-basins.

2.5 Heavy metals in precipitation

Twelve stations have delivered data for Cd and Pb in precipitation, and six have delivered data for Hg in precipitation. Annual averages of Cd and Pb are presented in Figure 2.11. The yearly mean concentrations in precipitation have been calculated from weekly or monthly reported values as precipitation-weighted averages. The lowest concentrations were seen at the Estonian site, but here there are problems with the detection limit especially for Cd. Besides EE09, the lowest concentrations for both Cd and Pb in precipitation were reported for the Swedish site SE05 with 0.02 and 0.4 µg/l, respectively. For lead, similar low concentrations were seen at DE09, and for cadmium at LV10 and SE15. The highest concentration of Cd was measured at SE11 with 0.13 µg/l, while the highest level of lead was seen at DK05 with 5.2 µg/l. This is much higher concentration than the second highest level, which was observed at DK08 with 1.4 µg/l. For mercury in precipitation, the highest levels are seen in Latvia, but the detection limit for their analysis is very high and the highest level of the remaining sites is seen at SE11 and SE14 with 11 ng/L and the lowest at FI17 with 4 ng/L.

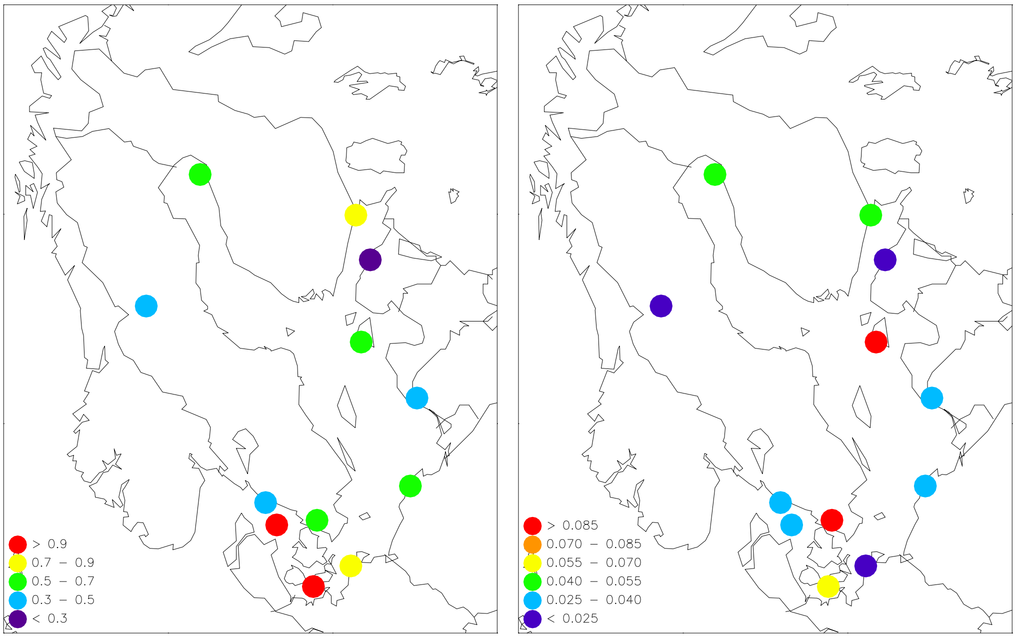


Figure 2.11. Concentrations of left: lead (Pb), right: cadmium (Cd) in precipitation in 2012. Units: µg/l.

2.6 Conclusions for Chapter 2

- Measurement data was reported from eighteen HELCOM stations in 2012, but few sites have a complete measurements program with measurements in both air and precipitation.

- There is a general tendency of decreasing concentrations from south to north for all relevant species; and for many species an east west gradient.

- Many of the components measured in air show a winter maxima due to longer atmospheric residence time.

- The seasonal patterns in precipitation are not as strong as for airborne components. This is due to the presence of the precipitation effect. Though the highest deposition of reduced nitrogen is seen in summer due to enhanced agricultural activity

3. Atmospheric Supply of Nitrogen to the Baltic Sea in 2012

All nitrogen depositions have been calculated for the year 2012 with the EMEP MSC-W model, based on the latest emission data submitted to CEIP (June 2013) and on comprehensive meteorological data from the European Centre for Medium-range Weather Forecasts (ECMWF). Nitrogen emission data, as well as the model results presented here have been approved by the 38th Session of the Steering Body of EMEP in Geneva in September 2014. The version rv4.5 of the EMEP/MSC-W Eulerian model has been used for all nitrogen computations presented in this Chapter.

All 2012 deposition calculations were performed for the EMEP extended domain introduced in 2007 which includes countries in Eastern Europe, Caucasus and Central Asia. Meteorological input data necessary for 2010 EMEP model calculations were provided by ECMWF Numerical Weather Prediction model (ECMWF-IFS). The meteorological fields used for 2012 are based on ECMWF-IFS model cycle 38r2, initialised by ECMWF Interim Reanalysis (ERA) data. The meteorological fields have been interpolated from longitude-latitude coordinates with a resolution of $0.1^{\circ}\times0.1^{\circ}$ to the polar stereographic 50×50 km^2 grid of EMEP.

The inter-annual variability of nitrogen depositions is mainly driven by the changes in emissions and changes in meteorological conditions. The NO_x emissions in the entire EMEP domain were decreased by 1.27% from 2011 to 2012 and the deposition of oxidized nitrogen, on average, decreased by 2.7% in the entire EMEP domain during the same period. In the case of reduced nitrogen, emissions of NH_3 increased only by 0.01% and the deposition of reduced nitrogen increased by about 0.6%. But in case of HELCOM countries, the total emissions of both NOx and NH3 were increased by 2.3 % and 2.8% respectively.

Emission input data for 2012 were prepared by the EMEP Centre on Emission Inventories and Projections (CEIP). The EMEP Parties reported emission inventory data using standard formats in accordance with the EMEP Reporting guidelines. For the EMEP models, reported by Parties sectoral (NFR09) emissions were aggregated into 10 SNAP sectors. To fill in the missing gaps in sectoral emissions, CEIP applied different methods including Expert estimates. For 2012 shipping data, interpolated NO_x, SO_x and PM emissions were used based on recent estimates by the International Institute for Applied System Analysis (IIASA), for the years 2010 and 2015.

Calculated annual deposition of total nitrogen to the Baltic Sea basin in 2012 was 230 kt, approximately 6% higher than in 2011. Deposition of oxidised nitrogen was 6% higher and deposition of reduced nitrogen was 10.5% higher in 2012 compared to 2011. Deposition of oxidized nitrogen accounted for 53% of total nitrogen deposition in 2012.

3.1 Nitrogen emissions

Table 3.1. Annual total 2012 emissions of nitrogen oxides and ammonia from the HELCOM Contracting Parties and ship traffic on the Baltic Sea. Sum of HELCOM emissions is also included. Units: kt N per year.

Emission source	**Pollutant**	
	NO_x	**NH_3**
Denmark	35	63
Estonia	10	9
Finland	45	30
Germany	387	449
Latvia	11	16
Lithuania	18	31
Poland	249	217
Russian Federation	901	1115
Sweden	40	42
HELCOM	**1695**	**1972**
Baltic Sea	105	

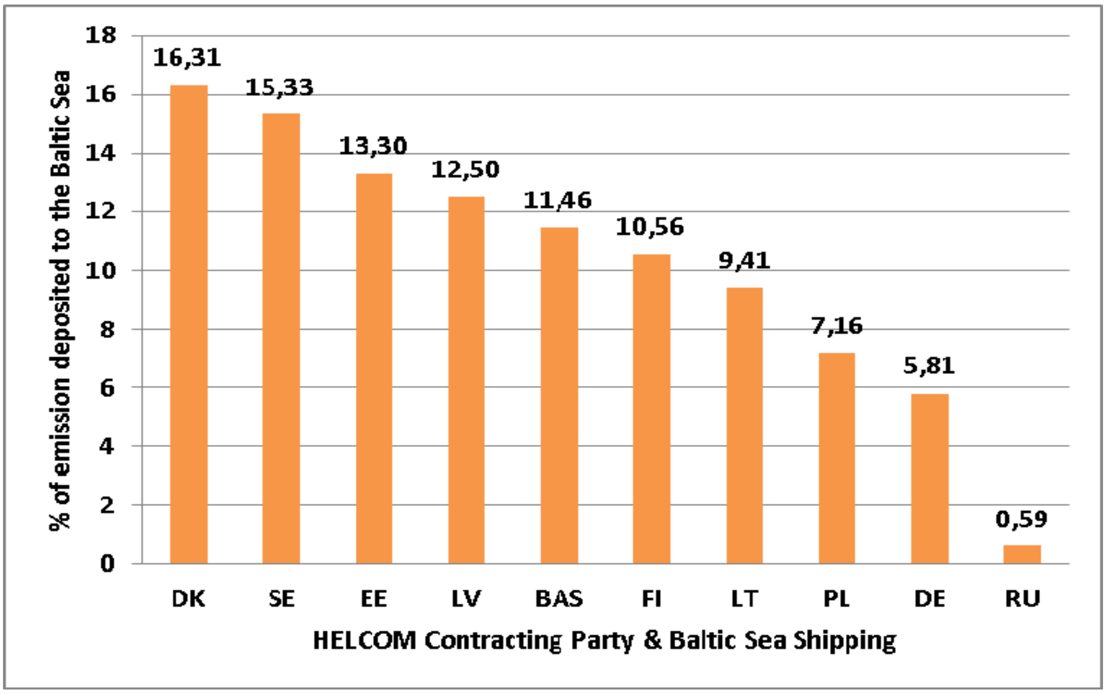


Figure 3.1. Percent of annual emissions of total (oxidized + reduced) nitrogen that is deposited on the Baltic Sea basin in 2012, for HELCOM Parties and international ship traffic on the Baltic Sea (BAS).

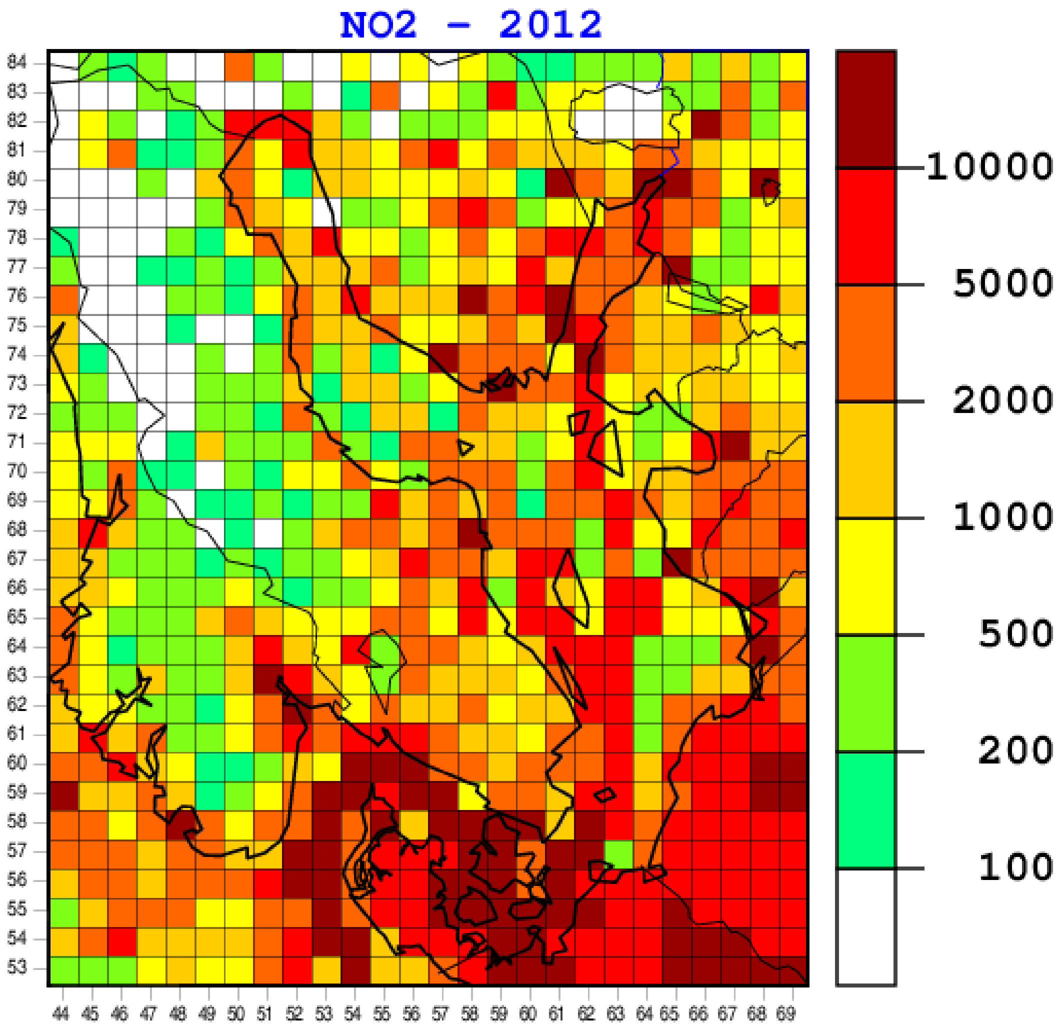


Figure 3.2. Map of annual emissions of oxidized nitrogen (including emissions from the ship traffic) in the Baltic Sea region in 2012. Units: Mg (tonnes) of NO_2 per year and per 50×50 km grid cell.

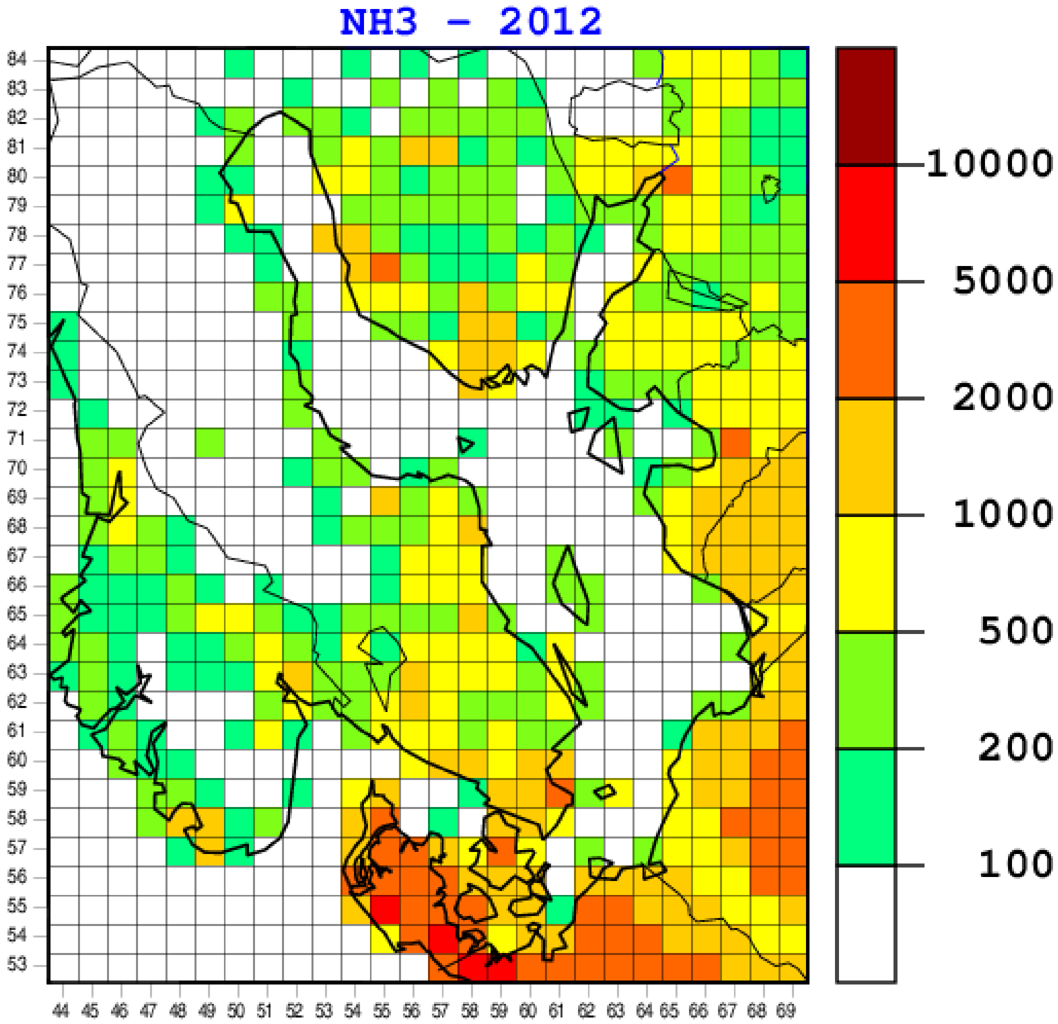


Figure 3.3. Map of annual emission of ammonia in the Baltic Sea region in 2012. Units: Mg of NH_3 per year and per 50×50 km grid cell.

Table 3.2. The list of 11 SNAP emission sectors as specified in the EMEP-CORINAIR Emission Inventory Guidebook.

Sector 1	Combustion in energy and transformation industry
Sector 2	Non-industrial combustion plants
Sector 3	Combustion in manufacturing industry
Sector 4	Production processes
Sector 5	Extraction and distribution of fossil fuels and geothermal energy
Sector 6	Solvent and other product use
Sector 7	Road transport
Sector 8	Other mobile sources and machinery (including ship traffic)
Sector 9	Waste treatment and disposal
Sector 10	Agriculture
Sector 11	Other sources and sinks

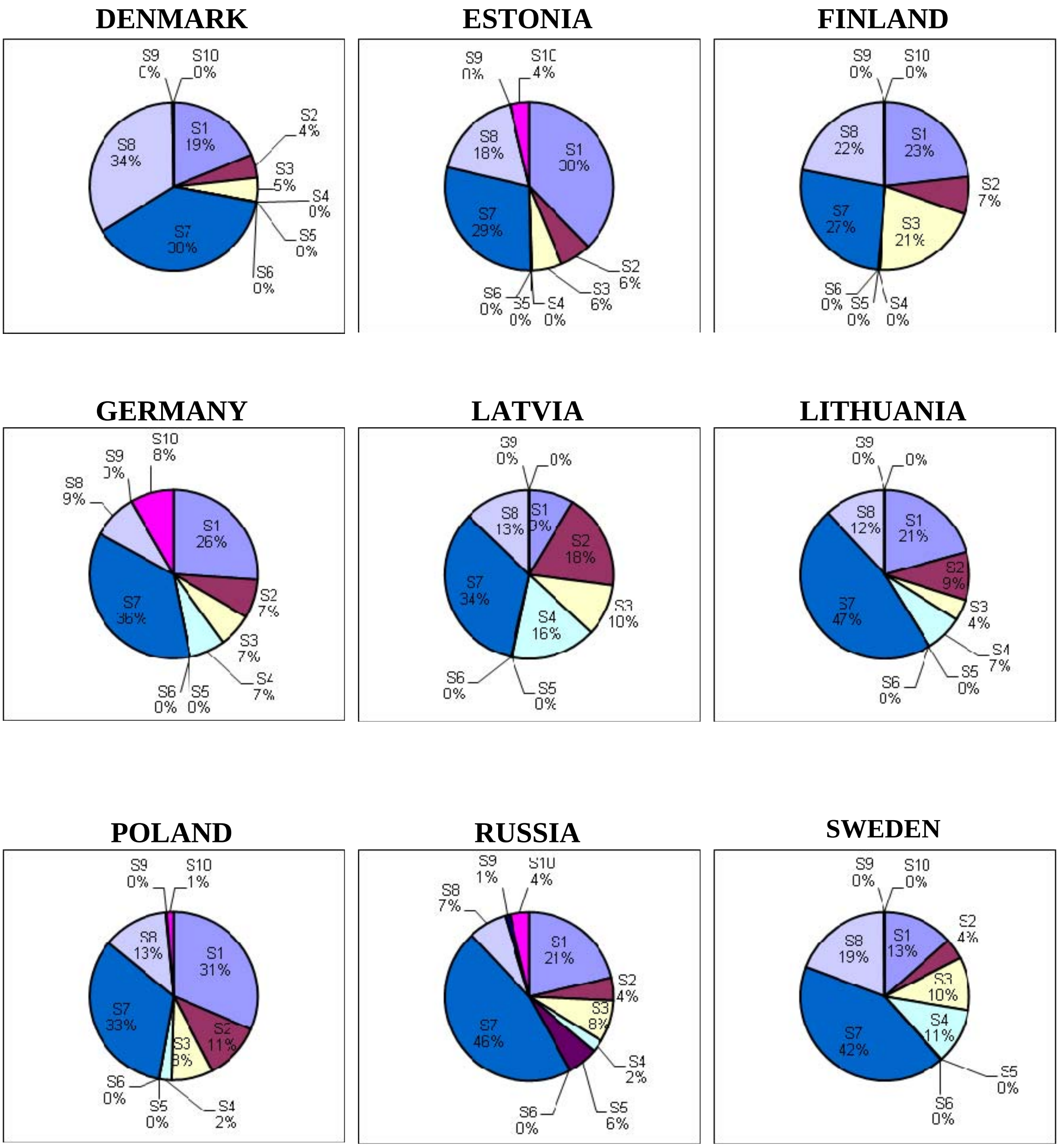


Figure 3.4. Annual 2012 nitrogen oxides emissions from the HELCOM Parties split into the SNAP sectors.

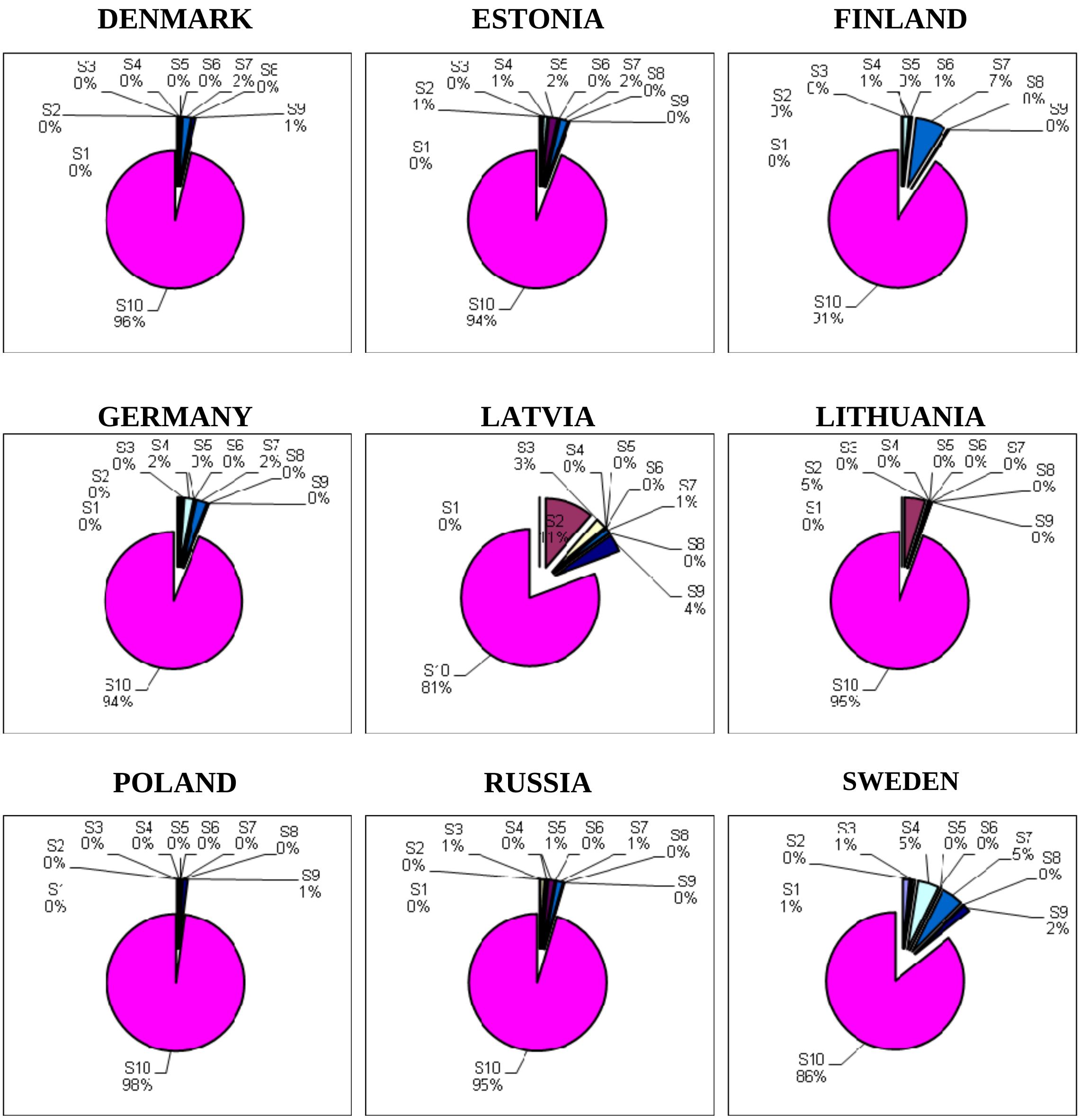


Figure 3.5. Annual 2012 ammonia emissions from the HELCOM Parties split into the SNAP sectors.

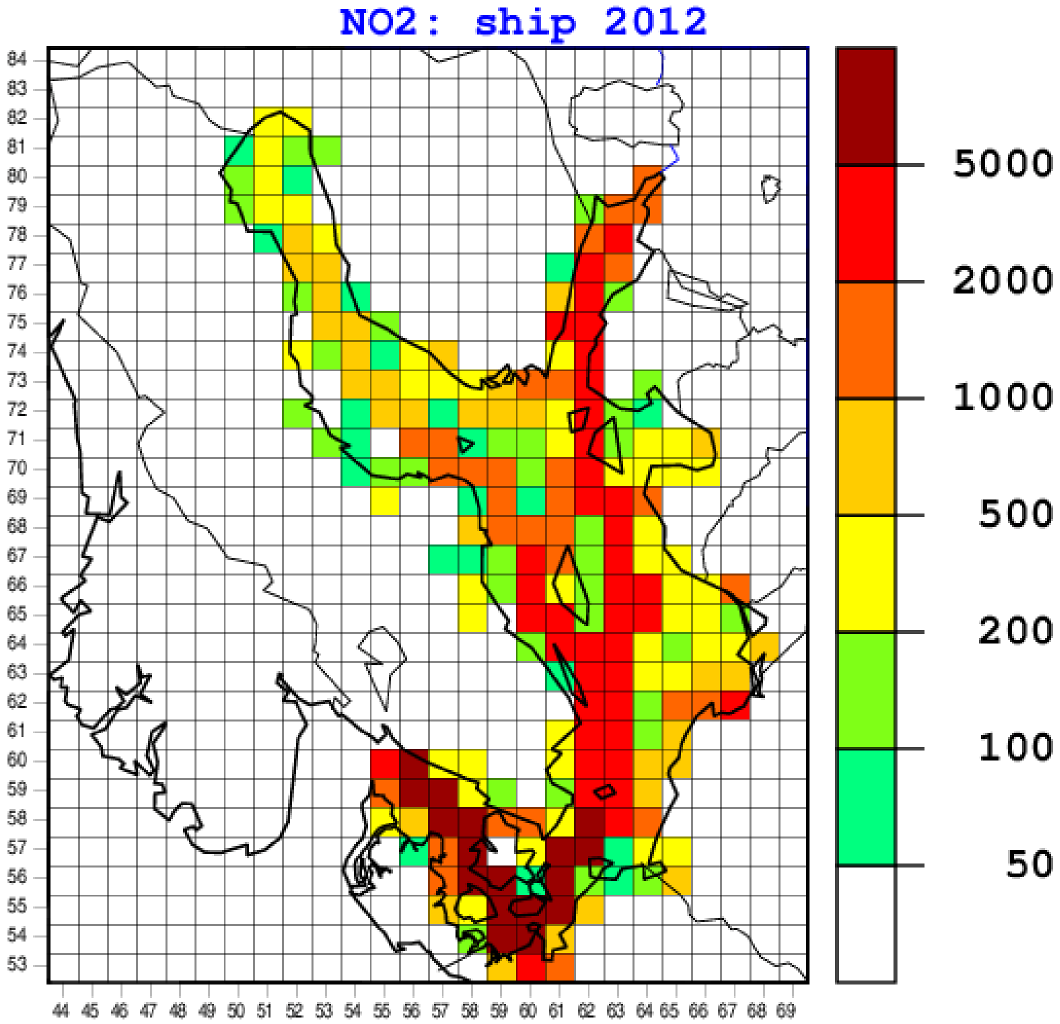


Figure 3.7 Map of annual emissions of nitrogen oxides from the international ship traffic on the Baltic Sea in 2012 used in the EMEP model calculations. Units: Mg of NO_2 per year and per 50×50 km grid cell. Emission input data for 2012 were prepared by the CEIP.

3.2 Annual deposition of nitrogen

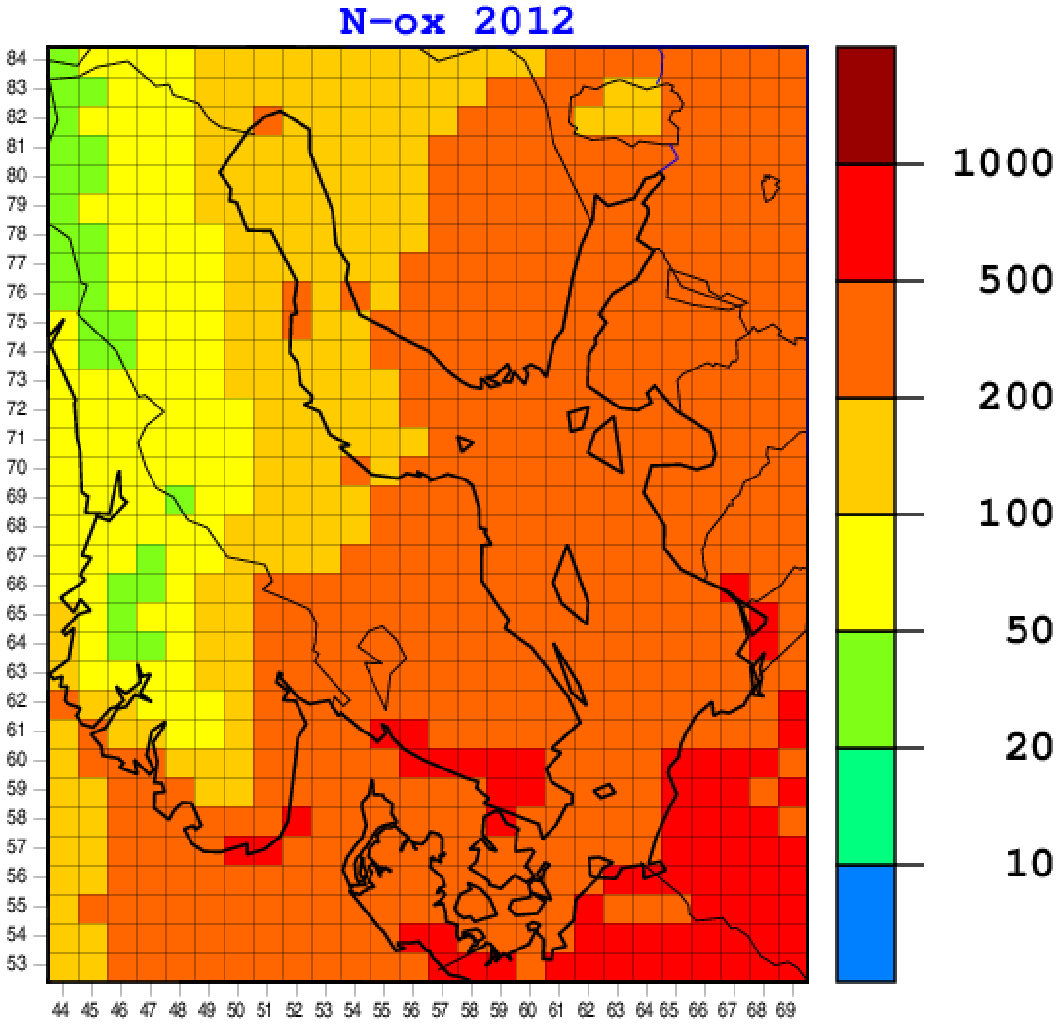


Figure 3.8. Map of annual deposition flux of oxidized nitrogen (dry + wet) in 2012. Units: mg N m^{-2} yr^{-1}.

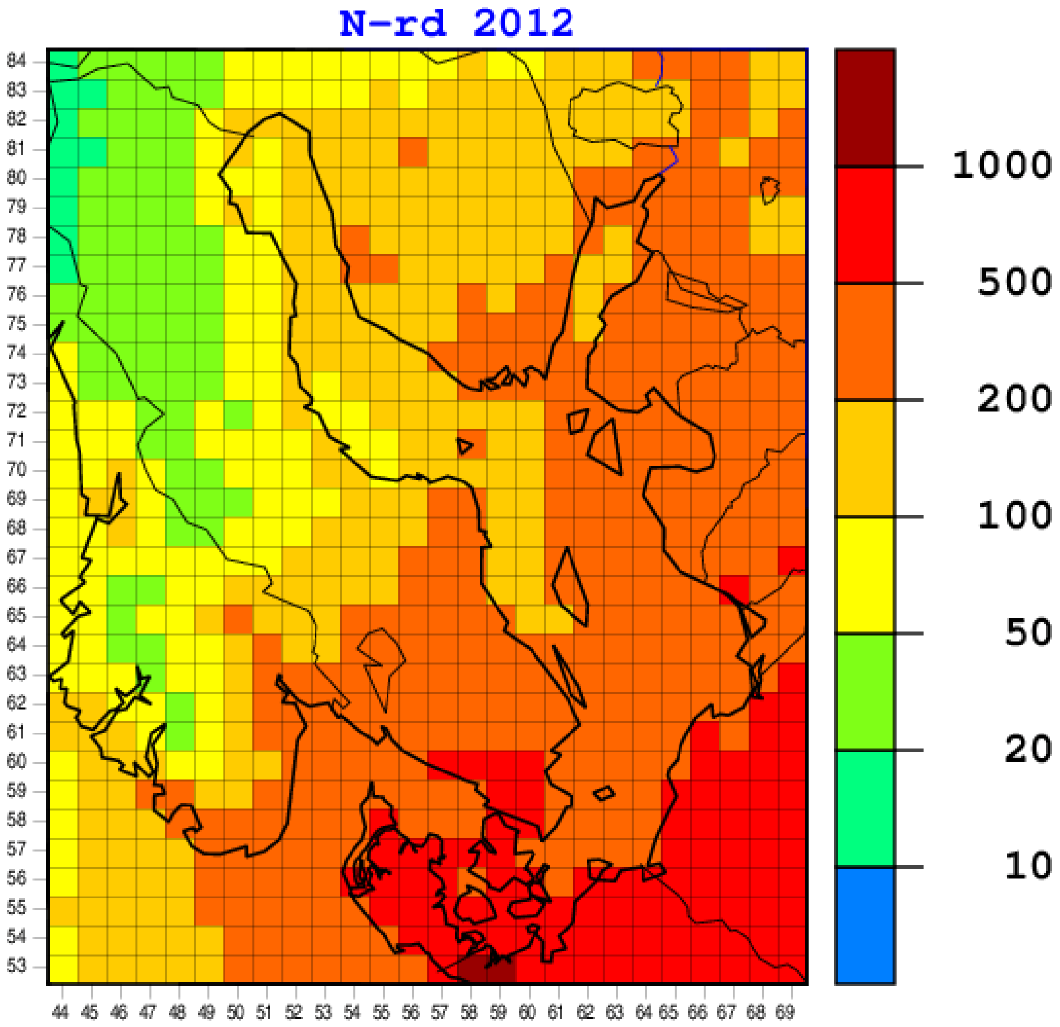


Figure 3.9. Map of annual deposition flux of reduced nitrogen (dry + wet) in 2012. Units: mg N m^{-2} yr^{-1}.

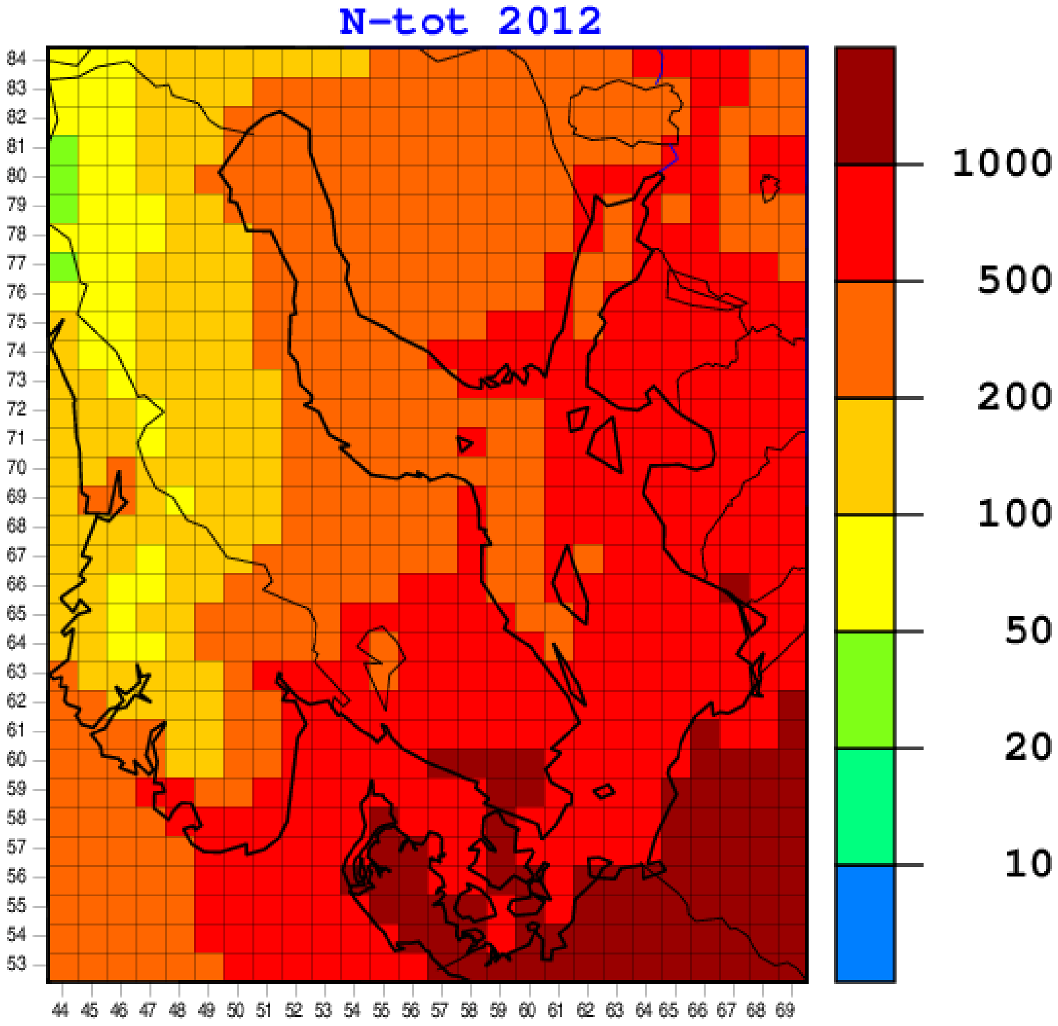


Figure 3.10. Map of annual deposition flux of total (oxidized + reduced) nitrogen in 2012. Units: mg N m^{-2} yr^{-1}.

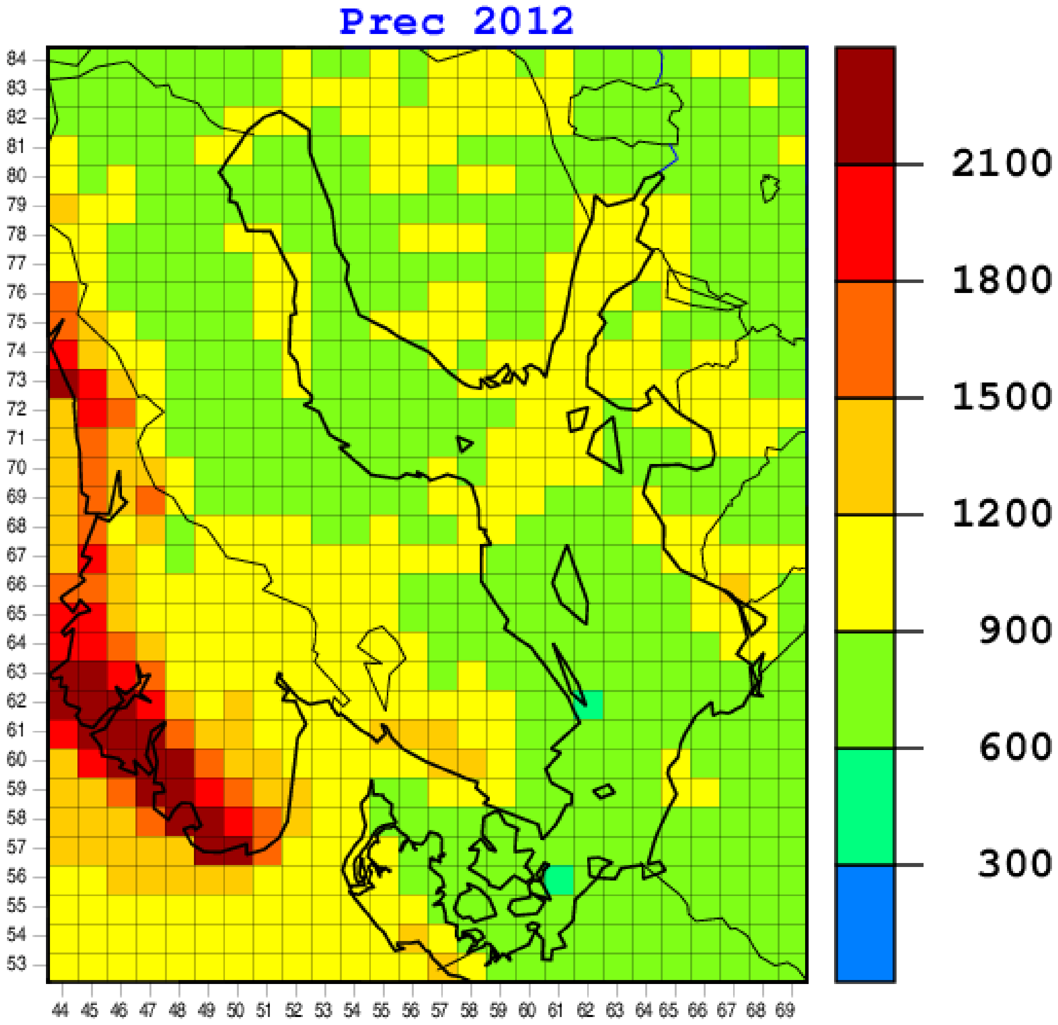


Figure 3.11. Map of annual precipitation in 2012. Units: mm yr^{-1}.

3.3 Normalised annual depositions

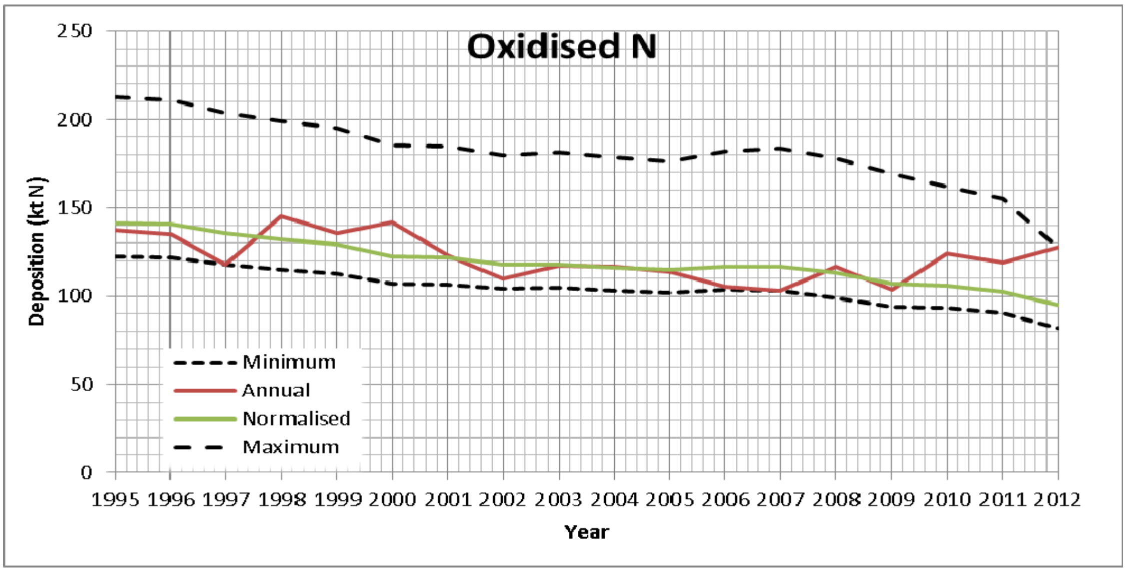


Figure 3.12. Normalised deposition of oxidised nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown. The minimum and maximum annual values are determined by the meteorological conditions for each particular year.

Table 3.3. Normalised deposition of oxidised nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown.

YEAR	MINIMUM	ANNUAL	NORMALISED	MAXIMUM
1995	122.3	137.1	141.0	213.0
1996	121.9	135.0	140.3	211.0
1997	117.9	117.8	135.7	203.7
1998	115.2	145.3	132.2	199.2
1999	112.5	135.6	128.9	194.8
2000	107.0	142.0	121.9	185.4
2001	106.4	122.9	121.5	184.6
2002	103.9	109.8	117.2	180.0
2003	104.5	117.4	117.4	181.0
2004	103.1	116.6	116.1	179.0
2005	102.0	114.1	115.0	176.0
2006	103.5	105.0	116.2	182.0
2007	102.7	102.7	116.5	184.0
2008	98.8	116.6	112.6	178.0
2009	94.0	103.8	106.5	169.0
2010	93.2	124.2	105.6	162.0
2011	90,1	119,0	102,3	155.0
2012	81.6	127.3	94.7	127.3

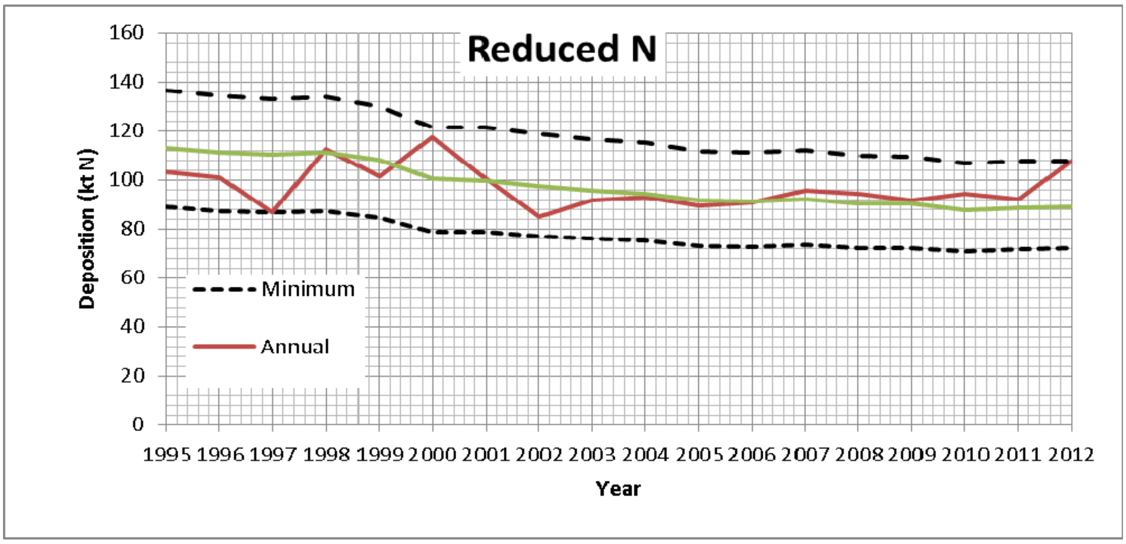


Figure 3.13. Normalised deposition of reduced nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown. The minimum and maximum annual values are determined by the meteorological conditions for each particular year.

Table 3.4. Normalised deposition of reduced nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown.

YEAR	MINIMUM	ANNUAL	NORMALISED	MAXIMUM
1995	89.0	103.0	111.8	136.7
1996	87.4	101.0	110.0	134.3
1997	86.7	86.7	109.3	133.1
1998	87.1	112.5	110.0	134.1
1999	84.5	101.6	107.2	130.0
2000	78.8	117.3	99.4	121.0
2001	78.7	100.5	98.9	121.4
2002	77.2	84.8	96.8	118.3
2003	76.1	91.7	95.1	116.8
2004	75.3	93.1	94.0	115.2
2005	73.1	89.5	91.2	111.8
2006	72.7	90.6	90.6	111.2
2007	73.5	95.7	91.4	111.9
2008	72.2	94.5	89.9	110.0
2009	72.1	91.8	89.6	109.3
2010	71.0	94.4	87.6	106.6
2011	71.9	97.2	88.6	107.5
2012	72.0	107.8	88.9	107.8

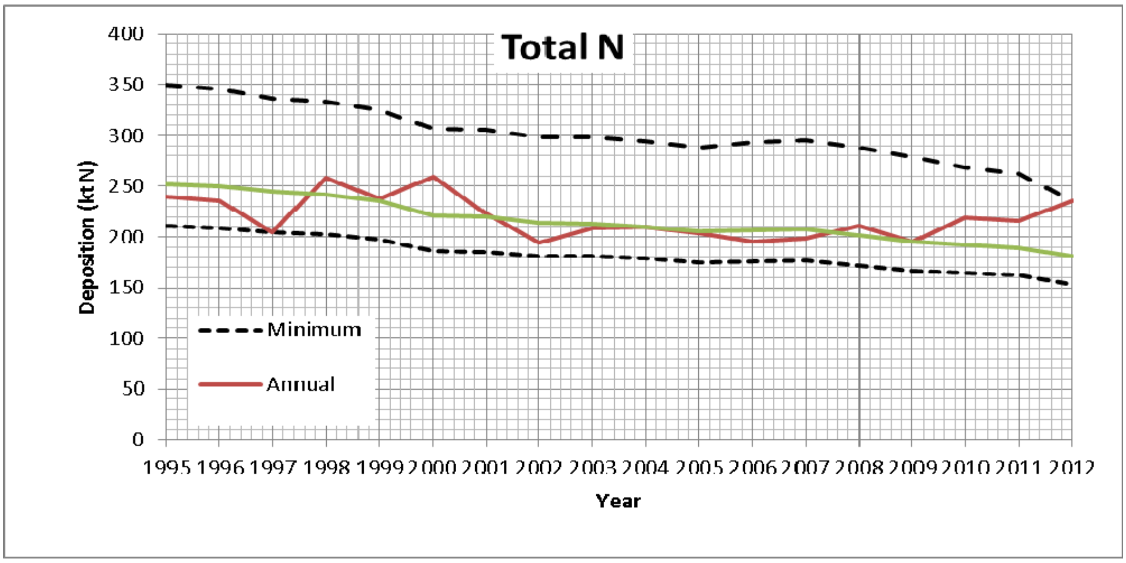


Figure 3.14. Normalised deposition of total nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown. The minimum and maximum annual values are determined by the meteorological conditions for each particular year.

Table 3.5. Normalised deposition of total nitrogen for the period 1995-2012. Minimum, maximum and actual annual values of the deposition are also shown.

YEAR	MINIMUM	ANNUAL	NORMALISED	MAXIMUM
1995	211.3	240.0	251.7	349.7
1996	209.3	235.9	249.2	345.3
1997	204.6	204.6	243.6	336.8
1998	202.3	257.8	240.7	333.2
1999	197.0	237.1	233.9	324.8
2000	185.8	259.3	219.9	306.6
2001	185.1	223.4	219.5	306.0
2002	181.1	194.6	214.0	298.4
2003	180.6	209.2	212.5	298.2
2004	178.4	209.7	209.7	293.9
2005	175.1	203.5	206.0	288.1
2006	176.2	195.5	206.7	293.0
2007	176.8	198.4	207.1	295.6
2008	171.7	211.1	201.9	287.7
2009	166.7	195.6	195.1	278.4
2010	164.2	218.6	191.4	268.4
2011	162.0	216.2	189.3	262.5
2012	153.6	235.1	180.9	235.1

3.4 Monthly depositions of nitrogen

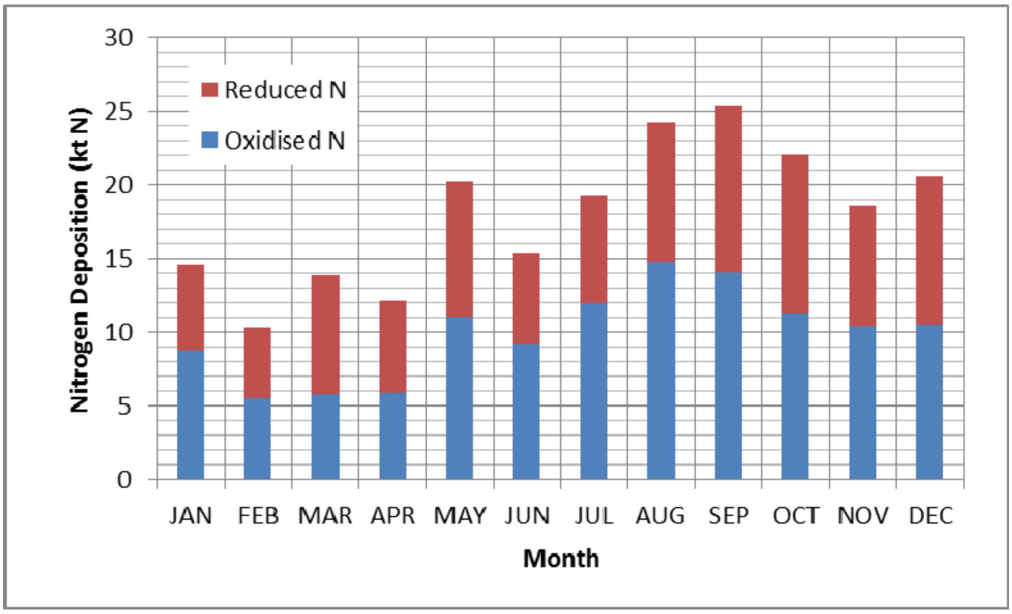


Figure 3.15. Monthly depositions of oxidized, reduced and total (oxidized +reduced) nitrogen to the entire Baltic Sea basin in 2012. Units: ktonnes N month^{-1}.

Table 3.6. Values of monthly depositions of oxidized, reduced and total (oxidized +reduced) nitrogen to the entire Baltic Sea basin in 2012. Units: ktonnes N month^{-1}.

Month	Oxidized	Reduced	Total
January	7.0	5.5	12.4
February	7.1	6.2	13.3
March	4.6	5.6	10.2
April	10.7	12.7	23.4
May	8.4	6.8	15.2
June	9.8	7.4	17.2
July	11.4	7.3	18.7
August	11.8	8.2	20.0
September	15.9	13.0	29.0
October	12.7	12.0	24.7
November	15.0	13.0	27.9
December	15.0	12.2	27.2

3.5 Comparison with observations

Model results of deposition of nitrogen and ammonia for 2012 are validated against measurements collected from the EMEP monitoring network for 2012. Figure 3.16 – 3.19 show the daily time series of concentration of nitrate and ammonium respectively in precipitation compared with observations for reported stations in HELCOM countries where daily observational data were available for the year 2012. The correlation (Corr.) and Root Mean Square Error RMSE = $\sqrt{\frac{1}{n}\sum_{1=1}^{n}(m_i - o_i)^2}$ where m_i and o_i, are modelled and measured concentration in monitoring station i), between the measurement and model results are summarized in Table 3.7 and Table 3.8 Slight underestimation can be found in most of the stations for both nitrate and ammonia mainly during summer. The precipitation field in EMEP model is an input variable from the meteorological data and thus deposition pattern depends very much on the seasonality of precipitation. But on an average the mean values of observation and model shows good agreement for the year.

Table 3.7. Annual mean values of measured and modeled nitrate concentration in precipitation in 2012 for selected stations. Units: mg N l^{-1}.

Station	Observation	Model	Corr.	RMSE
Lahemaa(EE)	0.29	0.37	0.24	0.49
Langenbruegge (DE)	0.54	0.82	0.04	2.70
Schauinsland (DE)	0.34	0.39	0.46	0.53
Neuglobsow (DE)	0.68	0.76	0.25	1.11
Preila (LT)	0.64	0.43	0.60	0.54
Jarczew (PL)	0.74	0.91	0.23	1.45
Sniezka (PL)	0.85	0.71	-0.03	1.81
Leba (PL)	0.61	0.52	0.43	0.62
Diabla Gora (PL)	0.79	0.88	0.55	1.29
Pinega (RU)	0.29	0.15	0.60	0.27
Janiskoski (RU)	0.19	0.11	0.35	0.26
Danki (RU)	0.53	0.55	0.49	0.65
Lesnoy (RU)	0.39	0.34	0.47	0.37
Raaoe (SE)	0.65	0.49	0.64	0.59

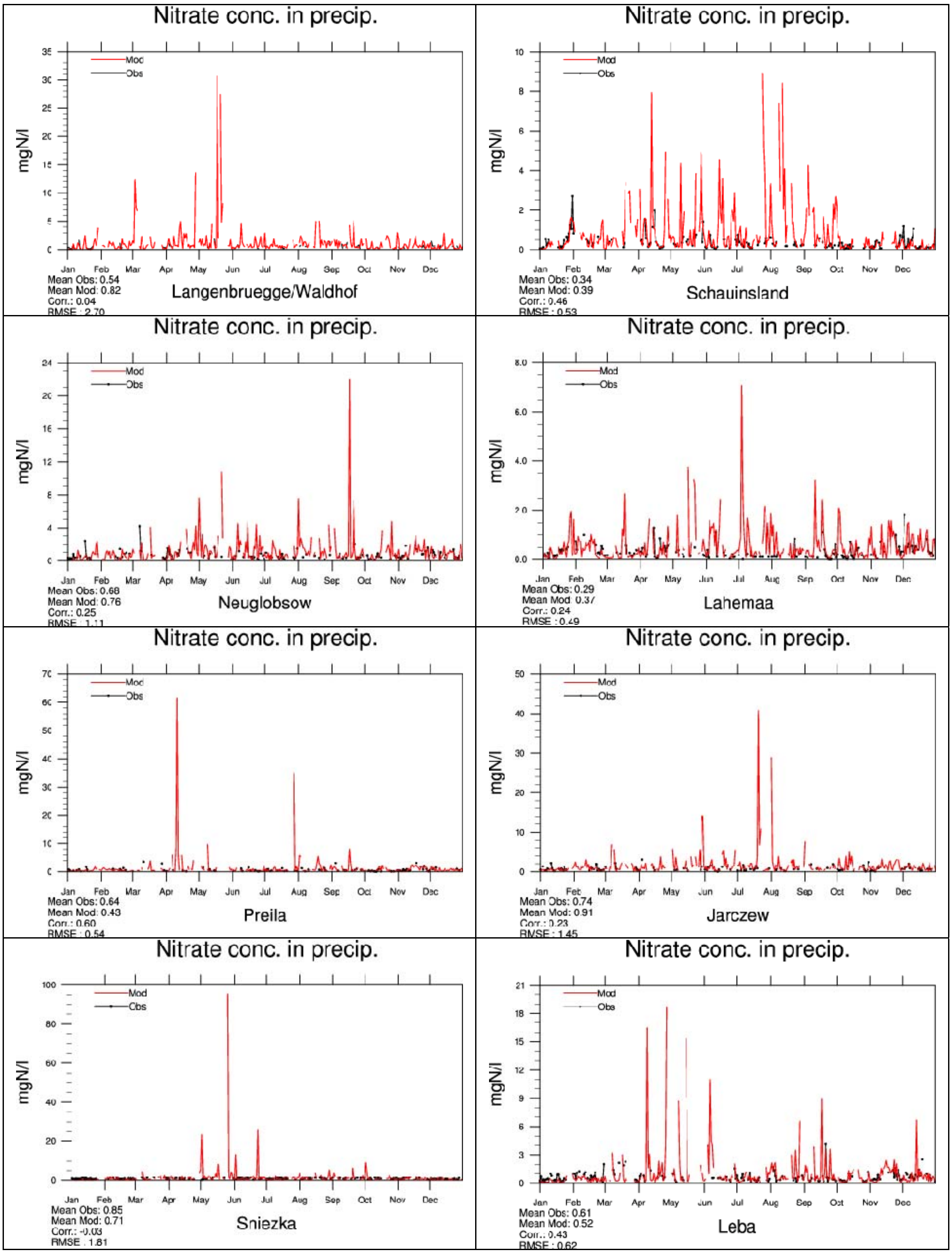


Figure 3.16. Time series of daily concentration of Nitrate in precipitation (mg N/l) to the stations in HELCOM countries (having measurements) for the year 2012.

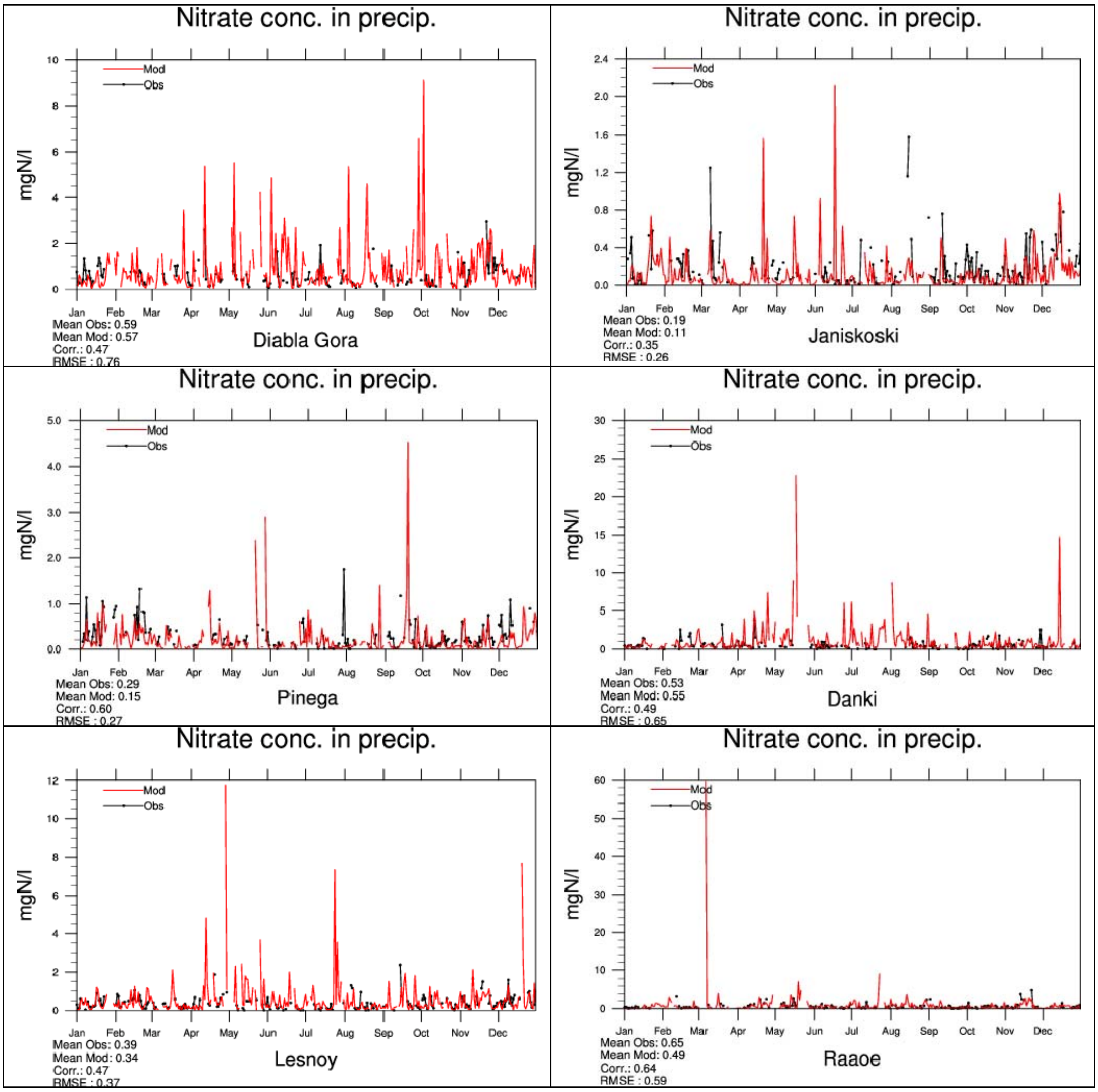


Figure 3.17. Time series of daily concentration of Nitrate in precipitation (mg N/l) to the stations in HELCOM countries (having measurements) for the year 2012.

Table 3.8. Annual mean values of measured and modeled ammonium concentration in precipitation in 2012 for selected stations. Units: mg N l^{-1}.

Station	Observation	Model	Corr.	RMSE
Lahemaa(EE)	0.17	0.27	0.69	0.29
Langenbruegge (DE)	0.83	1.18	0.06	3.59
Schauinsland (DE)	0.47	0.61	0.49	0.67
Neuglobsow (DE)	0.91	0.84	0.42	0.82
Preila (LT)	0.61	0.79	0.29	1.75
Jarczew (PL)	0.97	0.82	0.19	1.40
Sniezka (PL)	0.52	0.67	0.06	1.02
Leba (PL)	0.56	0.42	0.47	0.54
Diabla Gora (PL)	0.54	0.57	0.50	0.64
Pinega (RU)	0.53	0.09	0.08	0.63
Janiskoski (RU)	0.24	0.04	0.27	0.33
Danki (RU)	0.49	0.49	0.60	0.53
Lesnoy (RU)	0.43	0.28	0.47	0.42
Raaoe (SE)	0.58	0.43	0.67	0.54

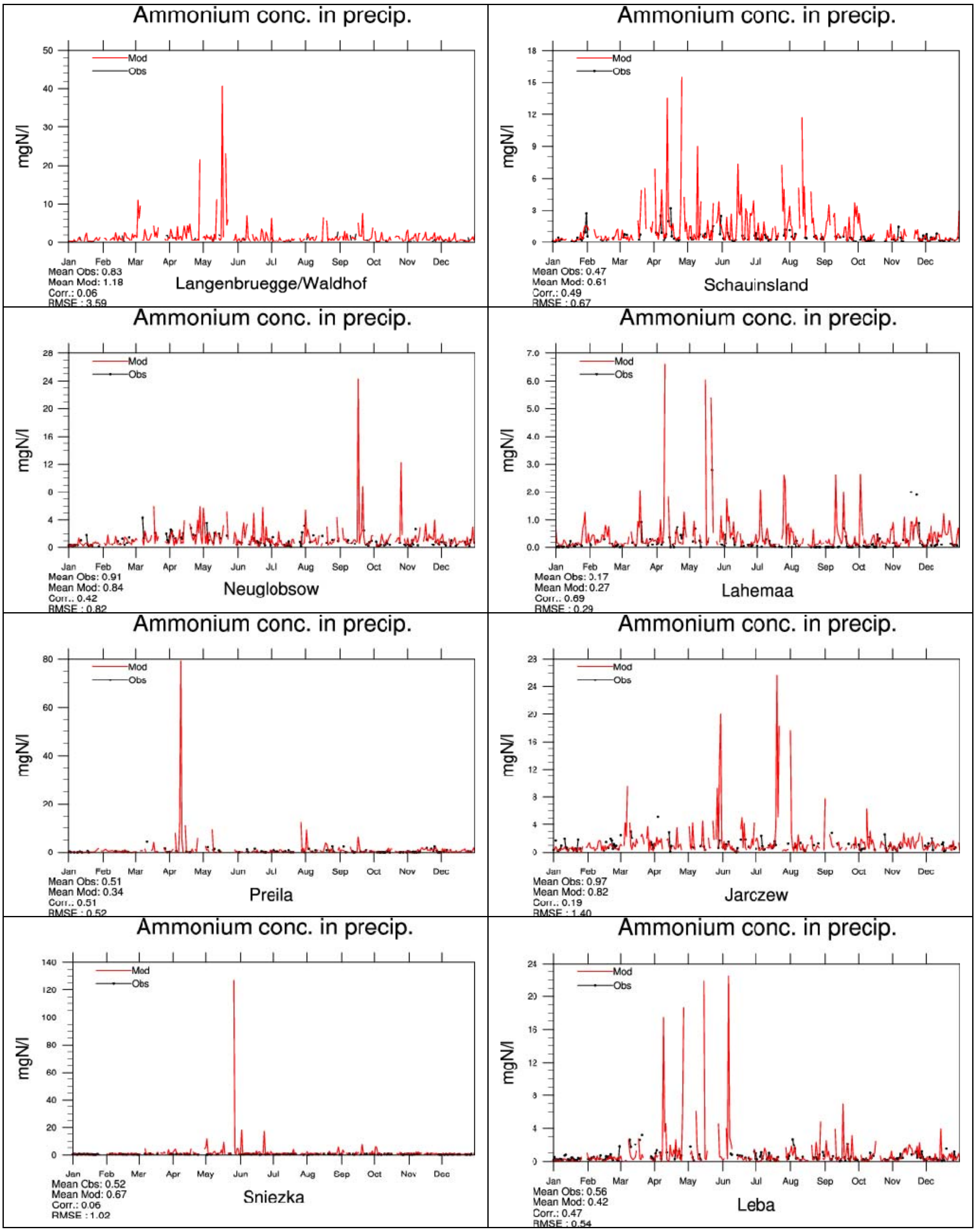


Figure 3.18. Time series of daily concentration of Ammonium in precipitation (mg N/l) to the stations in HELCOM countries (having measurements) for the year 2012.

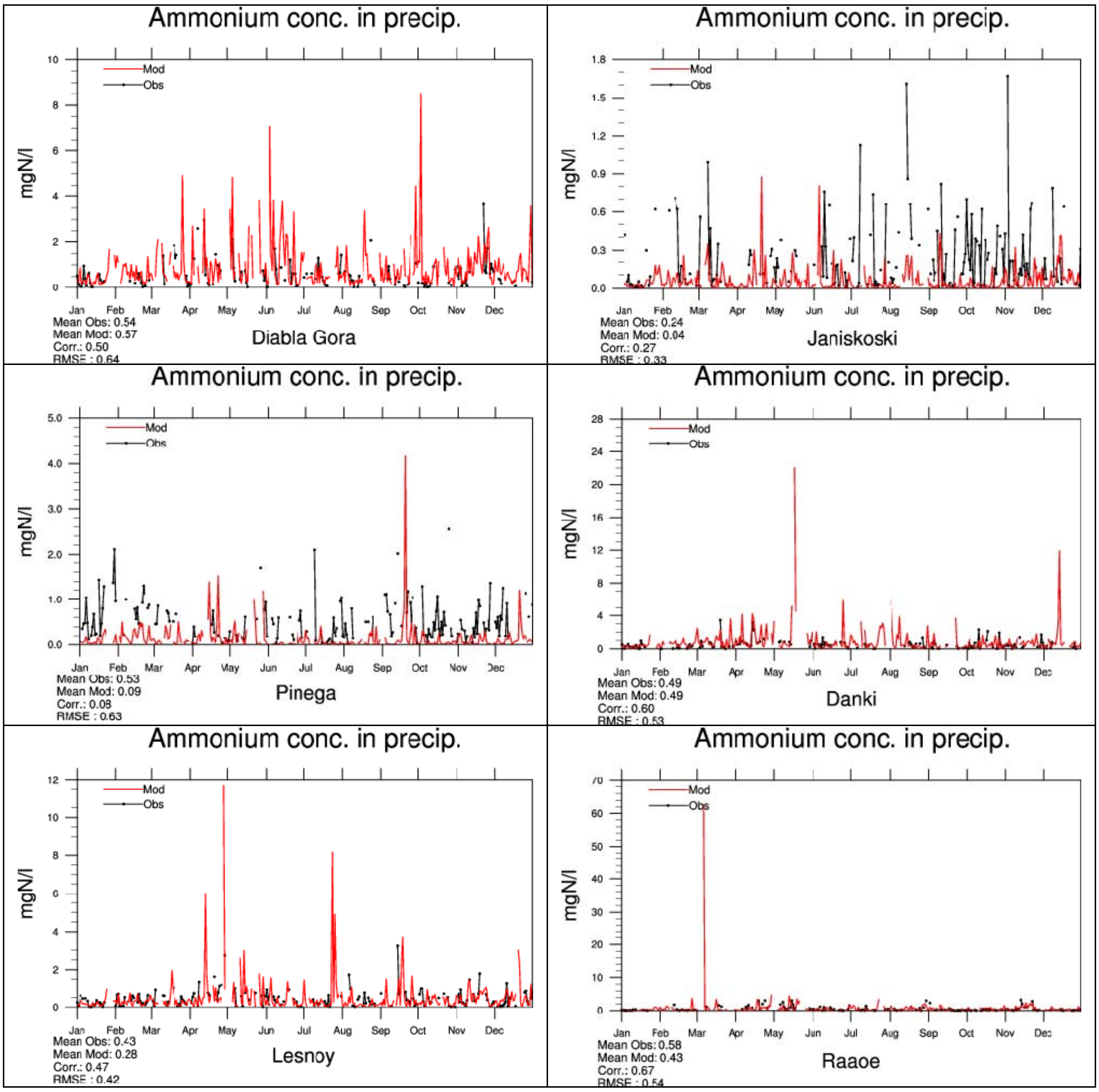


Figure 3.19. Time series of daily concentration of Ammonium in precipitation (mg N/l) to the stations in HELCOM countries (having measurements) for the year 2012.

3.6 Source allocation of nitrogen deposition

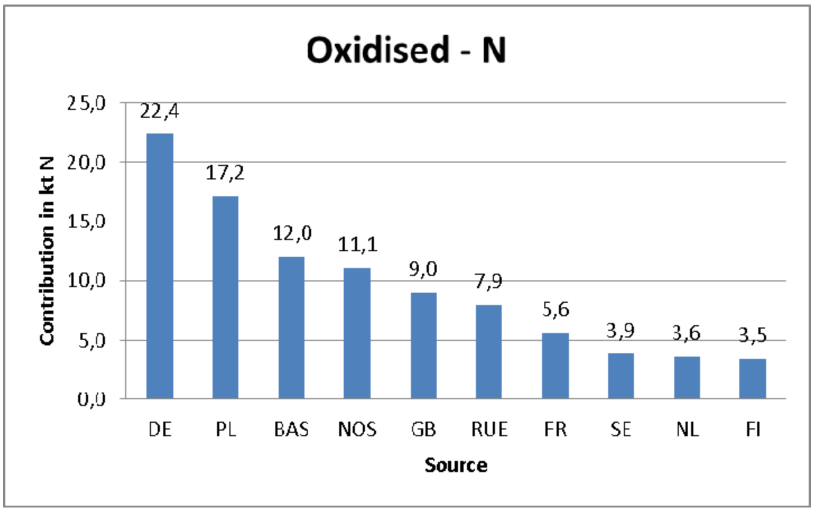


Figure 3.20. Top ten sources with highest contributions of nitrogen emissions to annual deposition of oxidised nitrogen into the Baltic Sea basin in the year 2012. BAS and NOS denote ship emissions form the Baltic Sea and from the North Sea, respectively. RUE denotes the contributions from emissions in extended Russian territory.

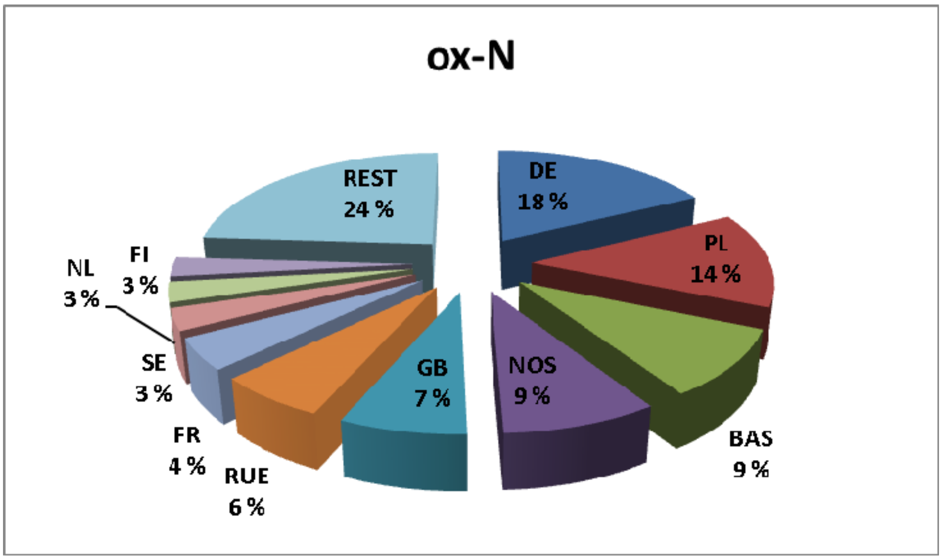


Figure 3.21. Relative top ten contributions of nitrogen emissions to annual deposition of oxidised nitrogen into the Baltic Sea basin in the year 2012. REST denotes remaining emission sources in the EMEP domain. RUE denotes the contributions from emissions in extended Russian territory. Units: % of total deposition of oxidized nitrogen.

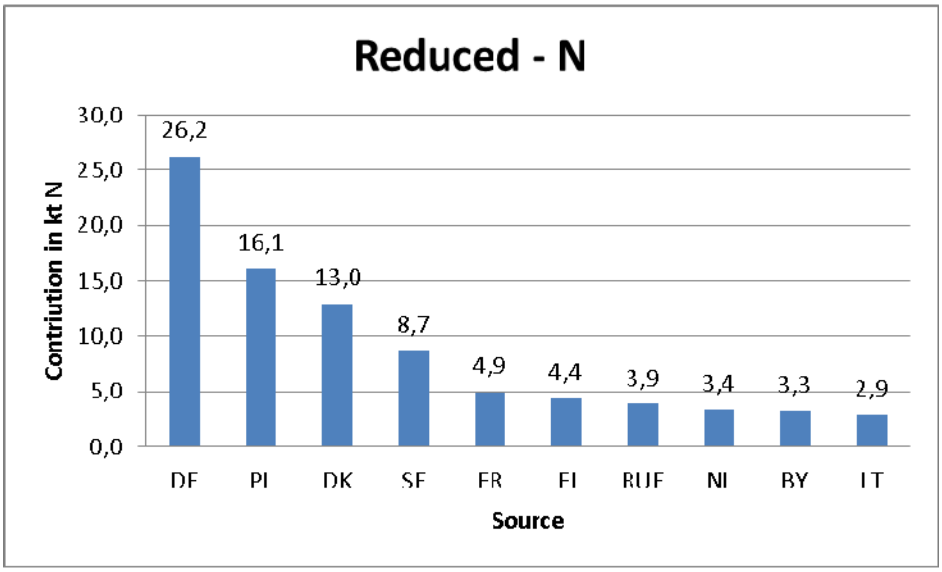


Figure 3.23. Top ten sources with highest contributions of nitrogen emissions to annual deposition of reduced nitrogen into the Baltic Sea basin in the year 2012. BAS and NOS denote ship emissions form the Baltic Sea and from the North Sea, respectively. RUE denotes the contributions from emissions in extended Russian territory.

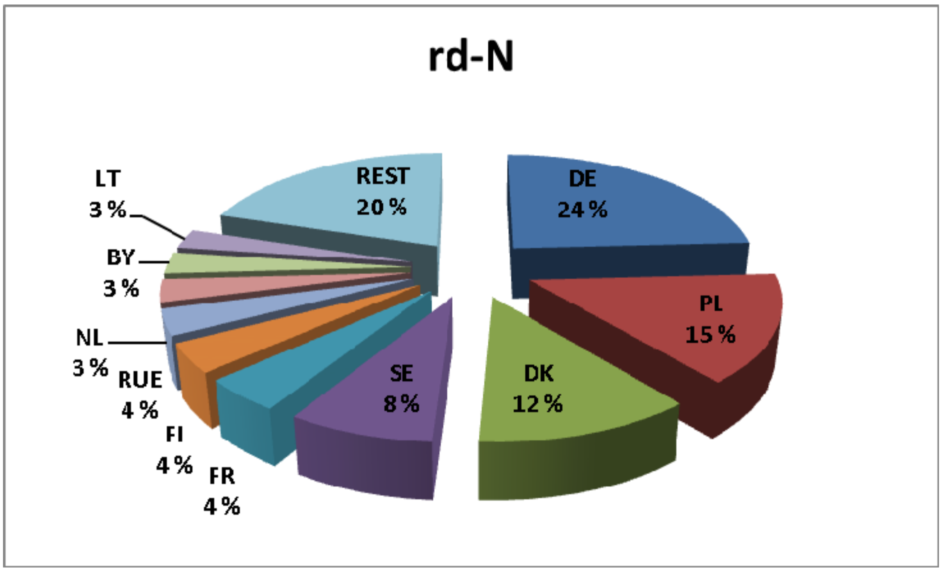


Figure 3.24. Relative top ten contributions of nitrogen emissions to annual deposition of reduced nitrogen into the Baltic Sea basin in the year 2012. REST denotes remaining emission sources in the EMEP domain. RUE denotes the contributions from emissions in extended Russian territory. Units: % of total deposition of oxidized nitrogen.

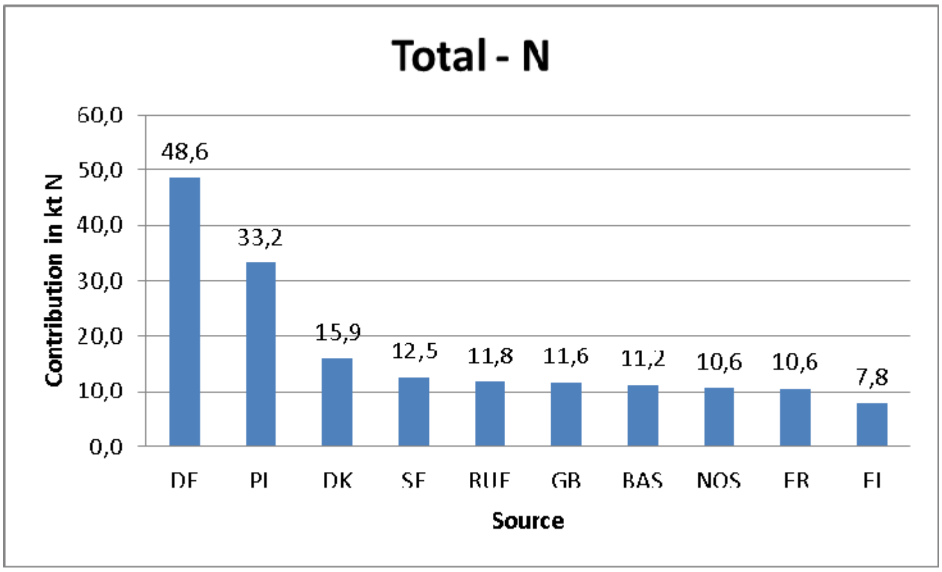


Figure 3.25. Top ten sources with highest contributions of nitrogen emissions to annual deposition of total nitrogen into the Baltic Sea basin in the year 2012. BAS and NOS denote ship emissions form the Baltic Sea and from the North Sea, respectively. RUE denotes the contributions from emissions in extended Russian territory.

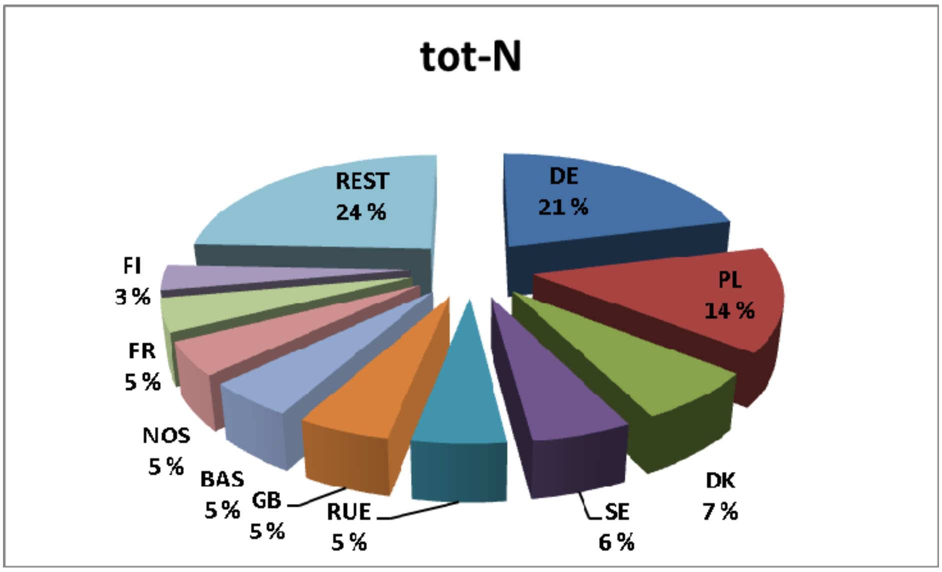


Figure 3.26. Relative top ten contributions of nitrogen emissions to annual deposition of reduced nitrogen into the Baltic Sea basin in the year 2012. REST denotes remaining emission sources in the EMEP domain. RUE denotes the contributions from emissions in extended Russian territory. Units: % of total deposition of oxidized nitrogen.

3.7 Conclusions for Chapter 3

- Compared to 2011 nitrogen oxides emissions in 2012 are lower (1-13%) in six out of nine HELCOM Contracting Parties and these are Denmark, Estonia, Finland, Germany, Poland and Sweden. Other three countries (Latvia, Lithuania and Russia) reported increased (2-15%) nitrogen oxides emissions in 2012 compared to 2011. Ship emissions from the Baltic Sea were also 1.6% higher in 2012 than in 2011.

- Annual 2012 ammonia emissions are higher than that of 2011 ammonia emissions in five out of nine HELCOM countries: Denmark, Estonia, Latvia, Lithuania and Russia. Maximum increase is reported in Latvia (30%) followed by Lithuania (23%), though the absolute values are very small in these countries. The 2012 emissions are lower than 2011 emissions in four CPs: Finland (2%), Germany (3%), Poland (3%) and Sweden (2%).

- Among the HELCOM Contracting Parties, the largest percent of 2012 nitrogen emissions deposited to the Baltic Sea basin can be noticed for Denmark (16.3%) and the lowest for Russia (0.59%).

- Spatial distributions of nitrogen oxides and ammonia emissions in 2012 are very similar to the distributions in 2011.

- Combustion and transportation SNAP sectors are the main sources of nitrogen oxides emissions, whereas agriculture is the dominating sector for ammonia emissions, for all HELCOM CPs.

- Calculated annual deposition of total nitrogen to the Baltic Sea basin in 2012 was 233 kt., approximately 8% higher than in 2011. Deposition of oxidised nitrogen accounted for 53% of total nitrogen deposition in 2012.

- Spatial distributions of nitrogen depositions to the Baltic Sea basin in 2012 are similar to those in 2011.

- Normalised depositions of oxidized, reduced and total nitrogen to the Baltic Sea show clear decreasing pattern in the period 1995-2012.

- No clear seasonal pattern can be found in monthly nitrogen depositions in 2012. The maximum of the deposition occurs in August and September.

- Germany, Poland, ship traffic on the North Sea and on the Baltic Sea are the main emission sources contributing to oxidized nitrogen deposition into the Baltic Sea basin in 2012. The main difference between 2011 and 2012 is higher contribution Germany and Poland to oxidized nitrogen deposition in 2011.

- As in previous years, Germany, Poland and Denmark are top three sources contributing to reduced nitrogen deposition into the Baltic Sea basin in 2012. Germany is the top contributor followed by Poland. Denmark was the second largest contributor to reduced nitrogen deposition into Baltic Sea in 2011.

- As in previous years, also in 2012 some distant sources like United Kingdom. France and ship traffic on the North Sea contribute significantly to total nitrogen deposition into the Baltic Sea basin.

- The main sources contributing to total nitrogen deposition to the Baltic Sea basin are: Germany, Poland, Denmark, and Sweden. Compared to 2011, contribution from the United Kingdom is lower by 23% and contribution from Russia is higher by 47% in 2012. Contribution of other distant sources like ship traffic on the North Sea, France and the Netherlands is also significant.

4. Atmospheric Supply of Lead to the Baltic Sea in 2012

This chapter presents the results of model evaluation of lead atmospheric input to the Baltic Sea and its sub-basins in 2012. Modelling of lead atmospheric transport and deposition was carried out using MSC-E Eulerian Heavy Metal transport model MSCE-HM (*Travnikov and Ilyin*, 2005). Latest available official information on lead emission from HELCOM countries and other European countries for 2012 was used in model simulations. Based on these data annual and monthly levels of lead deposition to the Baltic Sea region have been obtained and contributions of HELCOM countries emissions to the deposition over the Baltic Sea are estimated. Model results were compared with observed levels of lead concentrations in air and precipitation measured at monitoring sites around the Baltic Sea in 2012.

4.1 Lead emissions

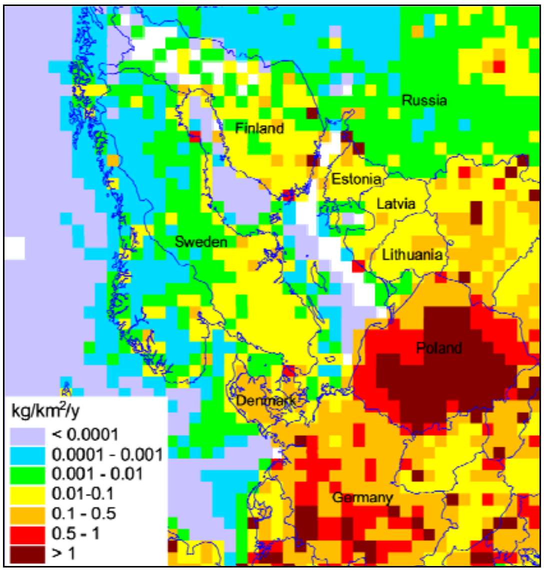


Figure 4.1. Annual total anthropogenic emissions of **lead** in the Baltic Sea region for 2012, kg/km^2/y.

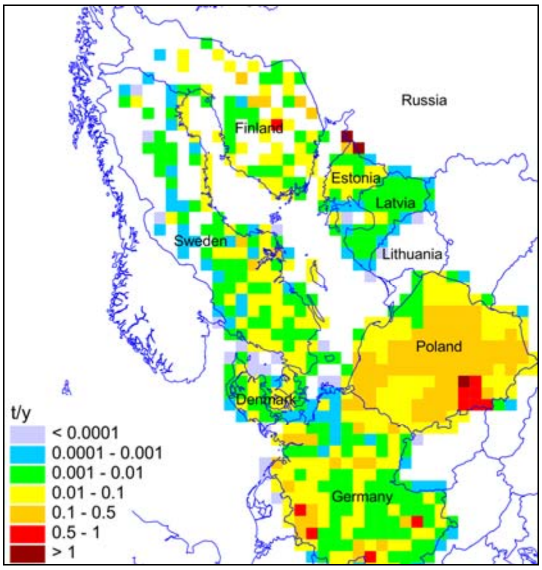


Figure 4.2. Annual lead emission from Public Power sector for 2012, t/grid cell/y (white color means no information).

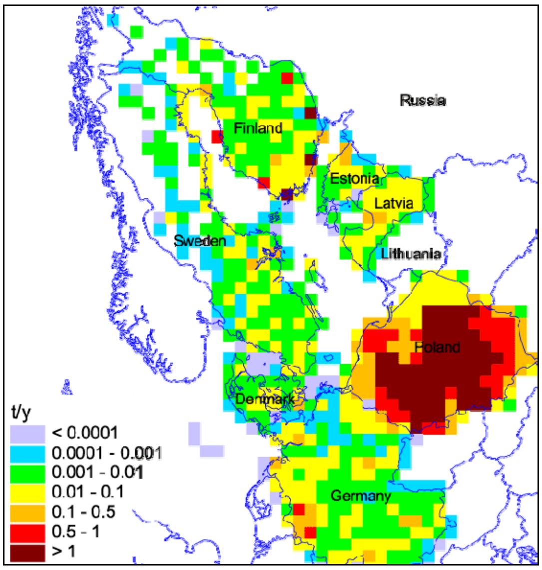


Figure 4.3. Annual lead emission from Industrial Combustion sector for 2012, t/grid cell/y (white color means no information).

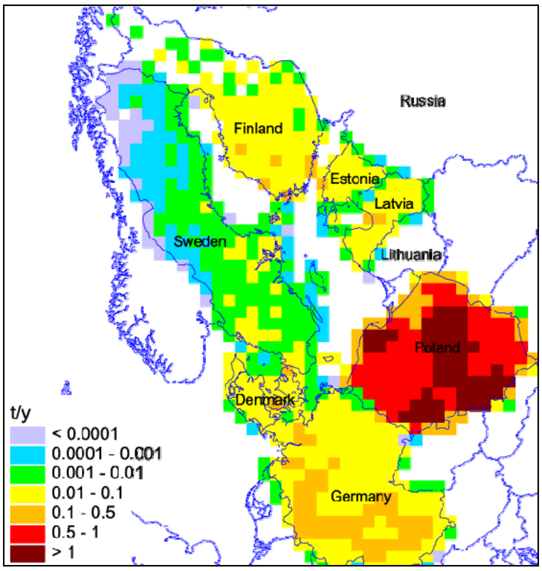


Figure 4.4. Annual lead emission from Small Combustion sector for 2012, t/grid cell/y (white color means no information).

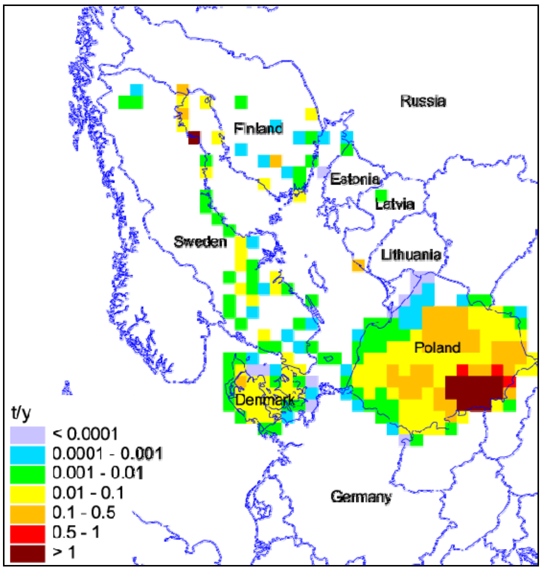


Figure 4.5. Annual lead emission from Industrial Processes sector for 2012, t/grid cell/y (white color means no information).

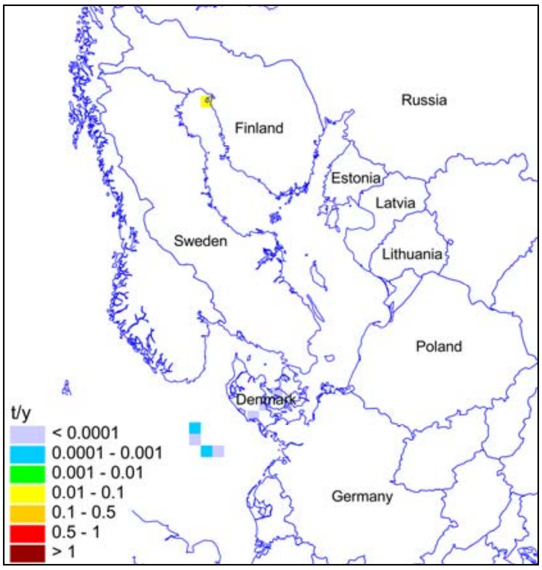


Figure 4.6. Annual lead emission from Fugitive Emissions sector for 2012, t/grid cell/y (white color means no information).

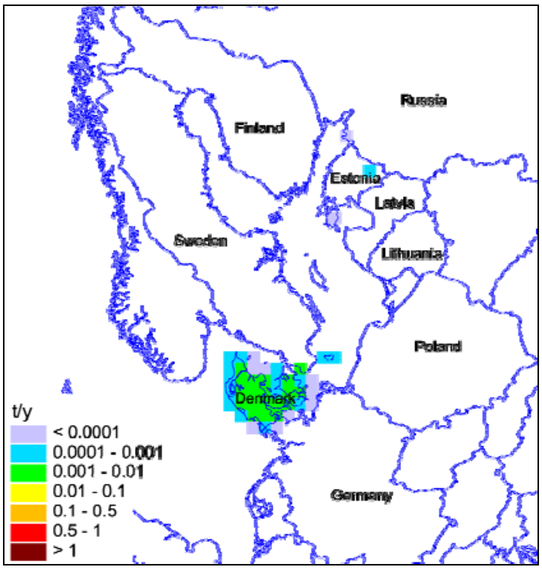


Figure 4.7. Annual lead emission from Solvents sector for 2012, t/grid cell/y (white color means no information).

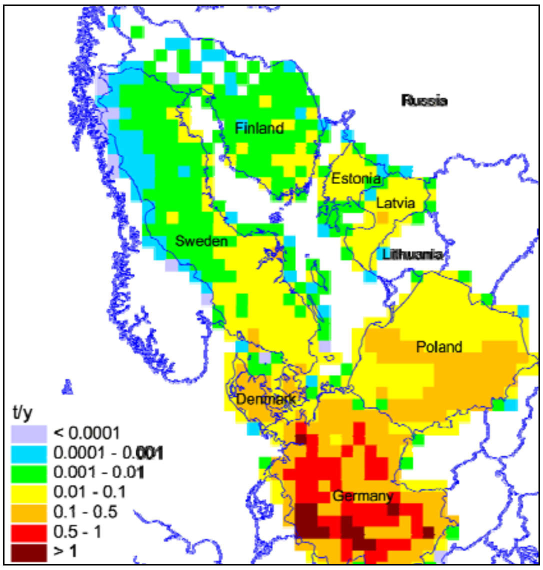


Figure 4.8. Annual lead emission from Road Rail sector for 2012, t/grid cell/y (white color means no information).

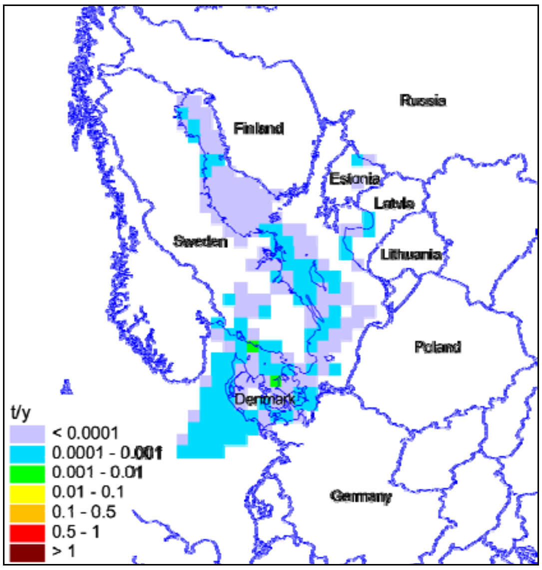


Figure 4.9. Annual lead emission from Shipping Emissions sector for 2012, t/grid cell/y (white color means no information).

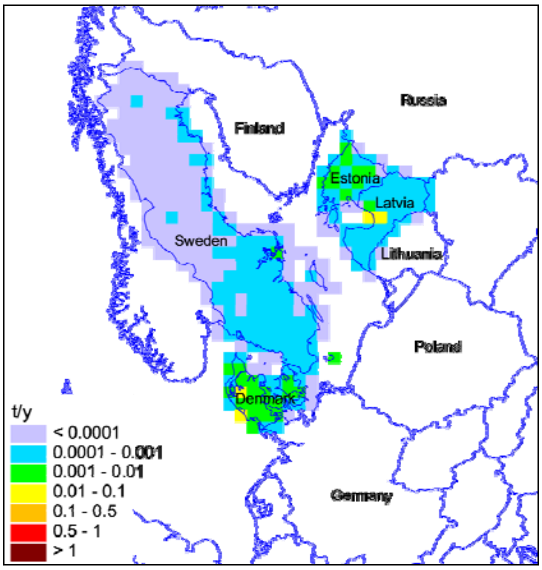


Figure 4.10. Annual lead emission from Off Road Mobility sector for 2012, t/grid cell/y (white color means no information).

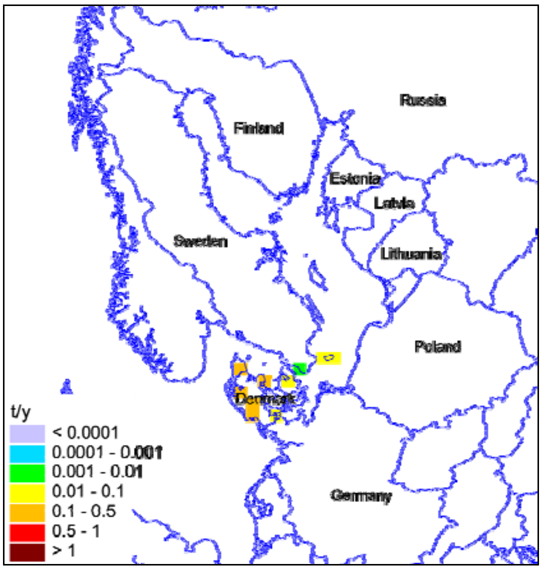


Figure 4.11. Annual lead emission from Civil Aviation sector for 2012, t/grid cell/y (white color means no information).

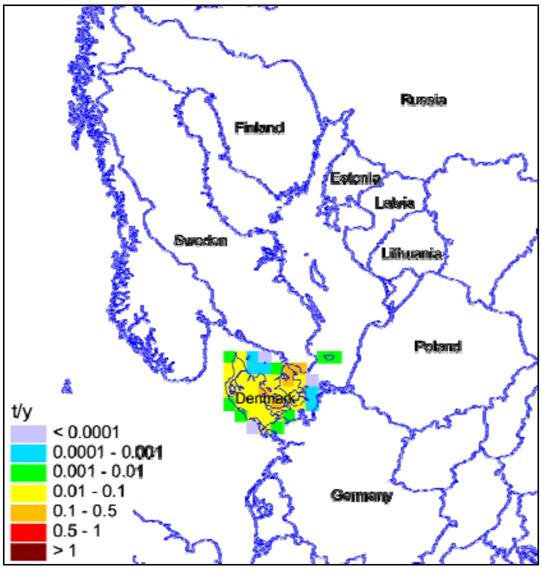


Figure 4.12. Annual lead emission from Other Waste Displacement sector for 2012, t/grid cell/y (white color means no information).

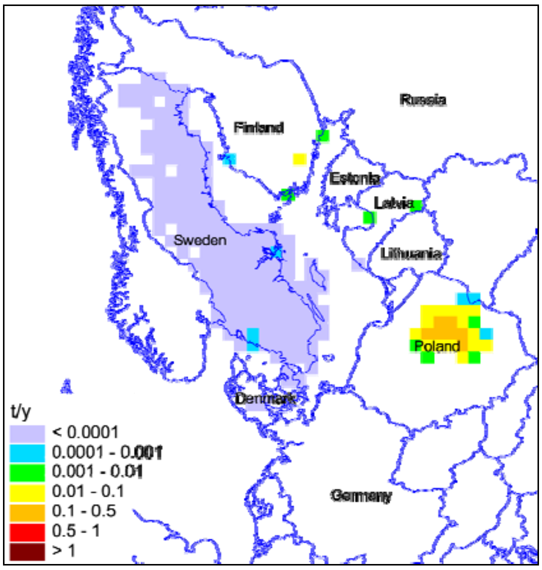


Figure 4.13. Annual lead emission from Waste Incineration sector for 2012, t/grid cell/y (white color means no information).

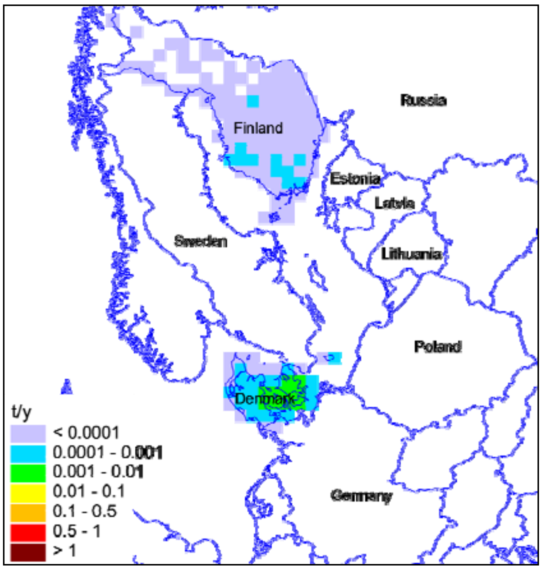


Figure 4.14. Annual lead emission from Agricultural Wastes sector for 2012, t/grid cell/y (white color means no information).

Table 4.1. Annual total **lead** anthropogenic emissions of HELCOM countries from different sectors for 2012, tonnes/year

GNFR emission sector	Sector name	Denmark	Estonia	Finland	Germany	Latvia	Lithuania	Poland	Russia	Sweden
A	Public Power	0.32	31.82	2.98	10.25	0.13	0.37	24.50	0.40	2.36
B	Industrial Combustion	0.45	0.47	10.67	6.61	0.70	0.83	293.41	12.11	1.55
C	Small Combustion	1.67	0.79	3.52	14.79	1.03	1.15	137.29	0.60	0.88
D	Industrial Processes	0.84	0.01	0.66	61.95	0.28	0.06	79.39	5.80	2.98
E	Fugitive Emissions	0.001	0.0001	0.023	NA			1.956	0.020	
F	Solvents	0.07	0.0003	5.3E-07	NA	NA		2.3E-06		2.1E-07
G	Road Rail	5.55	0.46	0.67	86.27	1.48	1.83	15.24		3.18
H	Shipping emissions	0.03	0.0002		0.0023	0.001	0.001	3.6E-07		0.02
I	Off Road Mobility	0.09	0.03	NA		0.05	0.08			0.03
J	Civil aviation	0.86	NA		5.29	NE	NE	NA		0.36
L	Other Waste Displacement	1.90	0.0007	NA	NA	NA		NA		
M	Waste water	NA	NA	NA	NA	NO	NE	NA	0.10	NA
N	Waste Incineration	0.0002		0.068	0.00001	0.005	0.34	1.77	0.03	0.003
Q	Agricultural wastes	0.04	NE	0.005	NO	NA	NO	NA		NO
R	Other	NO	NO	NO	NO	NA	NO	NO	13.04	NO
Total		11.8	33.6	18.6	185.2	3.7	4.7	553.6	32.1	11.4

NO – not occurring, an activity or process does not exist within a country.
NA – not applicable, the process or activity exists but emissions are considered never to occur.
NE – not estimated, emissions occur but have not been estimated or reported in this submission.
IE – included elsewhere, emissions by sources of compounds are estimated but included elsewhere in the inventory.

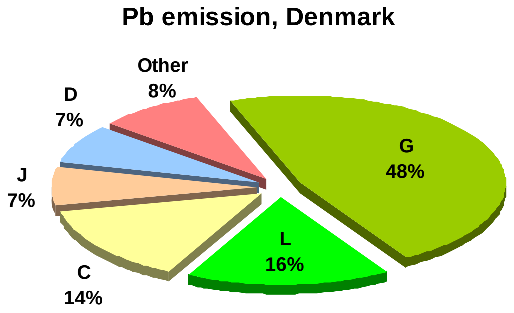


Figure 4.15. Contributions of different sector to total annual lead emission of Denmark in 2012.

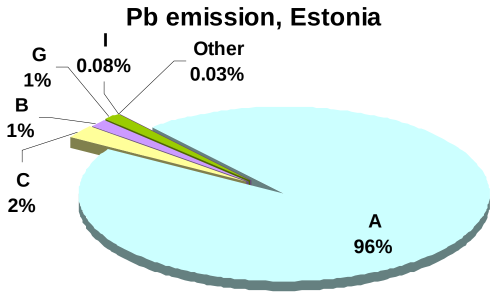


Figure 4.16. Contributions of different sector to total annual lead emission of Estonia in 2012.

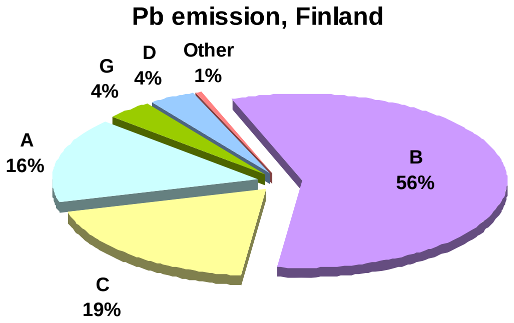


Figure 4.17. Contributions of different sector to total annual lead emission of Finland in 2012.

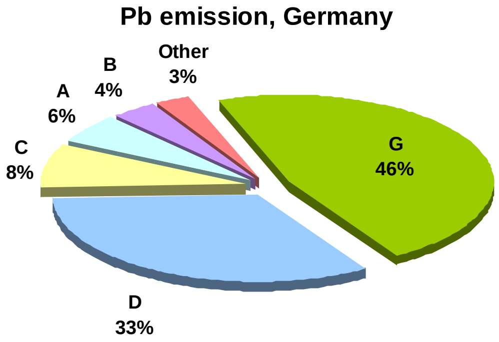


Figure 4.18. Contributions of different sector to total annual lead emission of Germany in 2012.

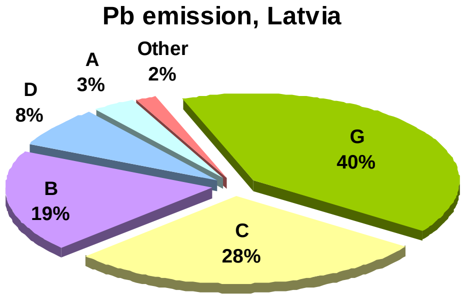


Figure 4.19. Contributions of different sector to total annual lead emission of Latvia in 2012.

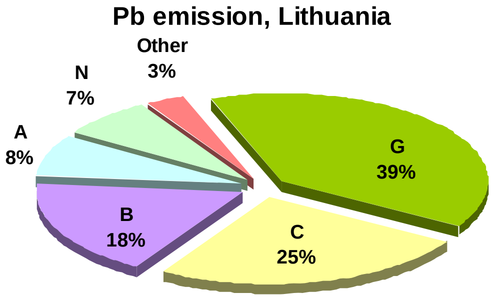


Figure 4.20. Contributions of different sector to total annual lead emission of Lithuania in 2012.

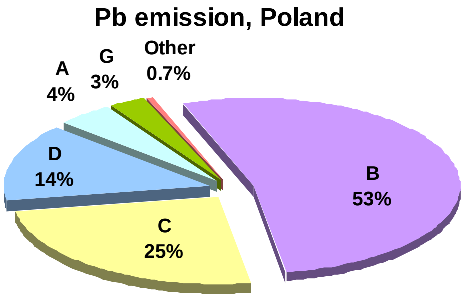


Figure 4.21. Contributions of different sector to total annual lead emission of Poland in 2012.

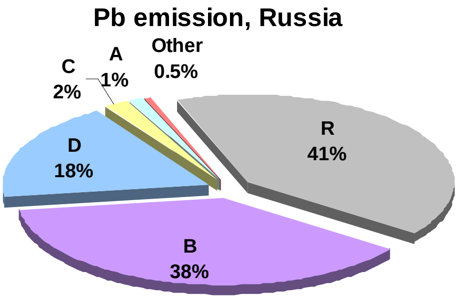


Figure 4.22. Contributions of different sector to total annual lead emission of Russia in 2012.

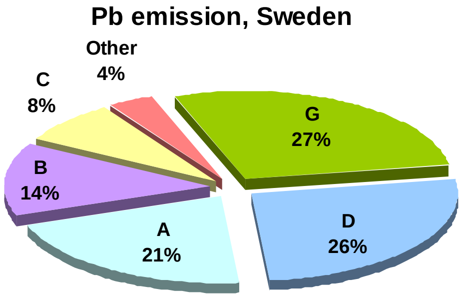
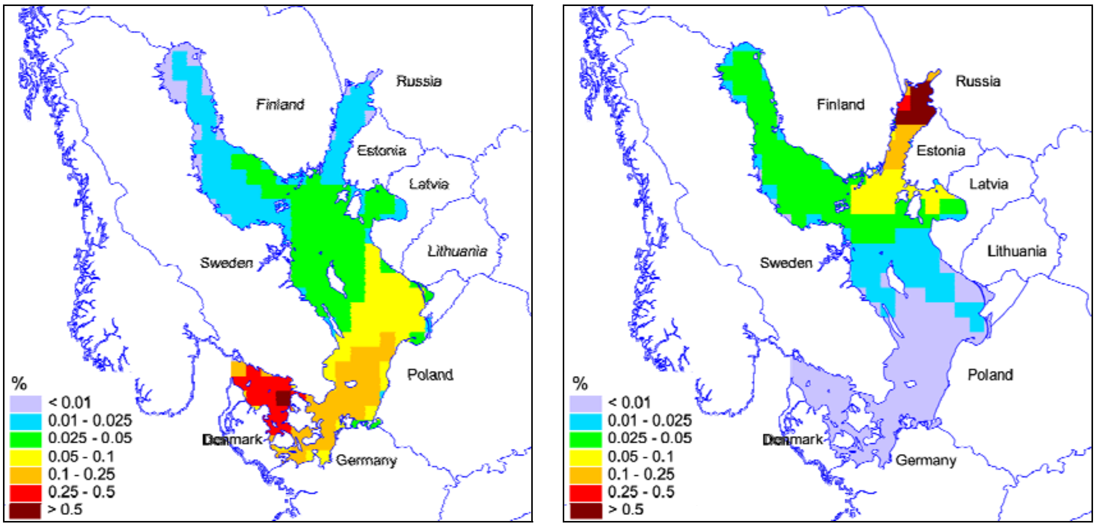
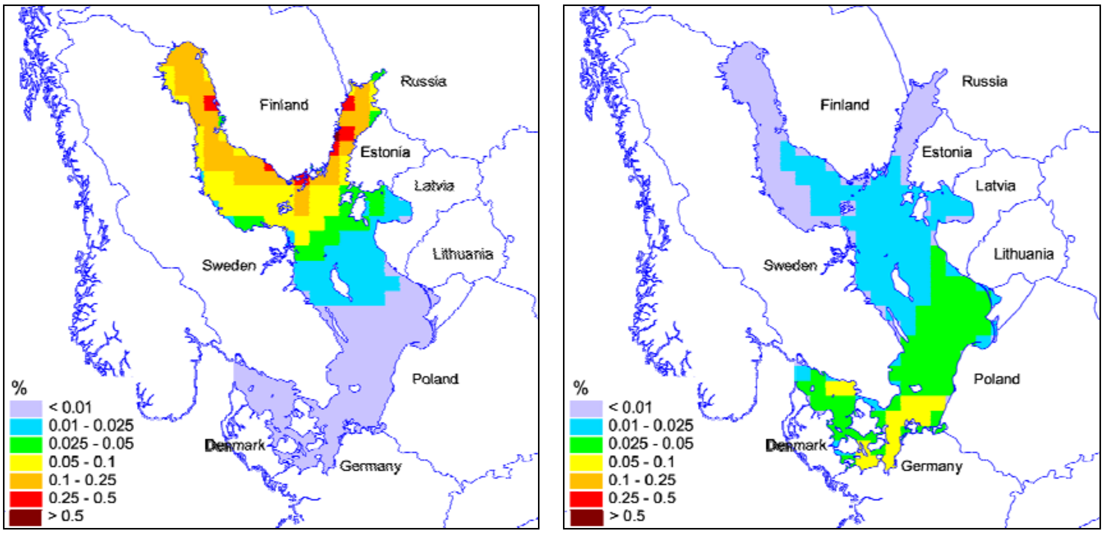


Figure 4.23. Contributions of different sector to total annual lead emission of Sweden in 2012.

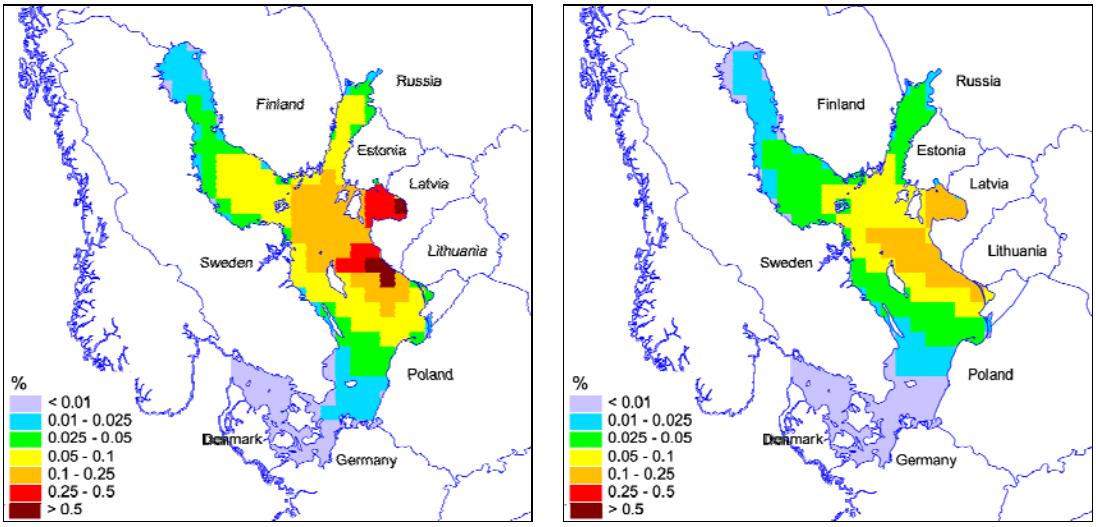


Denmark Estonia

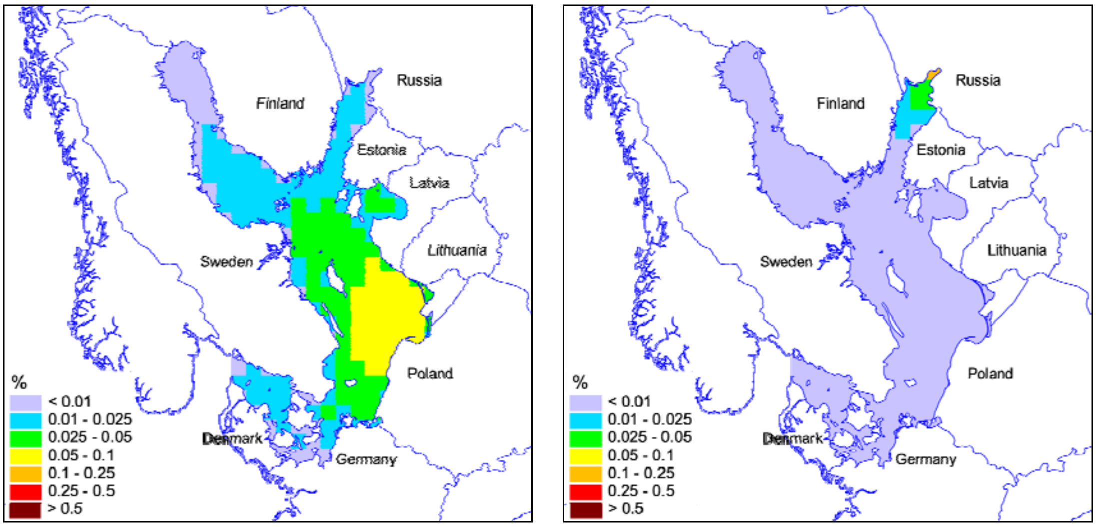


Finland Germany

Figure 4.24. Fractions of annual anthropogenic lead emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

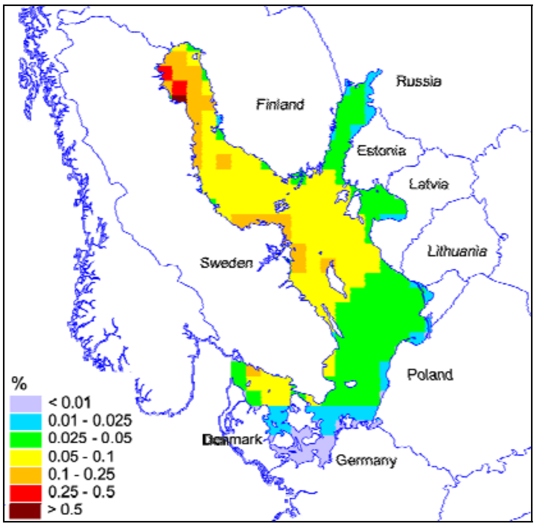


Latvia Lithuania



Poland Russia

Figure 4.24. **(cont.)** Fractions of annual anthropogenic lead emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).



Sweden

Figure 4.24. **(cont.)** Fractions of annual anthropogenic lead emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

Table 4.2. Annual total anthropogenic emissions of **lead** of HELCOM countries and other EMEP countries in period 1990-2012, tonnes/year. (Expert estimates of emissions are shaded)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012
DK	127	104	96	55	24	25	25	21	24	31	19	18	18	19	21	17	16	13	14	13	13	12	12
EE	205	188	124	102	123	85	66	46	39	39	36	36	36	38	36	35	31	40	35	28	39	38	34
FI	338	258	185	109	68	67	45	29	32	25	45	45	46	38	28	22	25	22	20	18	23	22	19
DE	2057	1289	974	806	615	685	550	440	426	408	425	412	391	375	365	345	340	331	192	169	187	183	185
LV	96	81	76	73	68	60	58	52	47	6	6	7	7	7	7	4	4,3	4,6	4,3	3,9	4	3,7	3,7
LT	151	170	89	82	67	92	100	100	10,7	8,6	7,3	7,1	7,2	7,5	7,2	4,7	3,4	6	5,7	4,9	4,8	4,8	4,7
PL	642	642	642	642	642	642	660	639	571	561	524	511	510	533	545	529	562	551	543	496	561	544	554
RU	3 591	3 553	3 095	3 276	2 643	2 426	2 304	2 247	2 262	2 339	2 352	2 235	2 118	2 207	330	355	355	247	140	32	32	32	32
SE	358	313	293	144	52	38	34	34	34	31	27	24	21	20	18	15	15	15	13,1	13	13	12	11
Total	**7 564**	**6 600**	**5 574**	**5 289**	**4 302**	**4 120**	**3 843**	**3 609**	**3 446**	**3 448**	**3 441**	**3 297**	**3 152**	**3 244**	**1 358**	**1 326**	**1 352**	**1 229**	**967**	**778**	**876**	**851**	**855**
AL	62	63	62	67	72	67	68	70	74	68	76	85	93	80	110	60	3,9	3,8	4	3,7	3,7	3,7	3,7
AM	11	0,82	0,61	0,79	0,34	0,33	0,01	0,01	0,01	0,01	0,01	0,5	1	2,5	2,8	3	4,1	8,9	11	9,2	5,6	6,9	31
AT	219	180	125	87	60	16	16	14	13	12	12	12	12	12	13	14	14	14	15	13	15	15	15
AZ	12	12	12	12	12	12	12	12	12	12	12	13	13	13	13	14	14	14	14	15	15	15	15
BY	794	519	450	377	348	147	46	42	41	38	46	41	44	43	45	50	57	59	63	66	70	69	68
BE	237	217	217	186	142	172	181	201	124	140	106	89	83	79	88	75	72	63	73	34	43	32	32
BA	97	97	97	97	97	97	97	97	97	97	97	91	85	79	72	66	60	54	48	42	36	30	24
BG	321	189	212	285	290	333	334	255	301	292	267	248	209	248	144	127	128	118	225	112	106	119	117
HR	536	376	301	285	314	323	270	272	278	281	274	240	150	65	58	51	47	43	39	35	29	26	19
CY	36	37	40	41	43	42	43	43	43	43	44	42	42	42	33	29	29	30	31	31	31	31	28
CZ	269	240	247	232	202	180	165	180	169	157	108	47	47	39	37	47	43	44	39	40	26	17	23
FR	4591	3297	2520	2304	2054	1827	1617	1428	1283	1000	297	262	255	204	188	183	176	172	158	134	144	134	138
GE	6,9	6,9	6,9	6,9	6,9	6,9	6,9	6,9	6,9	6,9	6,9	7	7,2	7,3	7,5	7,6	7,8	8	8,1	8,3	8,4	8,6	8,7
GR	505	499	493	488	482	476	470	386	301	217	132	133	133	133	133	133	133	133	133	133	133	133	134
HU	623	488	208	187	155	129	100	90	82	34	35	34	35	33	36	37	37	35	36	32	17	9,4	7,9
IS	6,4	5,8	5,1	4,5	3,9	3,3	2,7	2,1	1,4	0,82	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2	0,2
IE	125	113	120	104	92	81	69	73	51	32	21	20	19	22	22	23	22	22	22	19	17	17	16
IT	4415	3362	2488	2304	2125	2029	1916	1739	1600	1436	945	712	251	256	271	281	289	312	301	228	260	269	258
KZ	256	256	256	256	256	256	256	256	256	256	256	260	264	268	271	275	279	283	287	290	294	298	302
LI	0,64	0,56	0,47	0,37	0,31	0,24	0,18	0,15	0,1	0,06	0,02	0,02	0,02	0,02	0,02	0,02	0,03	0,03	0,03	0,03	0,03	0,03	0,03
LU	77	71	65	59	53	30	26	18	6,8	2,3	1,8	2	1,9	1,9	1,9	1,9	1,9	1,9	1,9	1,9	1,9	1,9	1,9
MT	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,7	0,8	0,8	0,8	0,8	0,8	0,7	0,7	3,4	5,8	13
MC	3,8	3,9	4,1	3,7	2,1	0,77	0,66	0,54	0,47	0,41	0,06	0,06	0,06	0,05	0,04	0,04	0,03	0,04	0,04	0,04	0,04	0,04	0,04
NL	331	276	234	207	179	154	105	56	44	36	28	32	36	33	34	30	31	36	31	32	38	23	16
NO	187	144	128	88	25	23	12	11	11	9,7	8,4	7,6	7,9	7,9	8,4	7,2	6,9	7,6	6,7	4,6	4,7	5,2	4,7
PT	559	613	688	729	746	765	320	323	333	338	51	51	53	56	76	96	115	135	191	157	211	183	179
MD	249	220	103	71	23	34	28	22	7,9	11	2,8	3,4	3,3	10,6	2,3	5,1	5	2,2	3,3	2,3	1	2	2
RO	585	573	561	550	538	526	514	502	491	420	402	476	384	291	199	107	103	106	91	56	62	61	56
ME	309	443	329	182	129	141	237	203	222	221	142	107	60	63	63	48	46	52	47	45	46	3,6	3,6
RS	235	241	225	123	205	210	221	238	249	201	255	268	266	260	268	261	259	257	269	229	147	160	115
SK	99	98	97	88	79	79	79	77	74	62	89	70	65	64	70	73	74	62	63	43	59	59	58
SI	350	305	298	316	315	200	84	74	58	51	47	30	18	19	17	16	17	17	18	16	17	17	16
ES	2753	1843	1230	1120	1106	942	927	858	789	728	576	344	211	206	200	208	206	203	201	179	185	184	186
CH	369	344	311	269	236	179	155	140	126	63	43	39	34	31	29	28	28	27	27	26	26	25	24
MK	104	90	97	100	89	98	100	103	103	100	105	104	100	93	21	23	6,7	5,6	7,3	7,1	7,5	8,5	6,1
TR	765	765	765	765	765	765	765	765	765	765	765	717	669	620	572	524	476	428	380	332	284	235	187
UA	3 878	3 586	3 293	3 001	2 709	2 417	2 124	1 832	1 540	1 248	955	663	145	123	195	304	297	309	213	171	159	159	159
UK	**3 042**	**2 772**	**2 535**	**2 246**	**1 929**	**1 604**	**1 357**	**1 188**	**871**	**499**	**150**	**143**	**132**	**117**	**117**	**107**	**88**	**80**	**73**	**64**	**62**	**59**	**61**
EMEP	**34 584**	**28 948**	**24 398**	**22 533**	**20 187**	**18 485**	**16 568**	**15 189**	**13 874**	**12 328**	**9 798**	**8 689**	**7 082**	**6 867**	**4 777**	**4 643**	**4 530**	**4 378**	**4 100**	**3 387**	**3 447**	**3 279**	**3 184**

Expert estimates:

- *Denier van der Gon, H.A.C., M. van het Bolscher A.J.H. Visschedijk P.Y.J. Zandveld [2006] Study to the effectiveness of the UNECE Persistent Organic Pollutants Protocol and costs of possible additional measures Phase I: Estimation of emission reduction resulting from the implementation of the POP Protocol, TNO report B&O-A R 2005/194*
- *Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H., Zandveld P.Y.J. [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03*

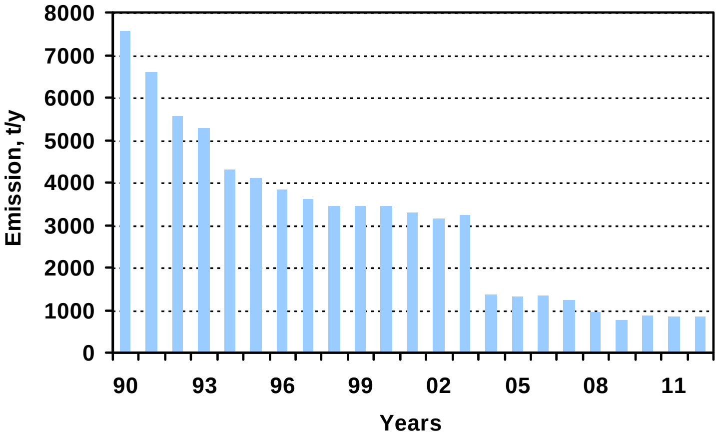


Figure 4.25. Time-series of total annual **lead** emissions of HELCOM countries in 1990-2012, tonnes/year.

4.2 Annual total deposition of lead

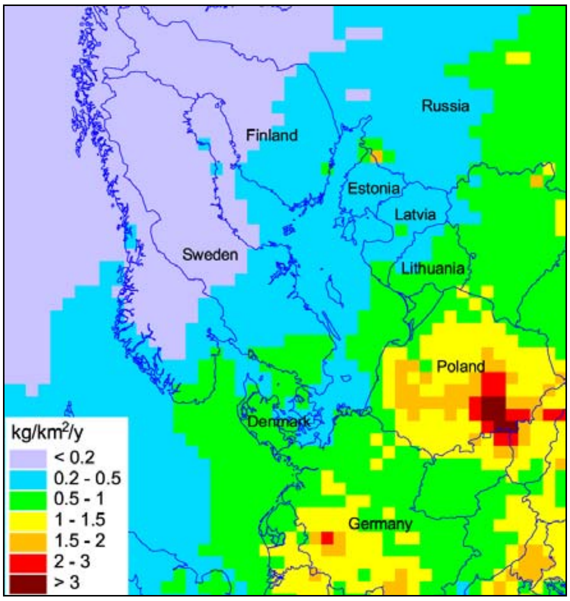


Figure 4.26. Annual total deposition fluxes of **lead** over the Baltic Sea region for 2012, kg/km^2/y.

4.3 Monthly total deposition of lead

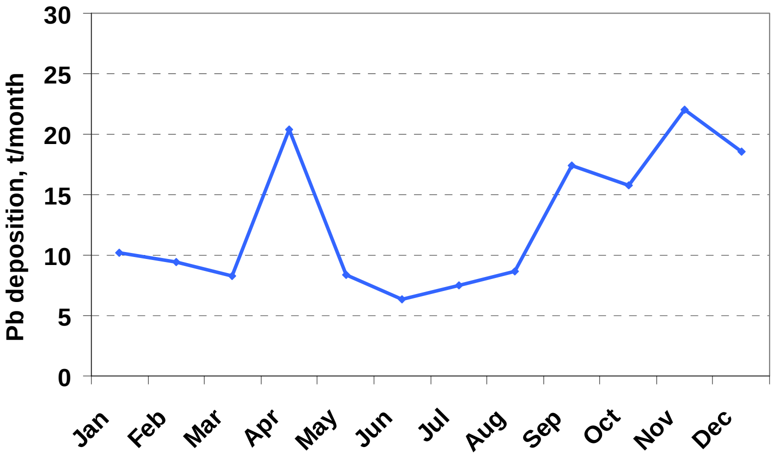


Figure 4.27. Monthly total deposition of **lead** to the Baltic Sea for 2012, tonnes/month.

Table 4.3. Monthly total deposition of **lead** to the Baltic Sea for 2012, tonnes/month.

Month	**Deposition**
Jan	10.2
Feb	9.5
Mar	8.3
Apr	20.4
May	8.3
Jun	6.4
Jul	7.5
Aug	8.6
Sep	17.4
Oct	15.8
Nov	22.0
Dec	18.5

4.4 Source allocation of lead deposition

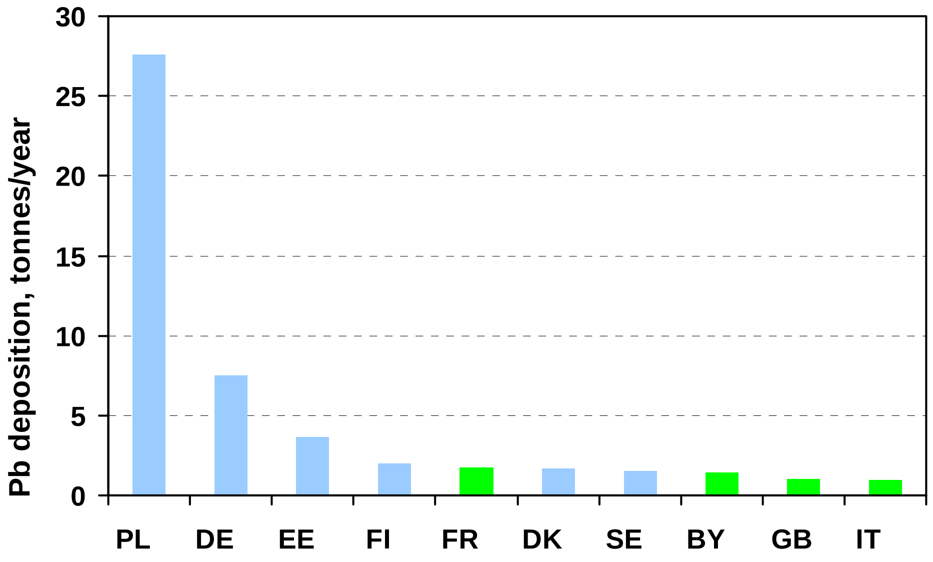


Figure 4.28. Top ten countries with the highest contribution to annual total deposition of lead into the Baltic Sea for 2012, tonnes/year.

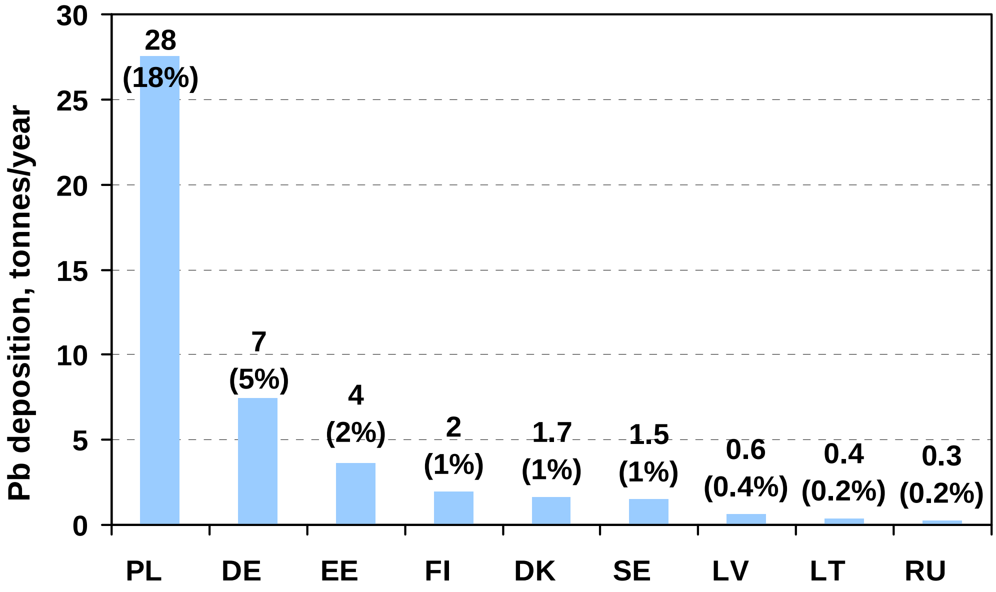


Figure 4.29. Sorted contributions (in tones/year and in %) of HELCOM countries to total deposition to the Baltic Sea for 2012. HELCOM countries emissions of lead contributed about 30% to the total annual lead deposition over the Baltic Sea. Contribution of other EMEP countries accounted for 8%. Significant contribution was made by other emission sources, in particular, remote emissions sources, natural emissions and wind re-suspension of **lead** (62%).

Table 4.4. Two most significant contributors to annual total deposition of **lead** to the nine Baltic Sea sub-basins for 2012.

Sub-basin	Country(1)	%	Country(2)	%	*, %
ARC	Poland	16	Finland	5	62
BOB	Poland	10	Finland	8	63
BOS	Poland	15	Finland	3	65
BAP	Poland	23	Germany	5	61
GUF	Estonia	23	Poland	10	50
GUR	Poland	18	Germany	3	61
KAT	Poland	7	Germany	6	74
SOU	Poland	10	Germany	9	66
WEB	Germany	11	Poland	7	70
BAS	Poland	18	Germany	5	62

* - contribution of re-emission, natural and remote sources.

4.5 Comparison of model results with measurements

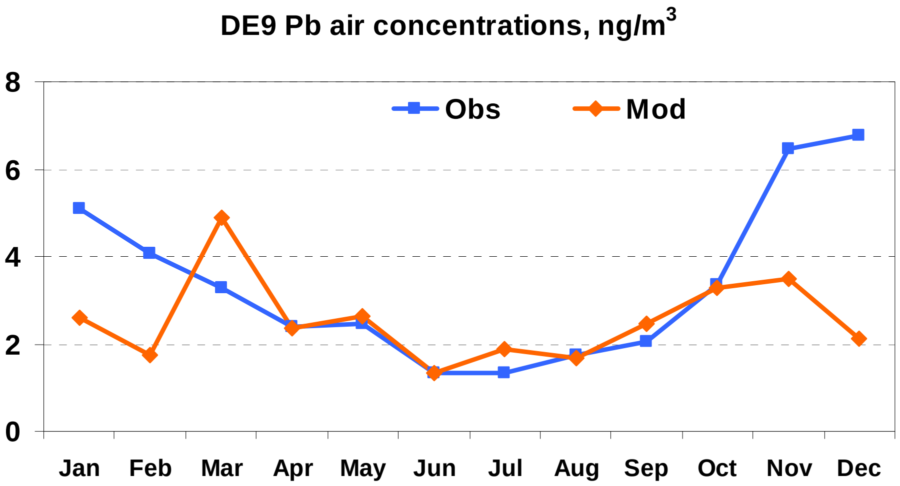


Figure 4.30. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Zingst (DE9). Units: ng / m^3.

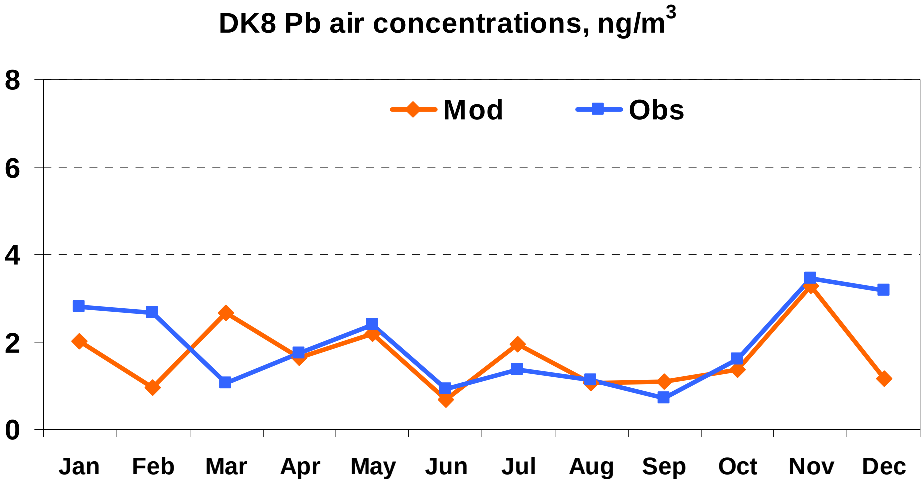


Figure 4.31. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Anholt (DK8). Units: ng / m^3.

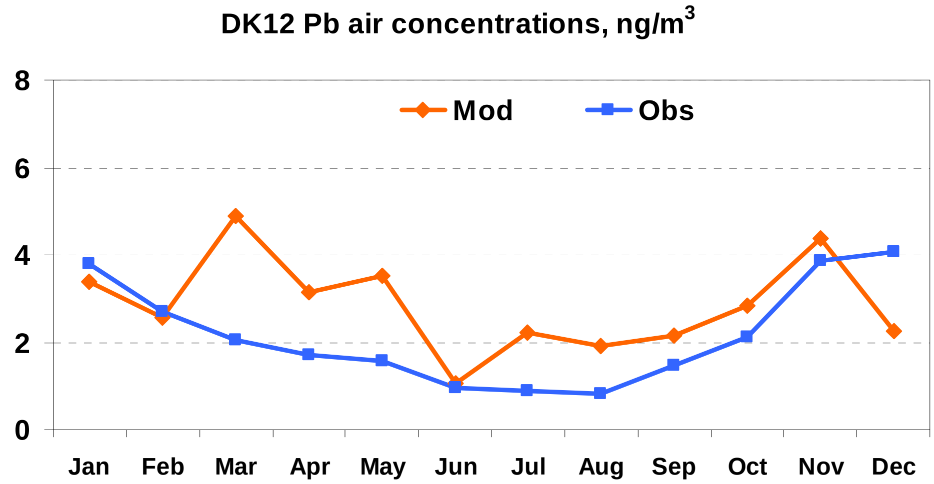


Figure 4.32. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Risoe (DK12). Units: ng / m^3.

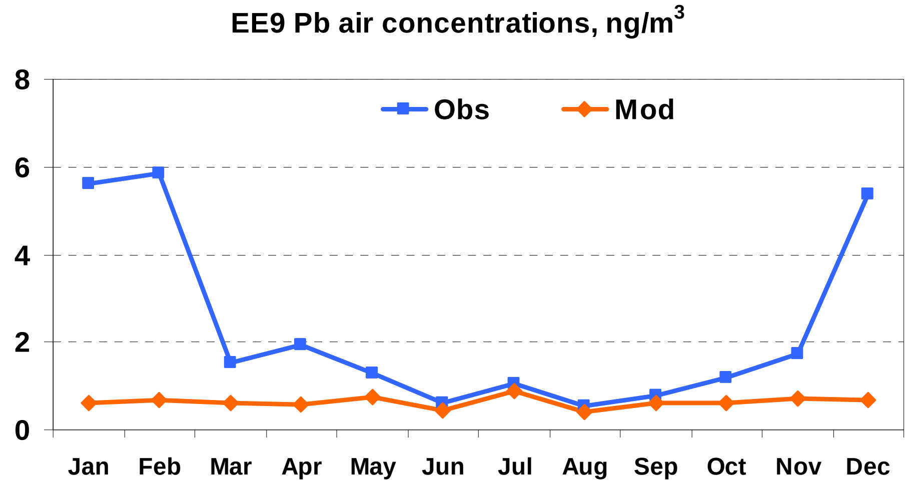


Figure 4.33. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Lahemaa (EE9). Units: ng / m^3.

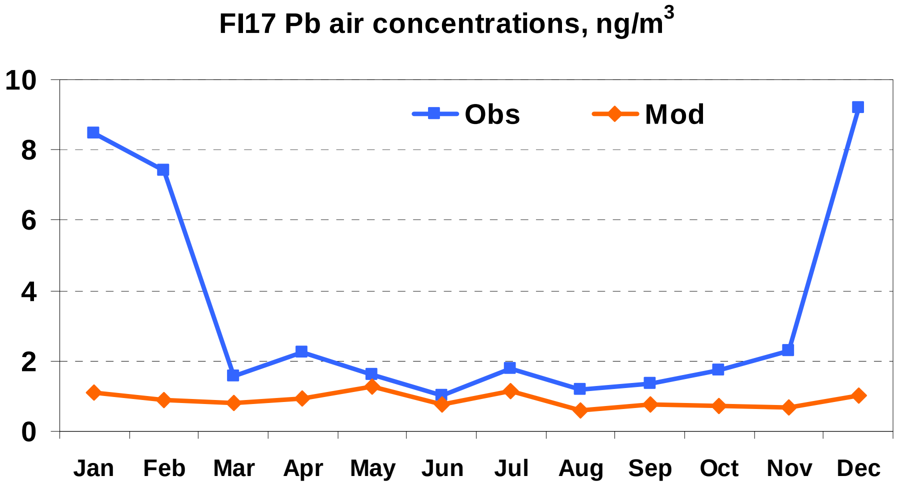


Figure 4.34. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Virolahti II (FI17). Units: ng / m^3.

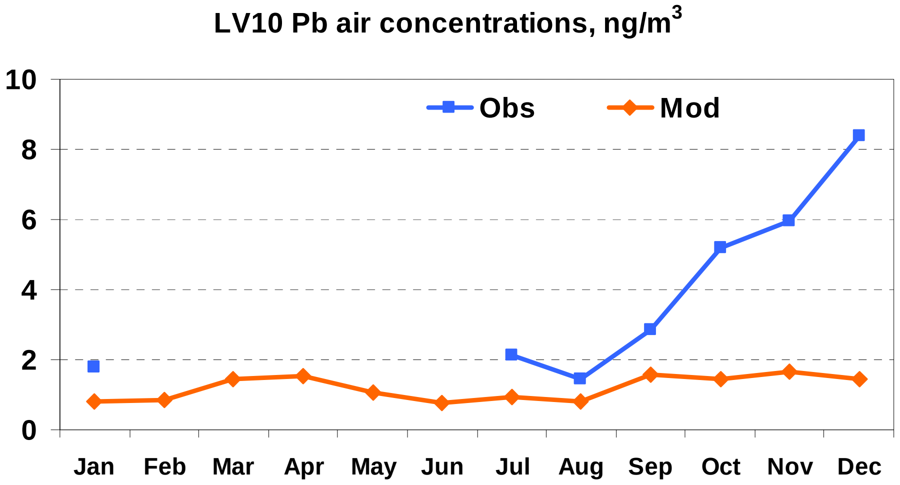


Figure 4.35. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Rucava (LV10). Units: ng / m^3.

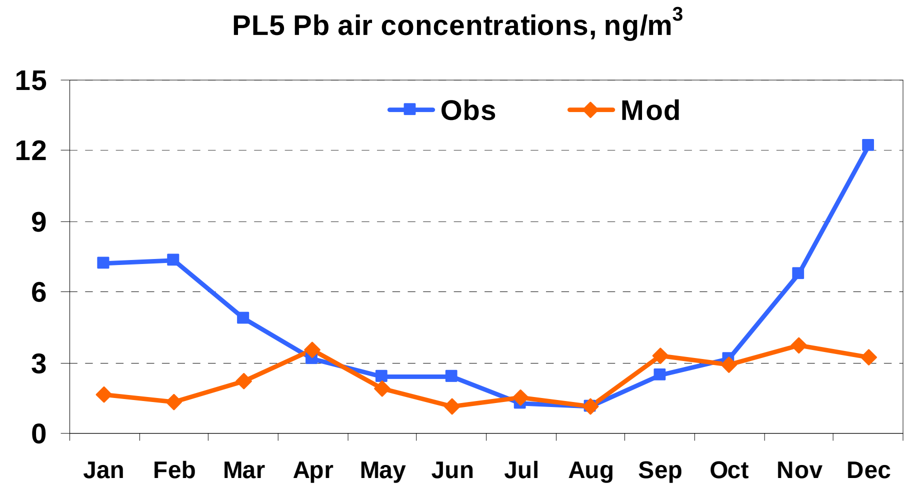


Figure 4.36. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Diabla Gora (PL5). Units: ng / m^3.

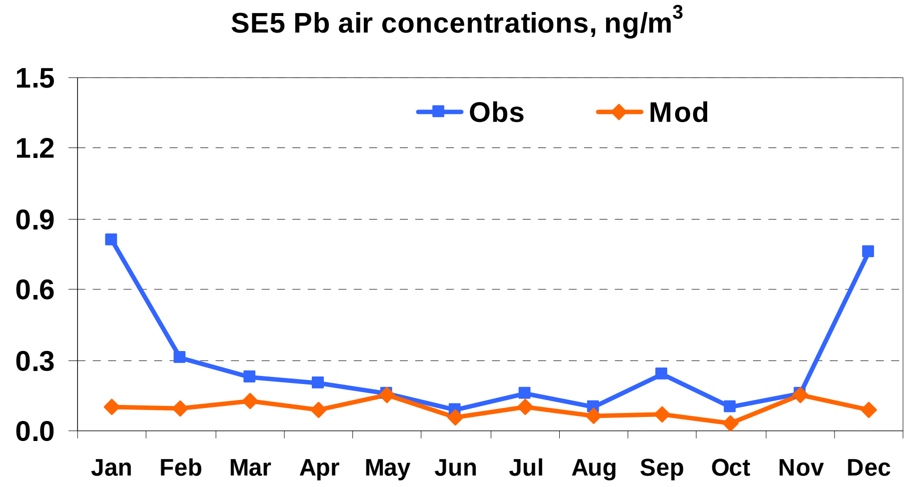


Figure 4.37. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Bredkälen (SE5). Units: ng / m^3.

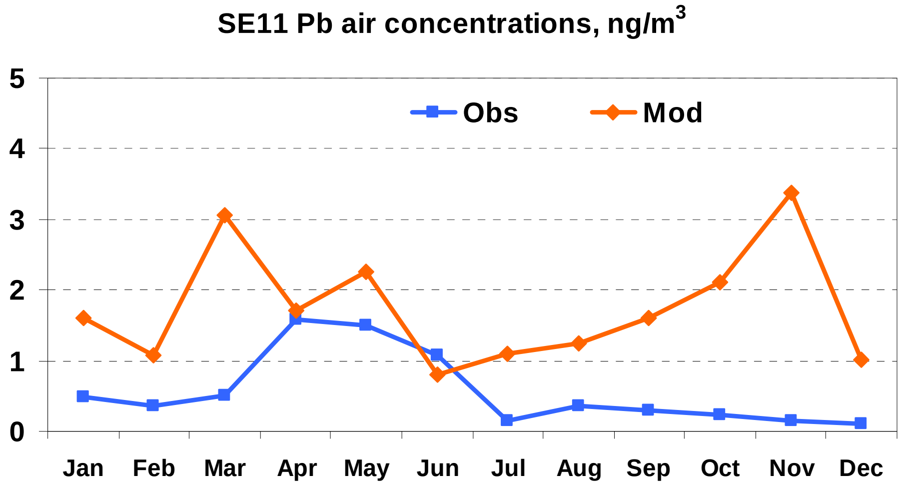


Figure 4.38. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Vavihill (SE11). Units: ng / m^3.

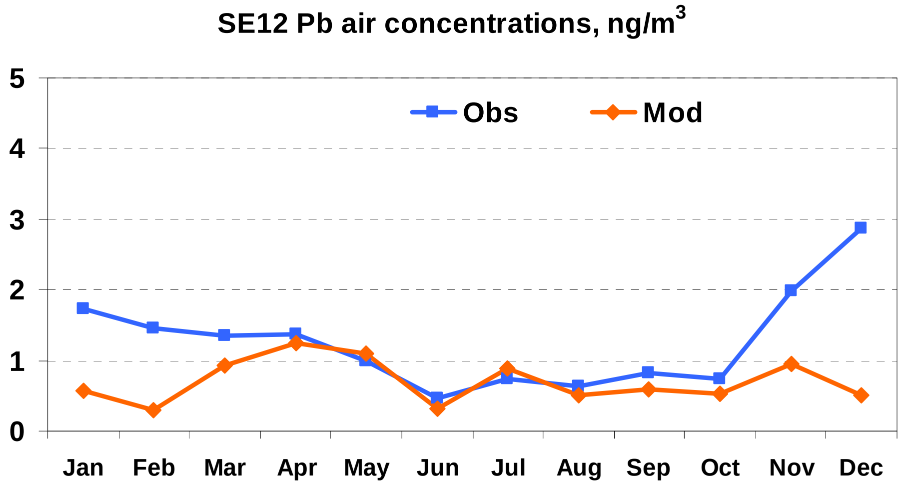


Figure 4.39. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Aspvreten (SE12). Units: ng / m^3.

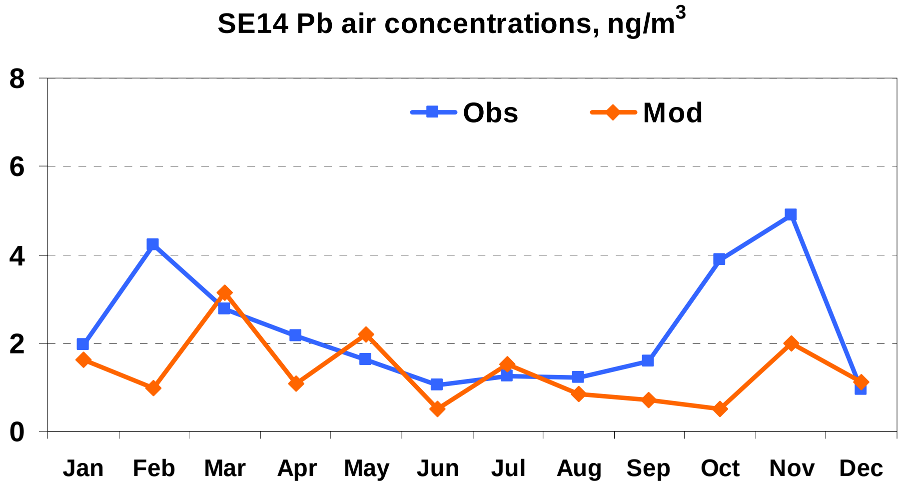


Figure 4.40. Comparison of calculated mean monthly lead concentrations in air for 2012 with measurements of the station Räo (SE14). Units: ng / m^3.

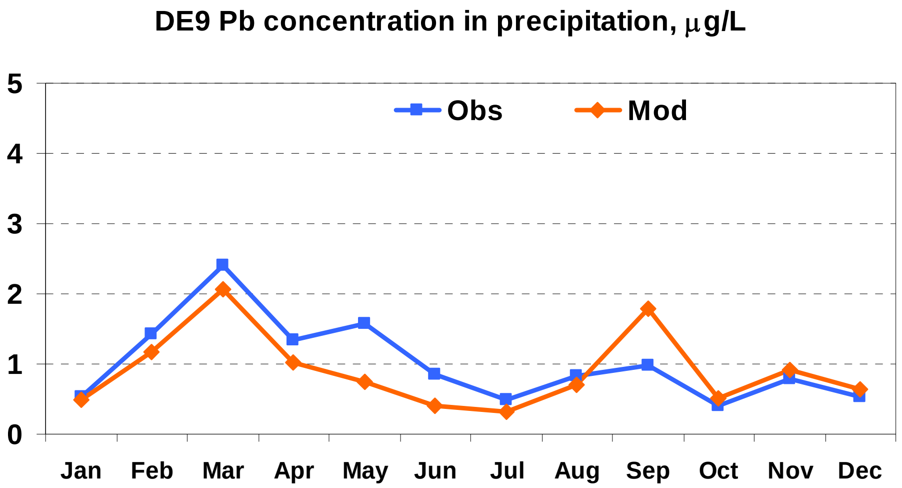


Figure 4.41. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Zingst (DE9). Units: µg / L.

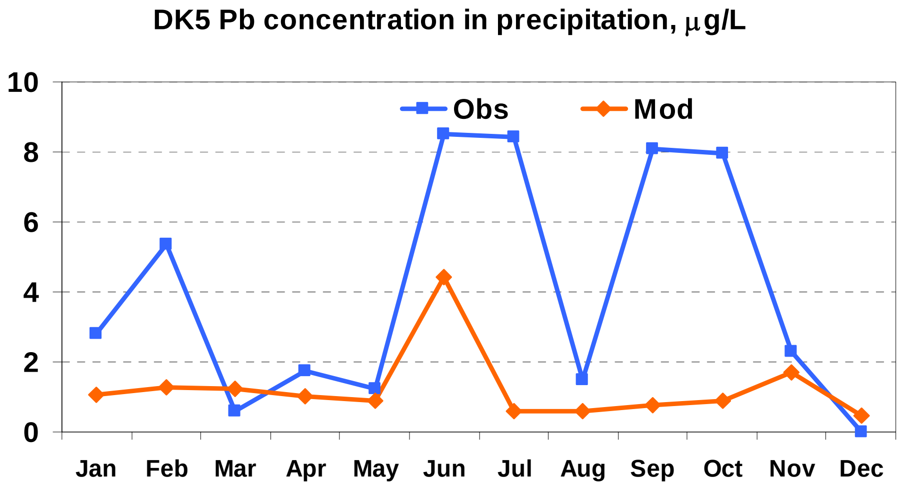


Figure 4.42. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Keldsnor (DK5). Units: µg / L.

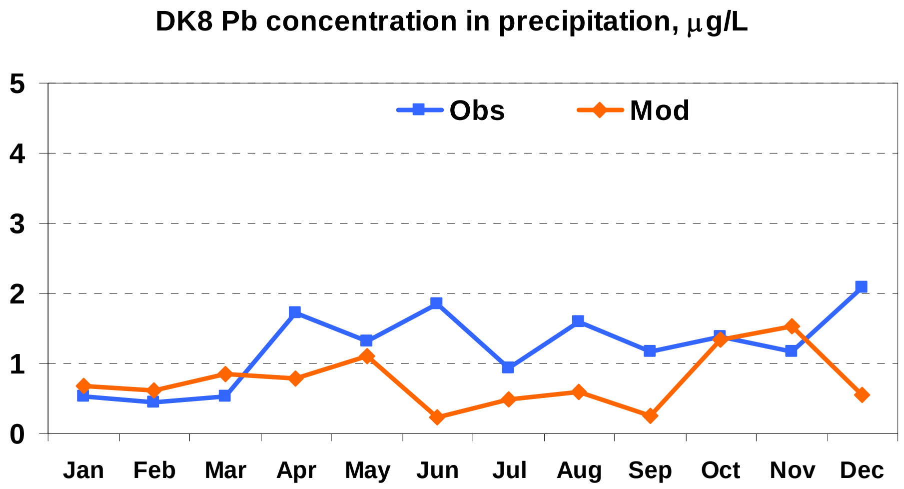


Figure 4.43. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Anholt (DK8). Units: µg / L.

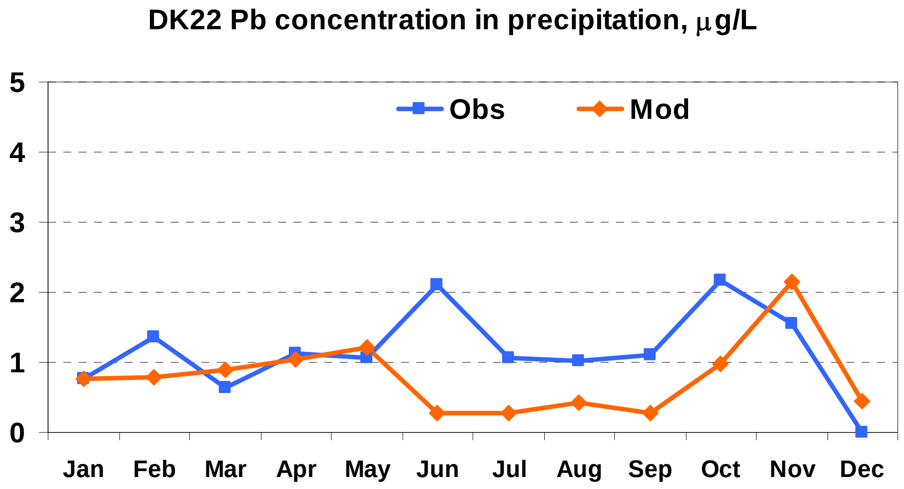


Figure 4.44. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Storebaelt (DK22). Units: µg / L.

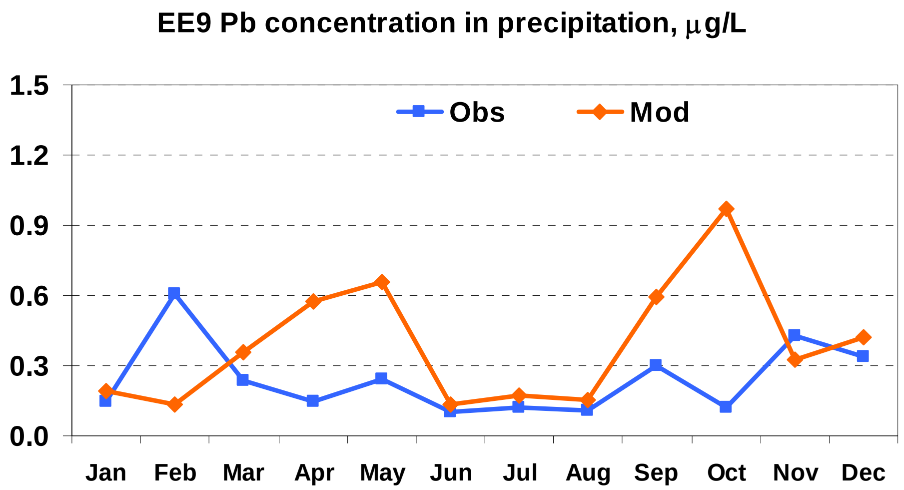


Figure 4.45. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Lahemaa (EE9). Units: µg / L.

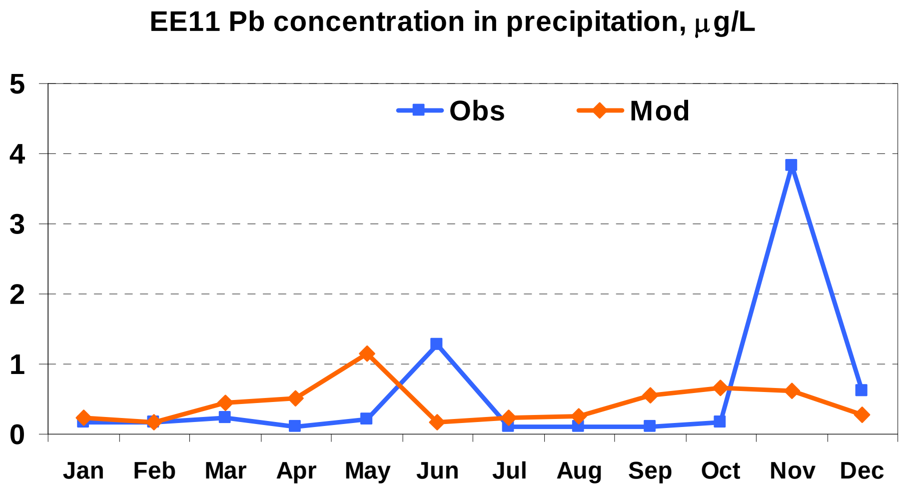


Figure 4.46. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Vilsandi (EE11). Units: µg / L.

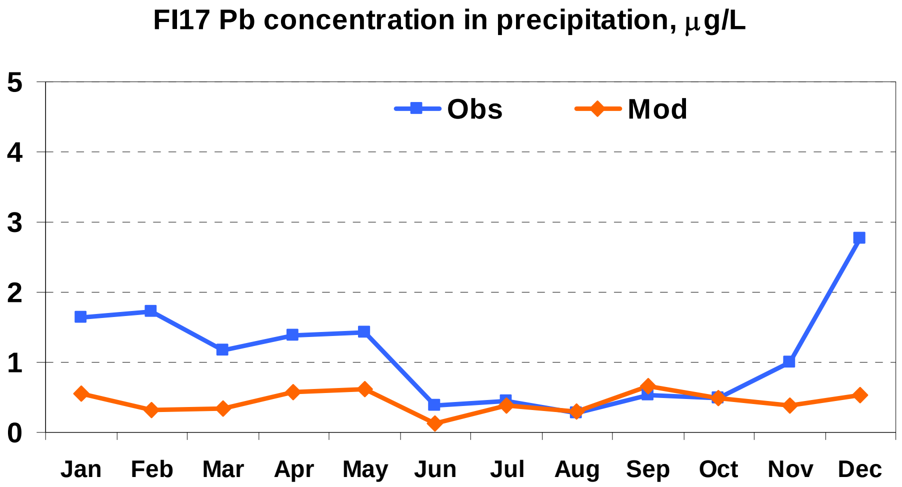


Figure 4.47. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Virolahty II (FI17). Units: µg / L.

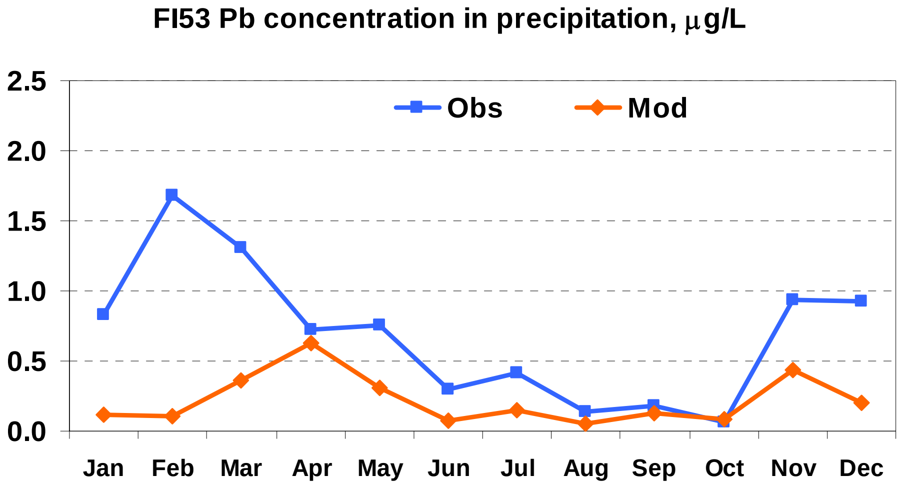


Figure 4.48. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Hailuoto (FI53). Units: µg / L.

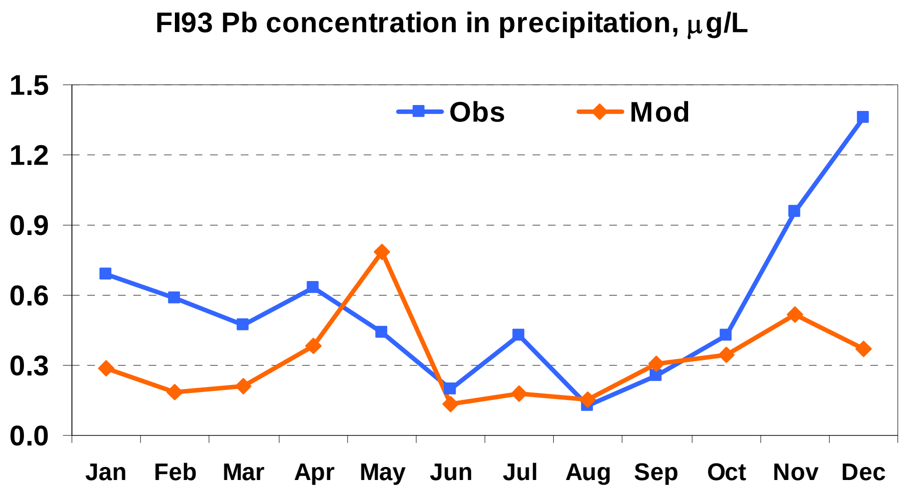


Figure 4.49. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Kotinen (FI93). Units: μg / L.

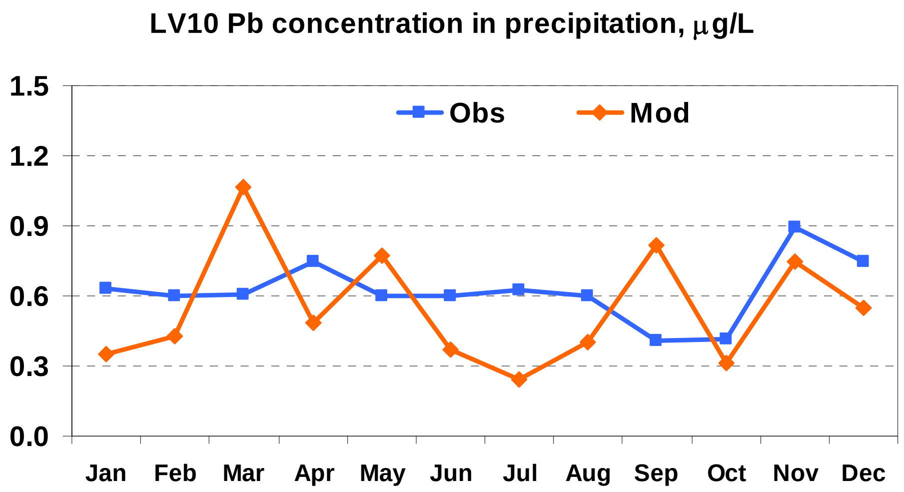


Figure 4.50. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Rucava (LV10). Units: μg / L.

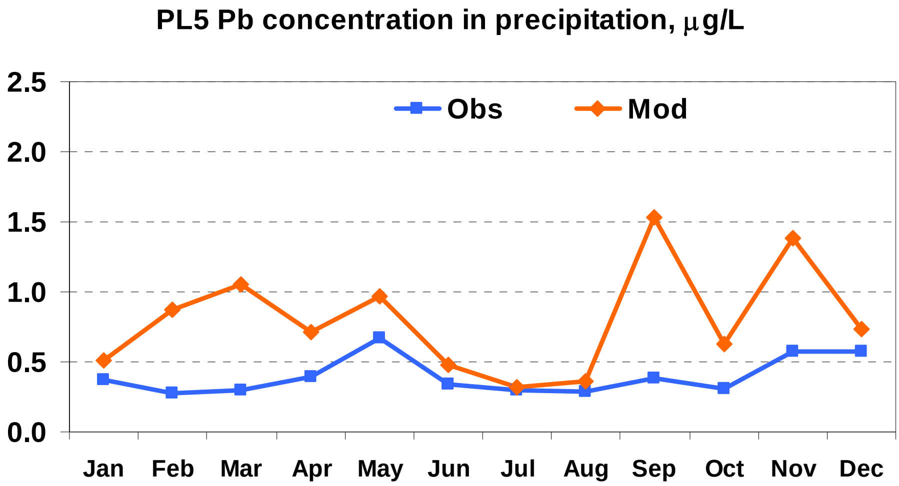


Figure 4.51. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Diable Gora (PL5). Units: µg / L.

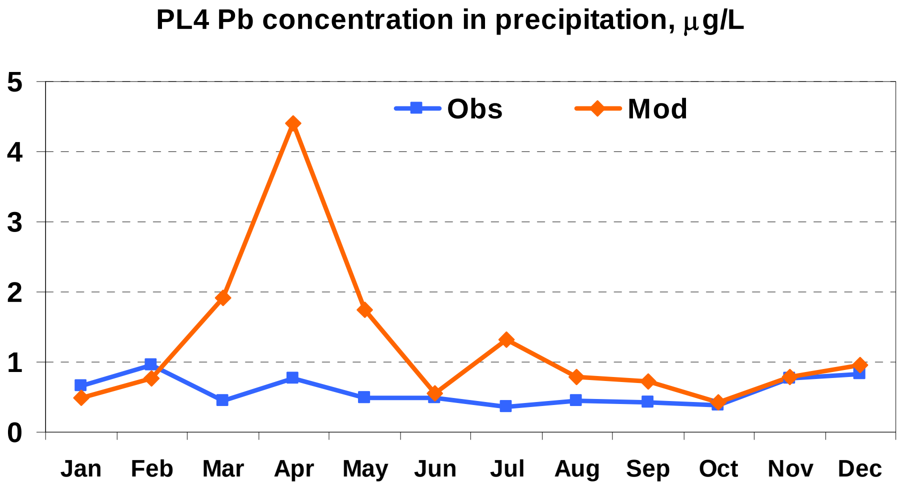


Figure 4.52. Comparison of calculated mean monthly lead concentrations in precipitation for 2012 with measurements of the station Leba (PL4). Units: µg / L.

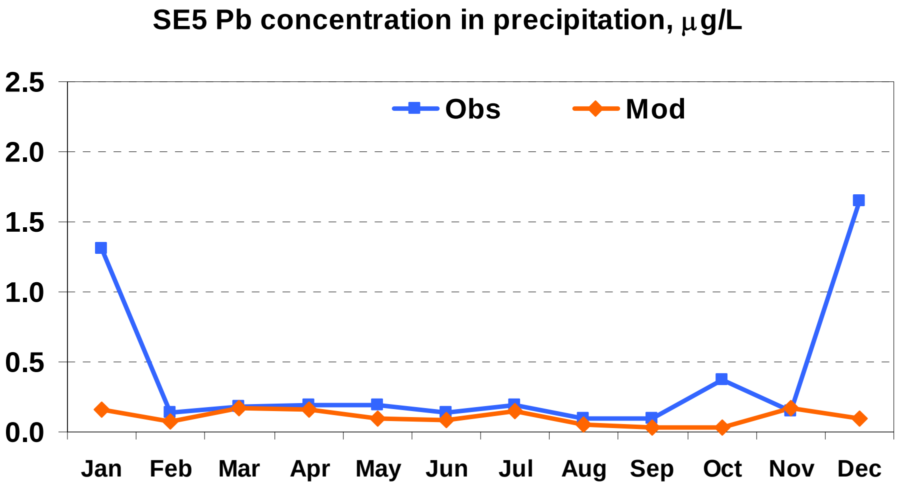


Figure 4.53. Comparison of calculated mean monthly lead concentrations in precipitation with measured at station Bredkälen (SE5). Units: µg / L.

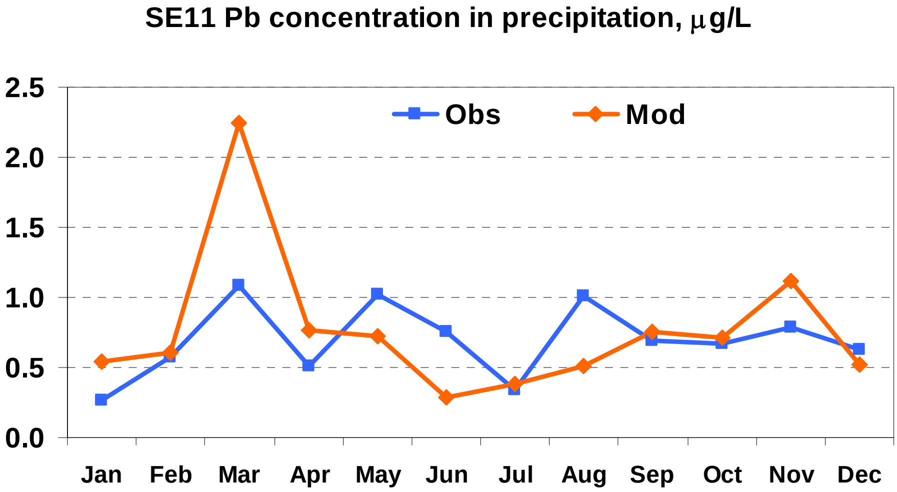


Figure 4.54. Comparison of calculated mean monthly lead concentrations in precipitation with measured at station Vavihill (SE11). Units: µg / L.

In general, computed concentrations of lead in air and in precipitation obtained for the selected monitoring sites around the Baltic Sea reasonably agree with the measured concentrations. Some deviations between the simulated and observed monthly mean concentrations of lead can likely be explained by the uncertainties in seasonal variation of lead emission used in modeling (underestimation of winter time emissions), differences between measured precipitation amount and the one used in the model, and difficulties in measurements of heavy metals.

4.6 Concluding remarks

- Emissions of lead from HELCOM countries have decreased from 1990 to 2012 by 89%. Lead emission in HELCOM countries have slightely increased from 2011 to 2012 by 0.4%.

- Annual deposition of lead to the Baltic Sea has dropped from 1990 to 2012 by 79%. From 2011 to 2012 the deposition of lead has decreased by 12%.

- The contribution of anthropogenic sources of HELCOM countries to total lead deposition over the Baltic Sea was estimated to 30%. Essential contribution belongs also to the anthropogenic sources of other EMEP countries (8%), natural sources and wind re-suspension (62%).

- The most significant contribution among the HELCOM countries to lead deposition over the Baltic Sea was made by Poland (18%) followed by Germany (5%).

- Modelling results for lead were generally within an accuracy of a factor of two in comparison with annual mean measured concentrations around the Baltic Sea in 2012.

5. Atmospheric Supply of Cadmium to the Baltic Sea in 2012

In this chapter the results of model evaluation of cadmium atmospheric input to the Baltic Sea and its sub-basins for 2012 is presented. Modelling of cadmium atmospheric transport and deposition was carried out using MSC-E Eulerian Heavy Metal transport model MSCE-HM (*Travnikov and Ilyin*, 2005). Latest available official information on cadmium emission from HELCOM countries and other European countries was used in computations. Based on these data annual and monthly levels of cadmium deposition to the Baltic Sea region have been obtained and contributions of HELCOM countries emission sources to the deposition over the Baltic Sea are estimated. Model results were compared with observed levels of cadmium concentrations in air and precipitation measured at monitoring sites around the Baltic Sea in 2012.

5.1 Cadmium emissions

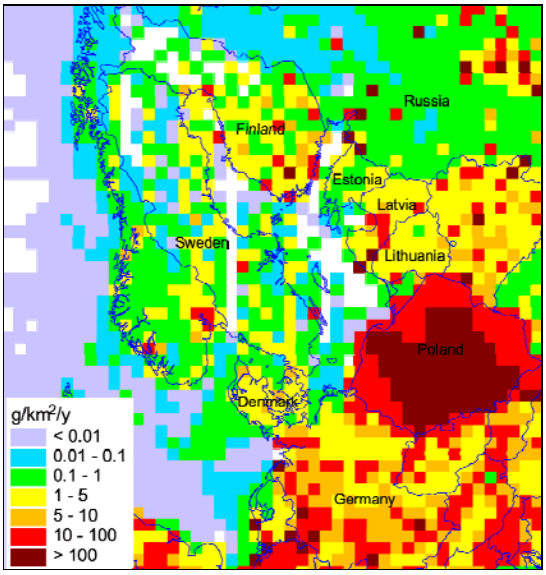


Figure 5.1. Annual total anthropogenic emissions of cadmium in the Baltic Sea region for 2012, g/km^2/y.

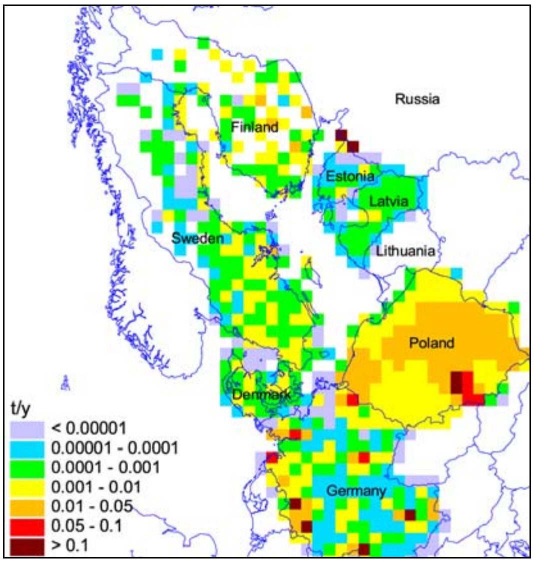


Figure 5.2. Annual cadmium emission from Public Power sector for 2012, t/grid cell/y (white color means no information).

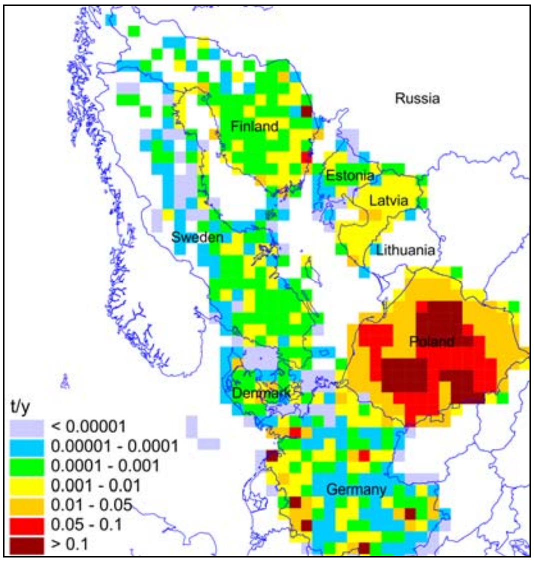


Figure 5.3. Annual cadmium emission from Industrial Combustion sector for 2012, t/grid cell/y (white color means no information).

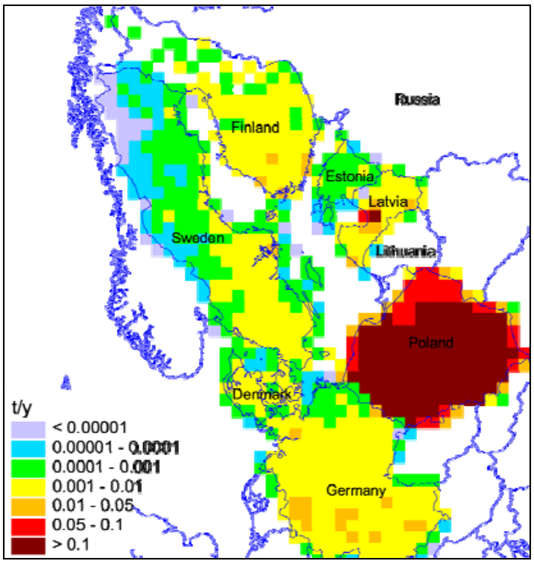


Figure 5.4. Annual cadmium emission from Small Combustion sector for 2012, t/grid cell/y (white color means no information).

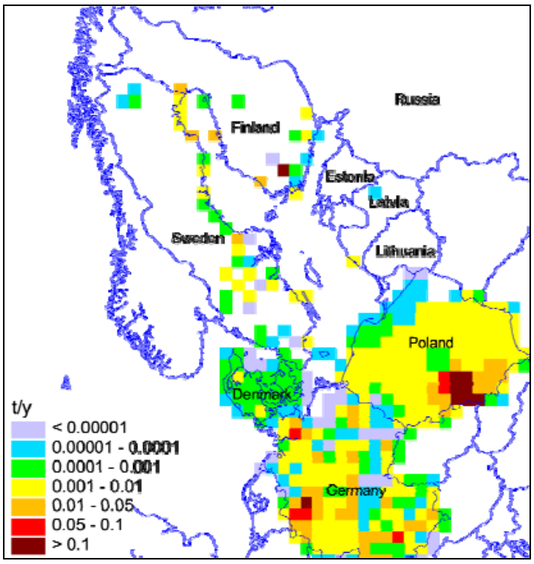


Figure 5.5. Annual cadmium emission from Industrial Processes sector for 2012, t/grid cell/y (white color means no information).

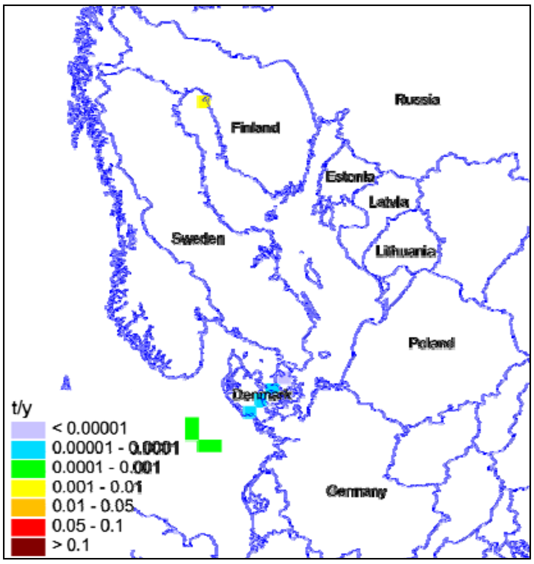


Figure 5.6. Annual cadmium emission from Fugitive Emissions sector for 2012, t/grid cell/y (white color means no information).

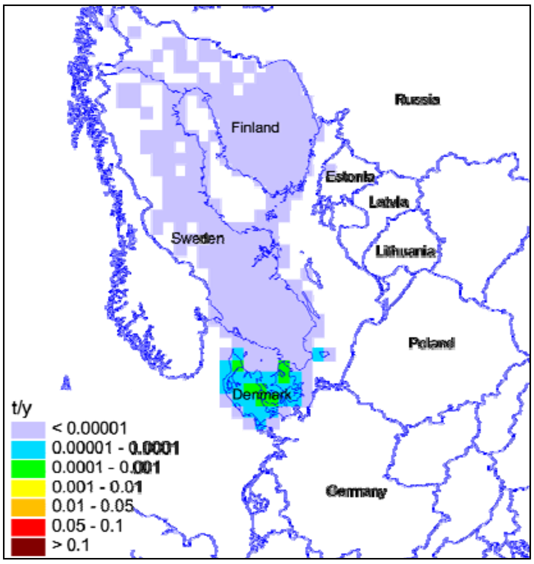


Figure 5.7. Annual cadmium emission from Solvents sector for 2012, t/grid cell/y (white color means no information).

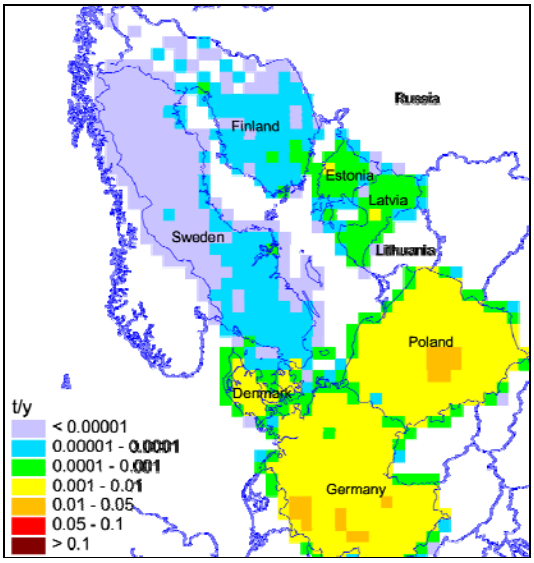


Figure 5.8. Annual cadmium emission from Road Rail sector for 2012, t/grid cell/y (white color means no information).

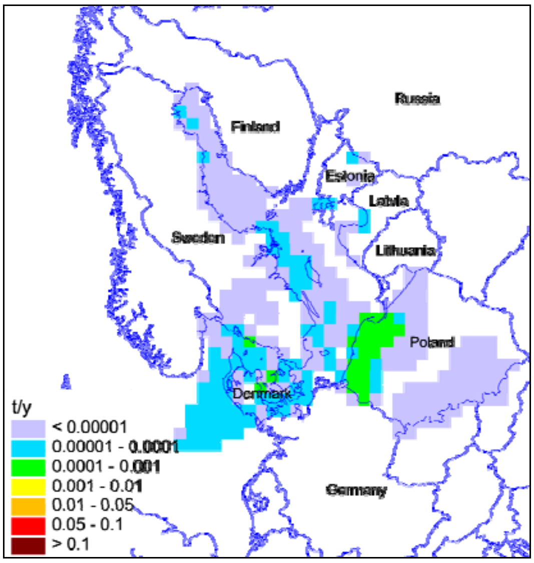
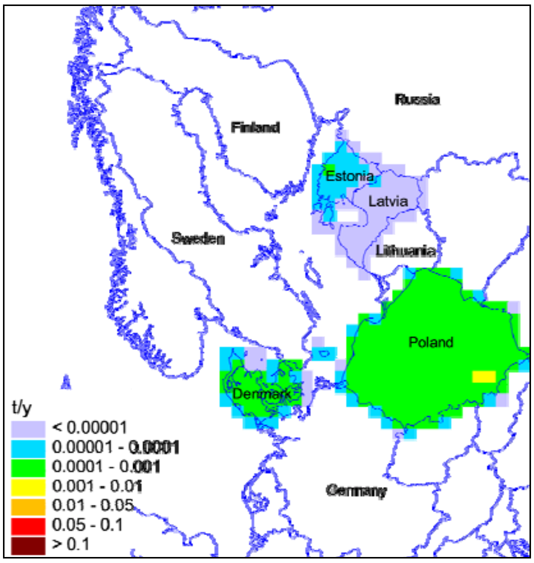


Figure 5.9. Annual cadmium emission from Shipping Emissions sector for 2012, t/grid cell/y (white color means no information).



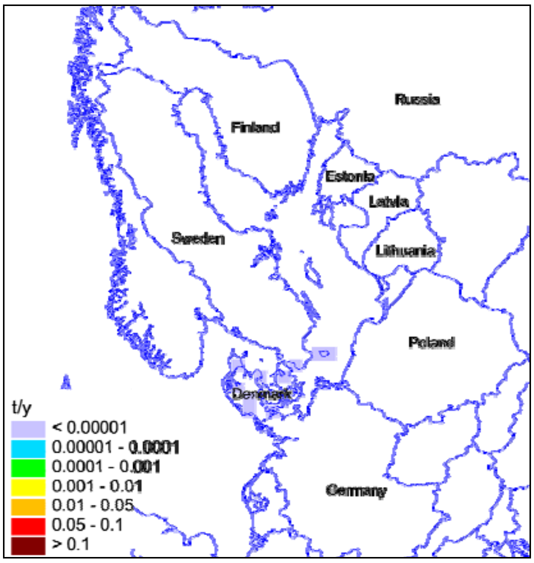
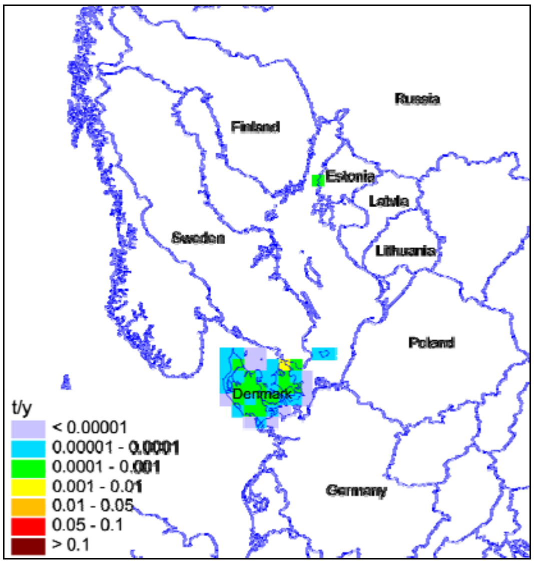


Figure 5.10. Annual cadmium emission from Off Road Mobility sector for 2012, t/grid cell/y (white color means no information).

Figure 5.11. Annual cadmium emission from Civil Aviation sector for 2012, t/grid cell/y (white color means no information).



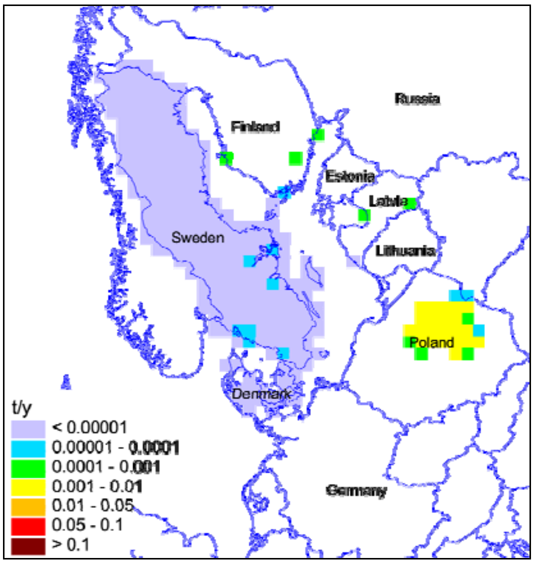


Figure 5.12. Annual cadmium emission from Other Waste Displacement sector for 2012, t/grid cell/y (white color means no information).

Figure 5.13. Annual cadmium emission from Waste Incineration sector for 2012, t/grid cell/y (white color means no information).

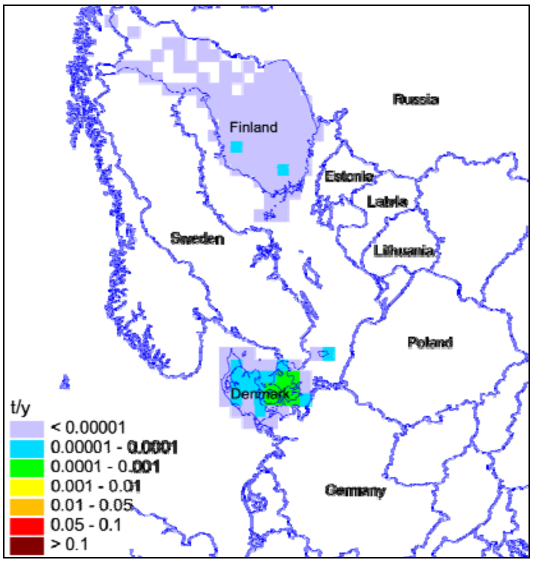


Figure 5.14. Annual cadmium emission from Agricultural Wastes sector for 2012, t/grid cell/y (white color means no information).

Table 5.1. Annual total anthropogenic emissions of **cadmium** of HELCOM countries from different sectors for 2012, tonnes/year

GNFR emission sector	Sector name	Denmark	Estonia	Finland	Germany	Latvia	Lithuania	Poland	Russia	Sweden
A	Public Power	0.027	0.532	0.175	1.206	0.011	0.045	2.483	0.130	0.154
B	Industrial Combustion	0.047	0.016	0.473	1.362	0.175	0.038	9.976	0.009	0.084
C	Small Combustion	0.043	0.02	0.335	0.799	0.435	0.399	23.045		0.171
D	Industrial Processes	0.018		0.343	1.41	0.003	0.005	2.054	0.09	0.107
E	Fugitive Emissions	0.002	2.7E-07	0.002	NA			0.445		
F	Solvents	0.003	1.0E-07	1.1E-06	NA	NA		4.7E-06		4.2E-07
G	Road Rail	0.046	0.008	0.004	0.765	0.010	0.013	0.470	20.4	0.003
H	Shipping Emissions	0.004	5.9E-05		0.003	7.0E-05	4.8E-05	0.005		0.001
I	Off Road Mobility	0.007	0.001	NA	0.025	1.9E-05	0.011	0.084		3.8E-05
J	Civil Aviation	1.6E-05	NA	NA	NE	NE	NE	NA		NE
L	Other Waste Disposal	4.9E-03	2.6E-04	NA	NA	NA		NA		
M	Waste water	NA	NA	NA	NA	NO	NE	NA	0.003	NA
N	Waste Incineration	1.4E-05		0.001	1.4E-06	3.7E-04	0.044	0.152	1.0E-06	0.001
Q	Agricultural waste	0.002	NE	2.9E-04	NO	NA	NO	NA		NO
R	Other	NO	NO	NO	NO	NA	NO	NO	1.968	NO
Total		0.20	0.58	1.33	5.57	0.63	0.55	38.71	22.60	0.52

NO – not occurring, an activity or process does not exist within a country.
NA – not applicable, the process or activity exists but emissions are considered never to occur.
NE – not estimated, emissions occur but have not been estimated or reported in this submission.
IE – included elsewhere, emissions by sources of compounds are estimated but included elsewhere in the inventory.

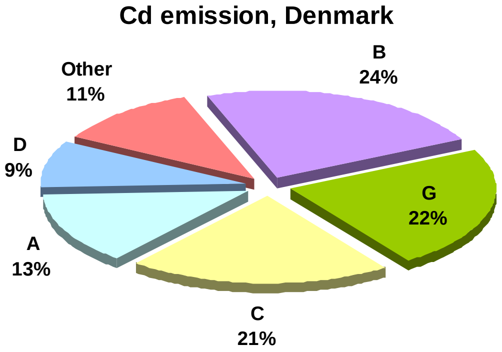


Figure 5.15. Contributions of different sectors to total annual cadmium emission of Denmark in 2012.

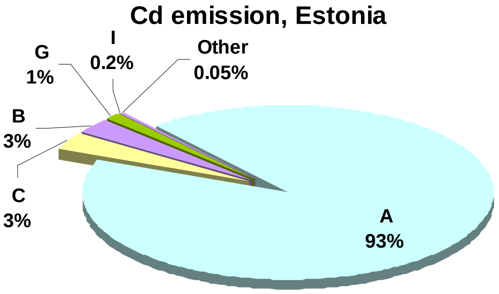


Figure 5.16. Contributions of different sectors to total annual cadmium emission of Estonia in 2012.

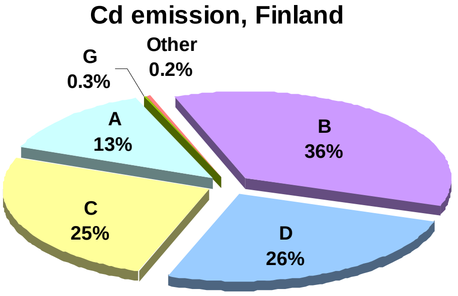


Figure 5.17. Contributions of different sectors to total annual cadmium emission of Finland in 2012.

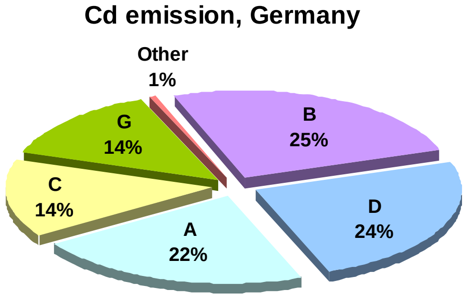
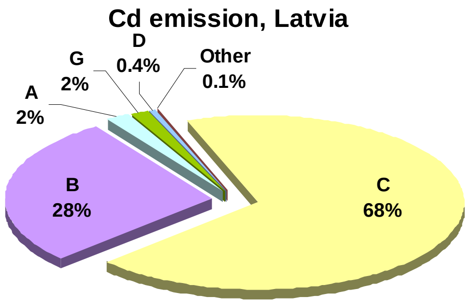


Figure 5.18. Contributions of different sectors to total annual cadmium emission of Germany in 2012.



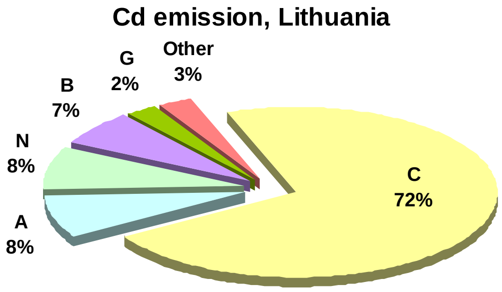
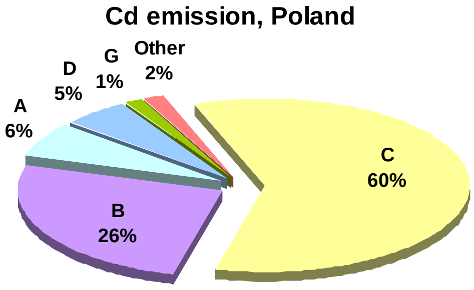


Figure 5.19. Contributions of different sectors to total annual cadmium emission of Latvia in 2012.

Figure 5.20. Contributions of different sectors to total annual cadmium emission of Lithuania in 2012.



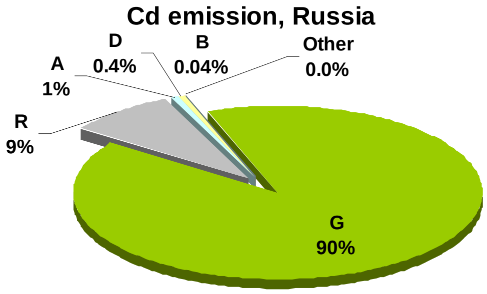


Figure 5.21. Contributions of different sectors to total annual cadmium emission of Poland in 2012.

Figure 5.22. Contributions of different sectors to total annual cadmium emission of Russia in 2012.

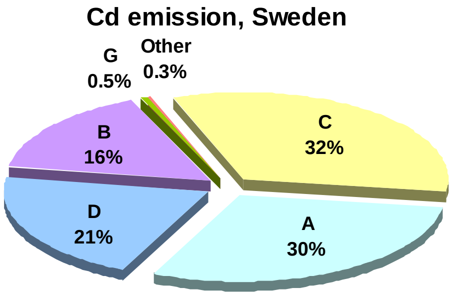
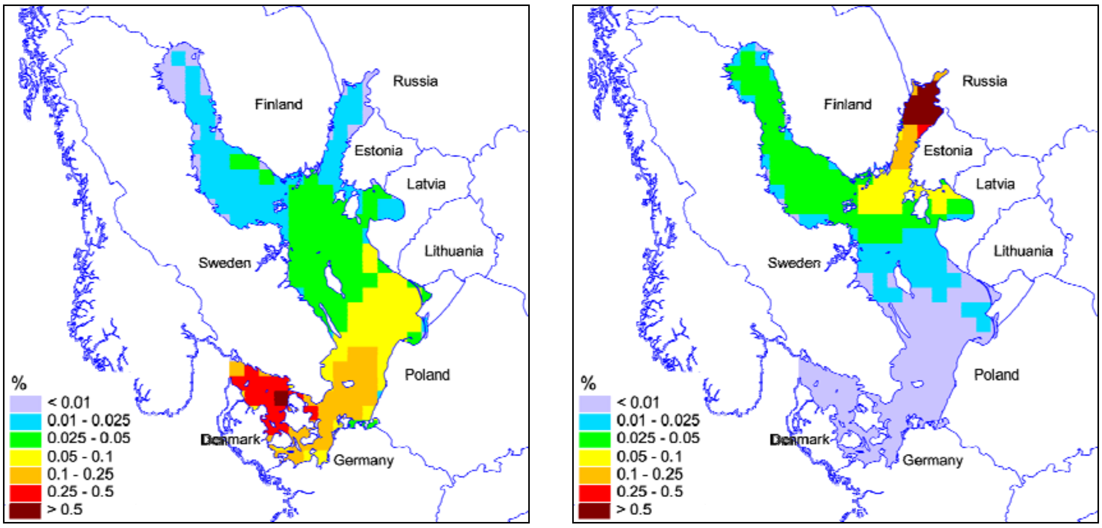
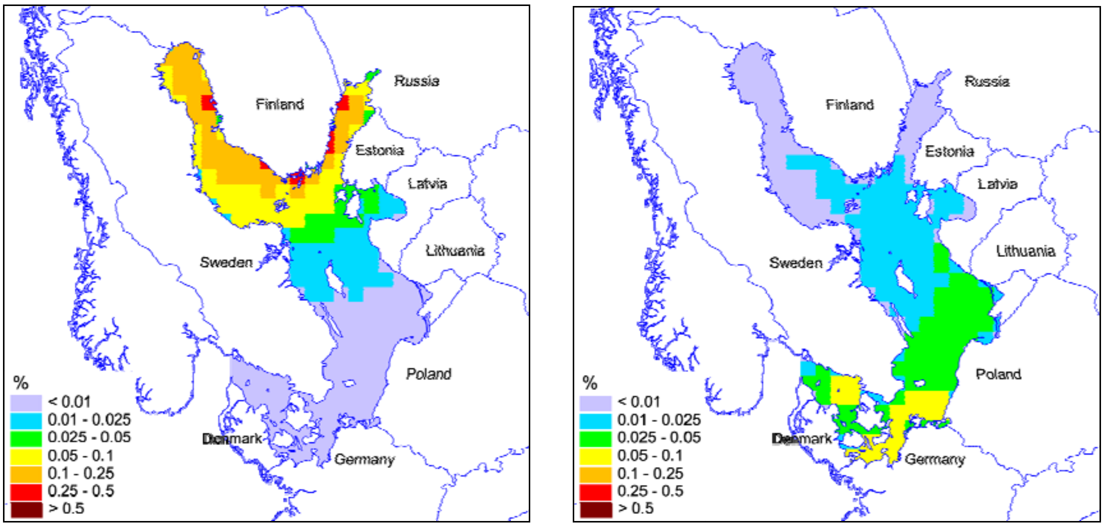


Figure 5.23. Contributions of different sectors to total annual cadmium emission of Sweden in 2012.

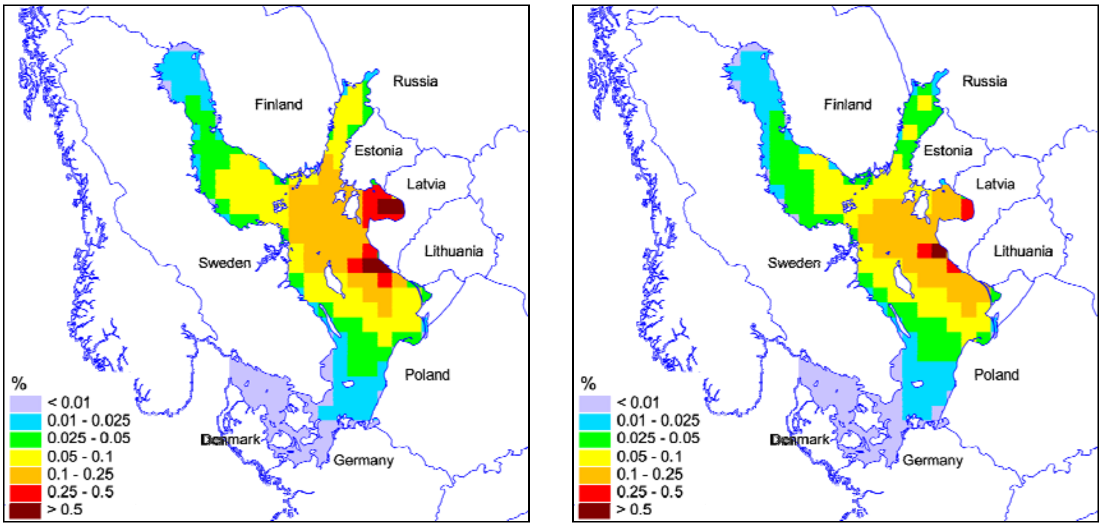


Denmark Estonia

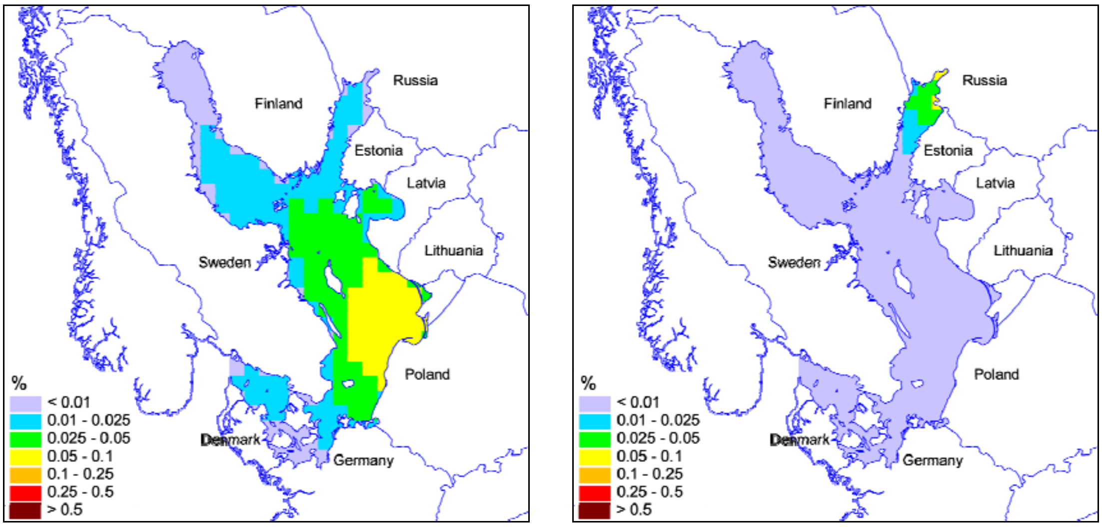


Finland Germany

Figure 5.24. Fractions of annual anthropogenic cadmium emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

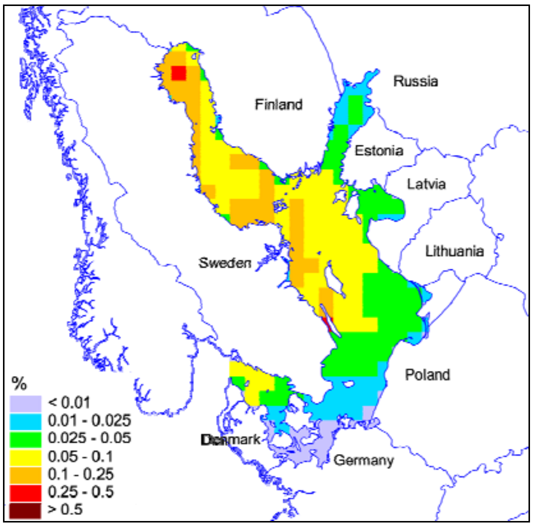


Latvia Lithuania



Poland Russia

Figure 5.24. **(cont.)** Fractions of annual anthropogenic cadmium emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).



Sweden

Figure 5.24. **(cont.)** Fractions of annual anthropogenic cadmium emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

Table 5.2. Annual total anthropogenic emissions of **cadmium** of HELCOM countries and other EMEP countries in period 1990-2012, tonnes/year (Expert estimates of emissions are shaded)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012
DK	0,99	1	0,85	0,76	0,63	0,5	0,52	0,44	0,42	0,38	0,36	0,4	0,34	0,34	0,32	0,27	0,23	0,24	0,24	0,21	0,21	0,21	0,2
EE	4,4	4,2	3	2,2	2,9	2	1	1,1	1	0,95	0,56	0,55	0,57	0,63	0,59	0,58	0,55	0,68	0,61	0,48	0,67	0,65	0,58
FI	6,3	3,5	3	2,8	2,2	1,7	1,5	0,86	1,3	0,57	1,3	1,7	1,3	1,2	1,5	1,3	1,3	1	1,2	1,2	1,4	1,3	1,3
DE	17	14	11	9,4	8,8	12	11	11	11	11	10	10	9,4	9	8,2	7,5	7,2	6,6	5,7	5	5,6	5,5	5,6
LV	0,47	0,51	0,5	0,53	0,54	0,59	0,62	0,58	0,57	0,55	0,51	0,56	0,55	0,57	0,61	0,61	0,61	0,6	0,57	0,65	0,63	0,58	0,63
LT	0,6	0,6	0,4	0,5	0,4	0,5	0,5	0,5	0,5	0,48	0,48	0,51	0,51	0,51	0,53	0,58	0,56	0,53	0,51	0,51	0,52	0,53	0,55
PL	58	58	58	58	58	58	60	54	44	42	36	36	34	34	35	35	38	36	37	35	42	38	39
RU	79	68	69	59	57	57	51	50	49	51	51	51	52	57	55	59	59	47	35	23	23	23	23
SE	2,3	1,7	1,4	1,1	0,75	0,73	0,7	0,69	0,61	0,53	0,51	0,59	0,51	0,51	0,53	0,53	0,55	0,55	0,51	0,54	0,54	0,52	0,52
Total	**170**	**151**	**147**	**134**	**131**	**133**	**127**	**120**	**109**	**107**	**100**	**101**	**99**	**104**	**103**	**106**	**108**	**94**	**81**	**66**	**74**	**70**	**71**
AL	0,31	0,15	0,08	0,08	0,06	0,08	0,06	0,03	0,05	0,05	0,04	0,04	0,04	0,05	0,05	0,06	0,06	0,07	0,08	0,12	0,12	0,12	0,12
AM	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21
AT	1,6	1,5	1,3	1,2	1,1	0,98	1	0,97	0,9	0,95	0,92	0,95	0,94	0,97	0,98	1,1	1,1	1,1	1,1	1,1	1,2	1,2	1,2
AZ	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,3	2,4	2,4	2,5	2,5	2,6	2,6	2,7	2,7	2,8	2,8	2,9
BY	2,1	2,2	2	1,7	1,3	1,1	1,2	1,3	1,5	1,4	1,4	1,8	1,9	1,8	1,8	2,1	2,5	2,6	2,8	3	3,2	3	2,9
BE	6,6	6,4	7,1	5,9	4,4	5,4	4,5	4,6	3,1	2,8	3,2	2,9	2,9	3	3,4	2,9	3	2,8	3,1	2,4	3	2,7	2,5
BA	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,7	1,6	1,6	1,6	1,6	1,6	1,6	1,6	1,6	1,6	1,6
BG	5,2	4,1	3,3	3,6	3,6	3,6	3,8	3,6	3,4	3	3,5	3,4	3,1	3,4	3,4	2,9	2,8	2,5	3,2	2,2	1,9	2,1	2
HR	1,3	0,88	0,67	0,7	0,87	0,92	0,85	0,73	0,83	0,75	0,65	0,6	0,6	0,64	0,62	0,6	0,58	0,51	0,47	0,45	0,42	0,42	0,38
CY	0,05	0,05	0,06	0,06	0,07	0,06	0,06	0,07	0,07	0,07	0,08	0,08	0,08	0,08	0,08	0,09	0,08	0,08	0,09	0,08	0,07	0,07	0,08
CZ	4,3	3,9	3,6	3,5	3,5	3,6	2,9	3	2,7	2,7	2,9	2,6	2,7	2,2	2,4	3,1	3,2	2,9	3,8	3,4	0,88	0,81	0,95
FR	20	20	20	19	19	18	17	16	15	14	14	13	12	8,8	6,3	6	4,5	4,1	4,1	2,8	2,7	2,6	2,5
GE	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,21	0,22	0,22	0,23	0,23	0,24	0,24	0,25	0,25	0,26	0,26	0,27	0,28
GR	4,5	4,2	4	3,7	3,5	3,2	3	3	2,9	2,9	2,8	2,8	2,8	2,7	2,7	2,6	2,6	2,5	2,5	2,4	2,4	2,3	2,3
HU	5,2	4,7	4	4,1	4,1	3,9	3,4	3,3	3,1	3,3	3,2	3,2	3	3	3	3,5	3,4	3,2	3,3	3,4	0,7	0,7	0,5
IS	0,17	0,16	0,15	0,14	0,13	0,12	0,12	0,11	0,1	0,09	0,08	0,08	0,08	0,08	0,08	0,08	0,08	0,08	0,08	0,08	0,09	0,09	0,09
IE	0,85	0,87	0,88	0,87	0,96	0,96	0,92	1,01	1,04	1,03	1,09	0,97	0,77	0,72	0,73	0,76	0,68	0,64	0,6	0,46	0,45	0,34	0,33
IT	10	11	10	9,8	9,4	9,4	9,1	9,1	8,6	8,5	8,8	8,7	7	7,3	7,8	8,1	8,3	8,9	8,7	7	6,8	7,1	6,7
KZ	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,7	5,8	5,9	6	6,1	6,2	6,3	6,3	6,4	6,5	6,6	6,7
LI	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,002	0,003	0,003	0,002	0,002	0,002	0,002	0,002	0,002
LU	0,6	0,58	0,55	0,53	0,5	0,4	0,4	0,3	0,2	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
MT	0,48	0,48	0,48	0,48	0,48	0,48	0,48	0,48	0,48	0,48	0,48	0,53	0,52	0,57	0,57	0,59	0,6	0,62	0,56	0,56	0,04	0,02	0,02
MC	0,06	0,06	0,06	0,07	0,006	0,006	0,007	0,008	0,007	0,007	0,008	0,008	0,007	0,006	0,005	0,005	0,004	0,005	0,005	0,005	0,004	0,005	0,005
NL	2,1	1,8	1,6	1,4	1,2	1,1	0,9	1	1,1	0,93	0,92	1,6	2,2	2,3	1,7	1,7	1,9	1,7	1,9	1,8	2,5	1,1	0,8
NO	1,2	1,1	1,1	1,2	1,2	1	1,1	1,1	1,2	1,1	0,78	0,77	0,77	0,75	0,68	0,64	0,69	0,63	0,61	0,49	0,58	0,58	0,48
PT	6	6,4	6,6	5,8	6,1	6,2	5,4	5,9	6,6	6,5	6	5,9	6,6	5,8	5,6	6,5	5,5	5,7	5,4	3,7	4,1	2,8	2,4
MD	2,4	3,5	1,7	1,4	0,82	0,59	0,66	0,36	0,33	0,15	0,17	0,11	0,23	0,12	0,11	0,15	0,16	0,07	0,16	0,1	0,1	0,12	0,12
RO	22	20	19	18	17	15	14	13	12	12	8,7	7,4	6,3	5,3	4,2	3,1	3,1	3,6	3,1	2	2,2	2,3	2
ME	0,08	0,08	0,06	0,05	0,04	0,02	0,06	0,06	0,07	0,06	0,06	0,05	0,06	0,06	0,07	0,06	0,07	0,07	0,09	0,05	0,06	0,07	0,07
RS	4,4	3,7	3,7	1,6	2	2,2	3	3,3	3,1	1,8	2	2	2,1	1,8	1,8	2	2,2	2,1	2,2	2,1	2,1	2,1	2
SK	9,7	10	11	8,7	6,6	10	9	10	7,6	6,5	9,1	7,4	5	5,8	6,8	6,2	6,1	1,4	1,4	1,1	1,4	1,3	1,3
SI	0,59	0,47	0,48	0,44	0,35	0,38	0,38	0,36	0,37	0,37	0,38	0,4	0,39	0,4	0,41	0,4	0,43	0,44	0,45	0,4	0,42	0,41	0,38
ES	25	24	23	21	22	23	21	21	21	21	19	17	18	17	16	16	15	12	11	10	9,8	9,2	9,1
CH	3,8	3,6	3,5	3,3	3	2,7	2,6	2,5	2,4	2	1,9	1,7	1,5	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,3	1,3
MK	0,41	0,42	0,38	0,33	0,29	0,38	0,44	0,38	0,38	0,36	0,4	0,25	0,25	0,22	0,15	0,15	0,16	0,14	0,17	0,17	0,18	0,19	0,16
TR	17	17	17	17	17	17	17	17	17	17	17	17	17	17	17	17	17	18	18	18	18	18	18
UA	54	50	46	42	38	34	30	26	22	18	14	10	2	28	3,1	6,8	5,1	9	8,3	4,4	2,8	2,8	2,8
UK	23	22	22	14	13	11	9	8,3	6,1	5,9	5,8	4,5	4,5	3,3	3,4	3,6	3,5	2,8	2,6	2,3	2,4	2,6	2,1
EMEP	**414**	**388**	**372**	**335**	**321**	**320**	**301**	**288**	**264**	**253**	**239**	**230**	**215**	**239**	**210**	**217**	**215**	**195**	**183**	**155**	**158**	**150**	**148**

Expert estimates:

Denier van der Gon, H.A.C., M. van het Bolscher A.J.H. Visschedijk P.Y.J. Zandveld [2006] Study to the effectiveness of the UNECE Persistent Organic Pollutants Protocol and costs of possible additional measures Phase I: Estimation of emission reduction resulting from the implementation of the POP Protocol, TNO report B&O-A R 2005/194

Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H., Zandveld P.Y.J. [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03

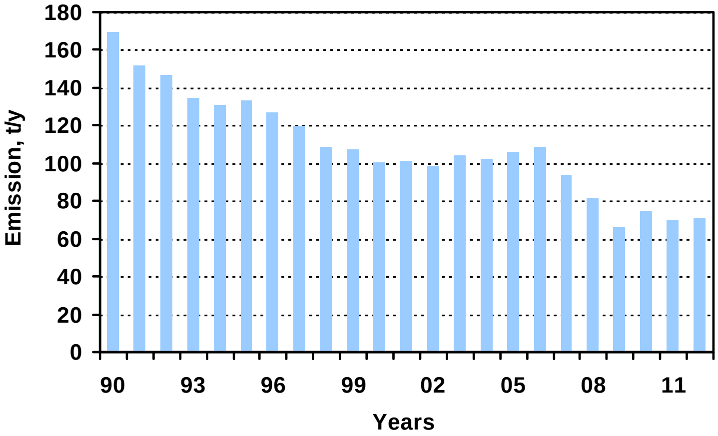


Figure 5.25. Time-series of annual **cadmium** emissions of HELCOM countries in 1990-2012, tonnes/year.

5.2 Annual total deposition of cadmium

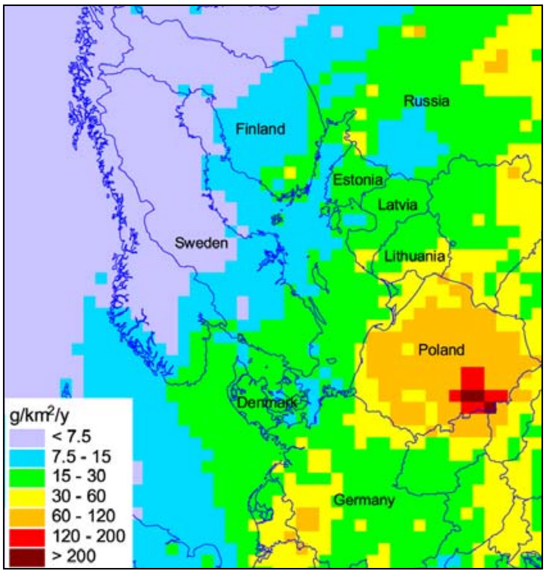


Figure 5.26. Annual total deposition fluxes of **cadmium** over the Baltic Sea region for 2012, g/km^2/y.

5.3 Monthly total deposition of cadmium

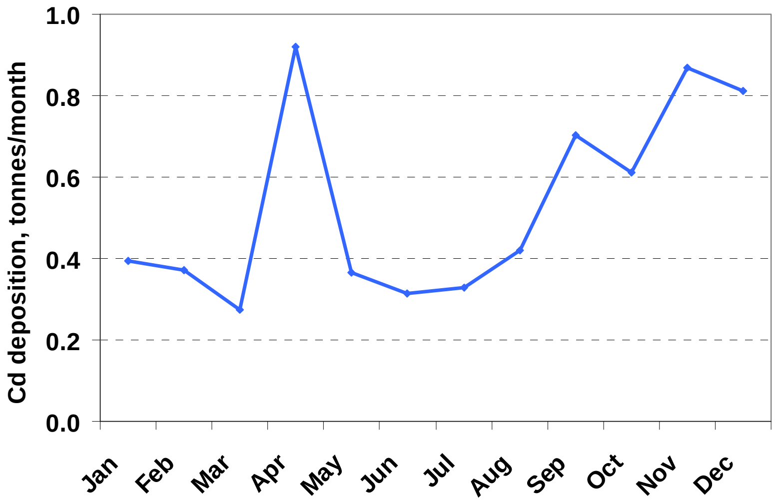


Figure 5.27. Monthly total deposition of **cadmium** to the Baltic Sea for 2012, tonnes/month.

Table 5.3. Monthly total deposition of **cadmium** to the Baltic Sea for 2012, tonnes/month.

Month	**Cd**
Jan	0.39
Feb	0.37
Mar	0.27
Apr	0.92
May	0.36
Jun	0.31
Jul	0.33
Aug	0.42
Sep	0.70
Oct	0.61
Nov	0.87
Dec	0.81

5.4 Source allocation of cadmium deposition

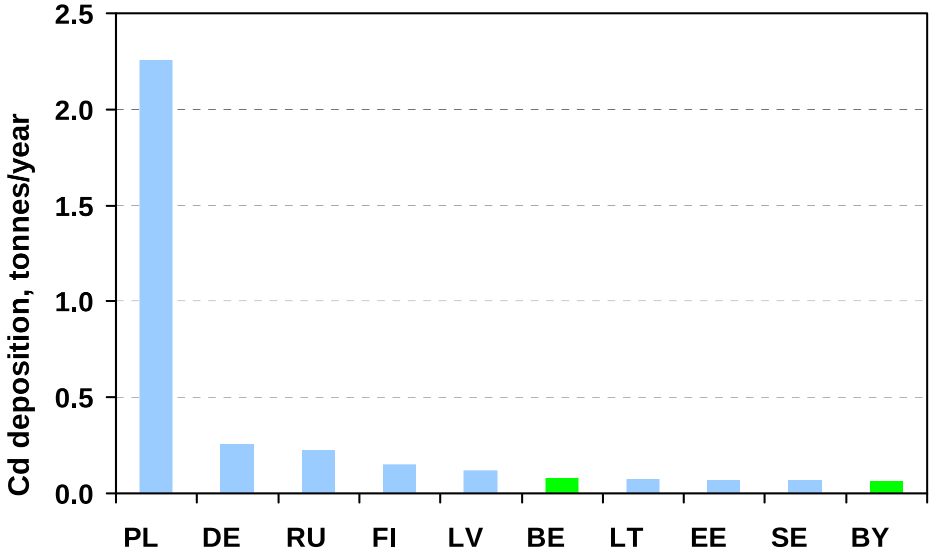


Figure 5.28. Top ten countries with the highest contribution to annual total deposition of **cadmium** over the Baltic Sea for 2012, tonnes/year.

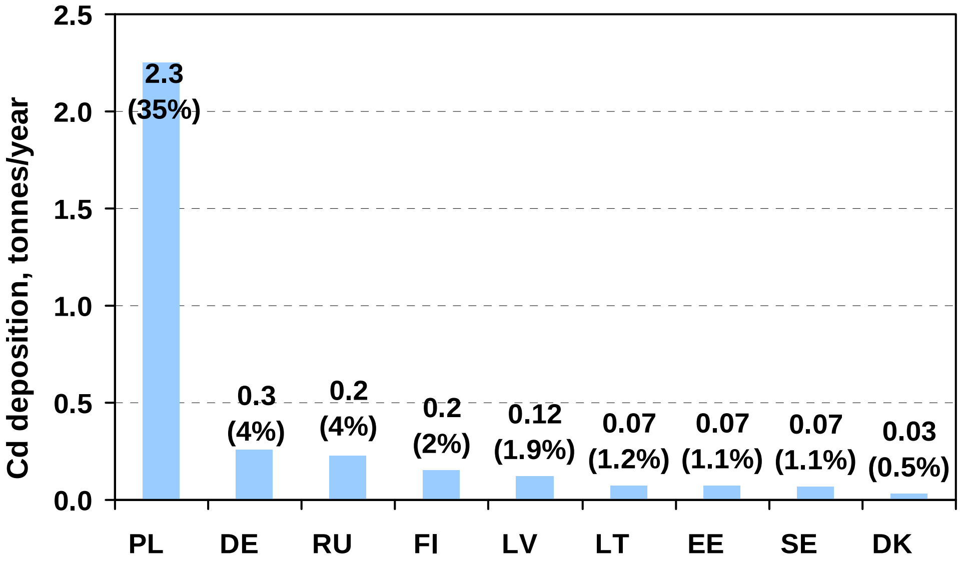


Figure 5.29. Sorted contributions (in tonnes/year and in %) of HELCOM countries to total **cadmium** deposition over the Baltic Sea for 2012. HELCOM countries emissions of **cadmium** contributed about 51% to the total annual cadmium deposition over the Baltic Sea. Contribution of other EMEP countries accounted for 7%. Significant contribution was made by other emission sources, in particular, remote emissions sources, natural emissions and wind re-suspension of cadmium (42%).

Table 5.4. Two most significant contributors to the annual total deposition of **cadmium** to the nine Baltic Sea sub-basins for 2012.

Sub-basin	**Country(1)**	**%**	**Country(1)**	**%**	***, %**
ARC	Poland	31	Finland	7	41
BOB	Poland	18	Finland	16	44
BOS	Poland	29	Finland	8	42
BAP	Poland	43	Germany	4	40
GUF	Russia	23	Poland	17	32
GUR	Poland	32	Latvia	9	38
KAT	Poland	17	Germany	7	61
SOU	Poland	22	Germany	9	55
WEB	Poland	14	Germany	13	59
BAS	Poland	35	Germany	4	42

* - contribution of re-emission, natural and remote sources (NSR).

5.5 Comparison of model results with measurements

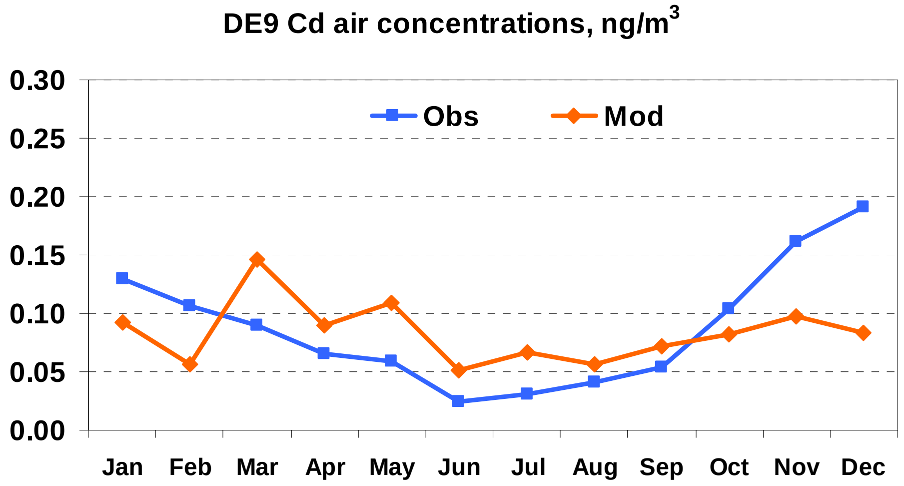


Figure 5.30. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Zingst (DE9). Units: ng / m^3.

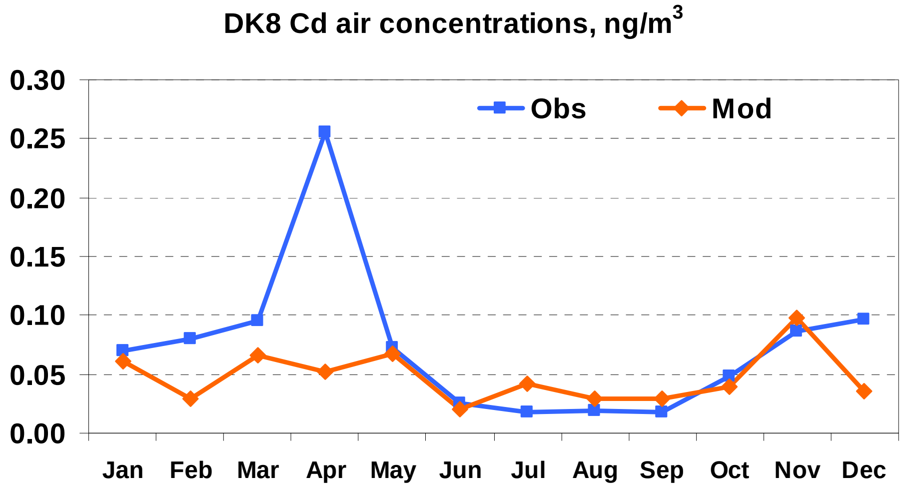


Figure 5.31. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Anholt (DK8). Units: ng / m^3.

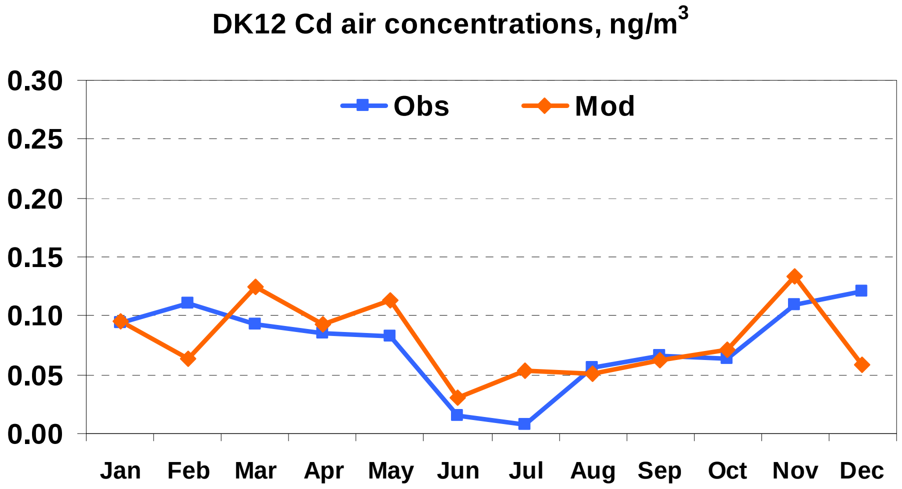


Figure 5.32. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Risoe (DK12). Units: ng / m^3.

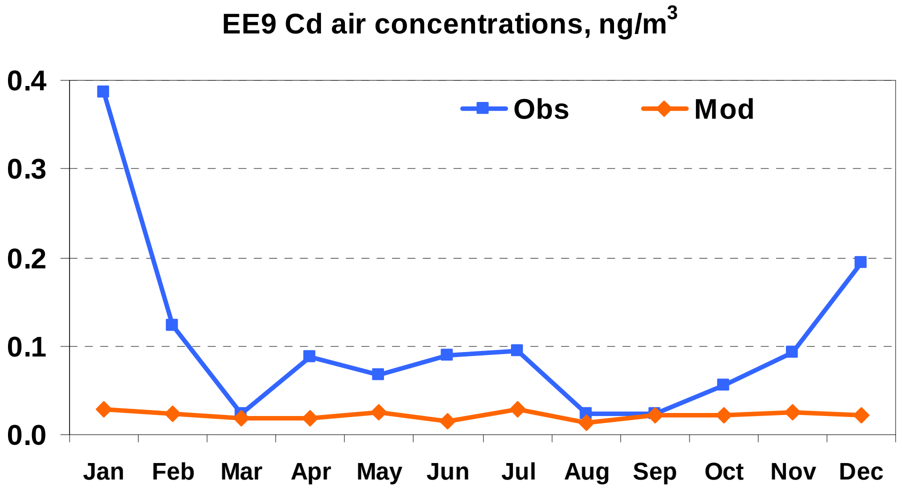


Figure 5.33. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Lahemaa (EE9). Units: ng / m^3.

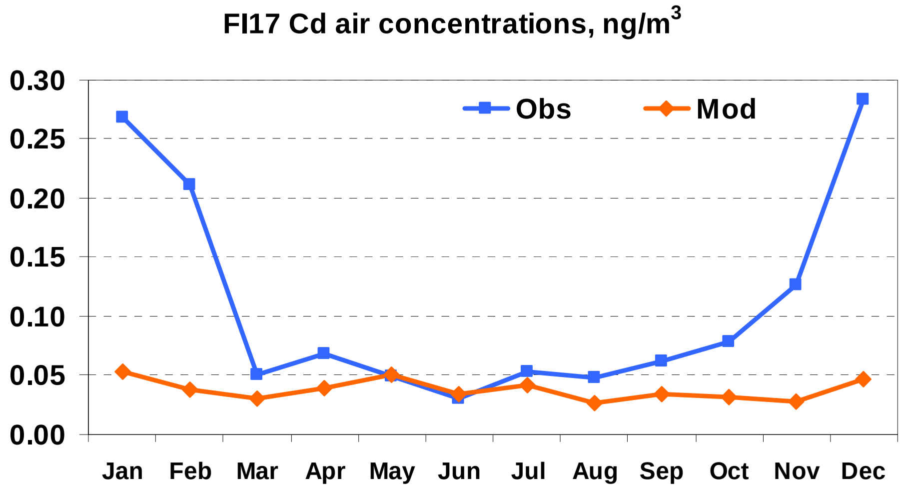


Figure 5.34. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Virolahty II (FI17). Units: ng / m^3.

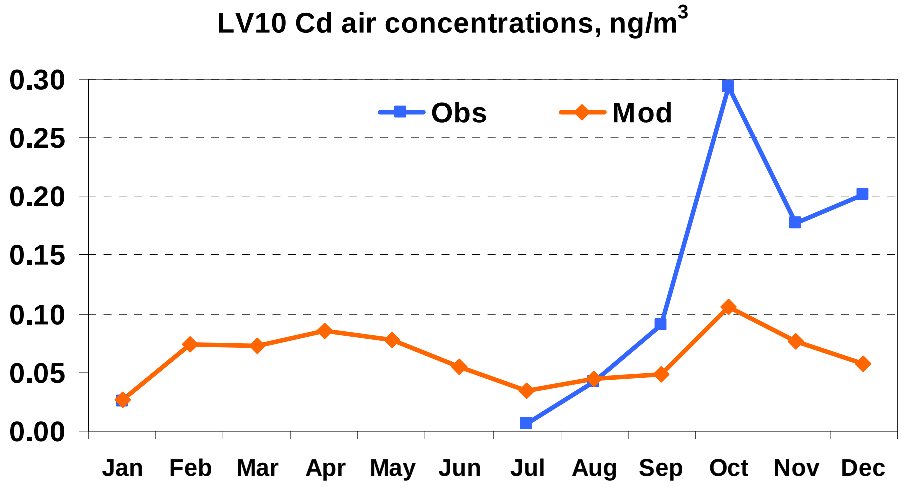


Figure 5.35. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Rucava (LV10). Units: ng / m^3.

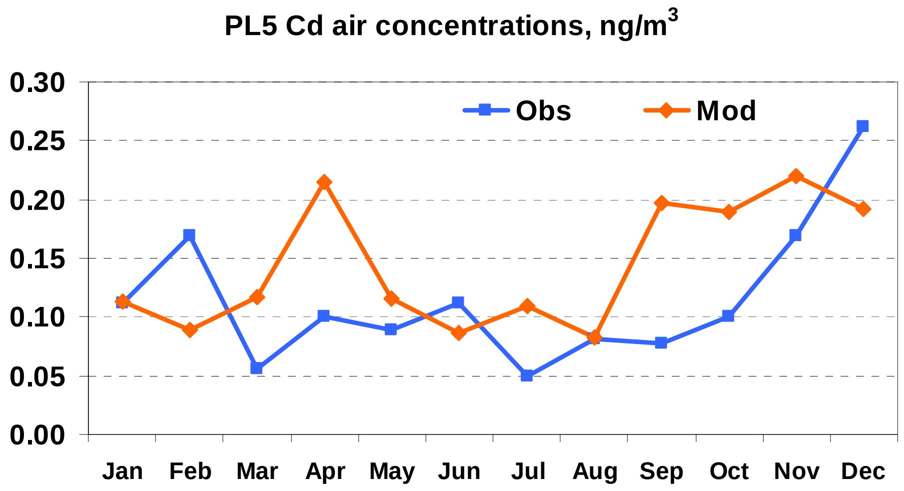


Figure 5.36. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Diabla Gora (PL5). Units: ng / m^3.

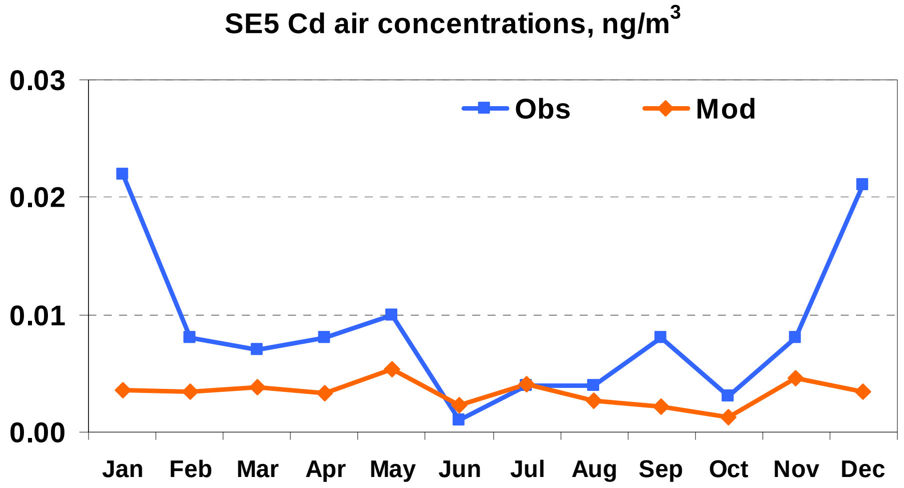


Figure 5.37. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Bredkälen (SE5). Units: ng / m^3.

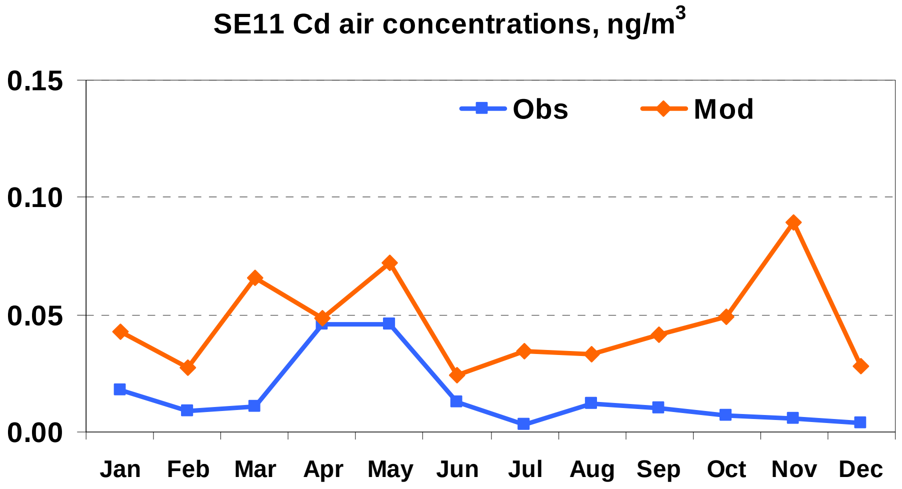


Figure 5.38. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Vavihill (SE11). Units: ng / m^3.

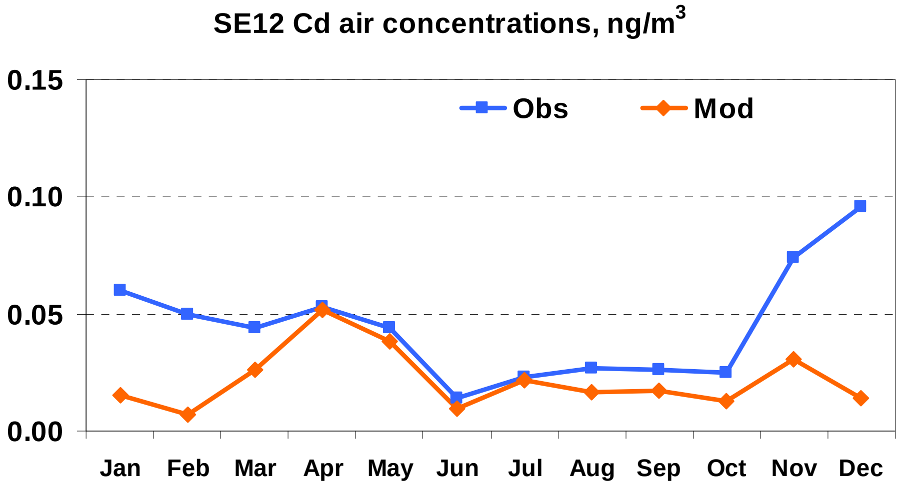


Figure 5.39. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Aspvreten (SE12). Units: ng / m^3.

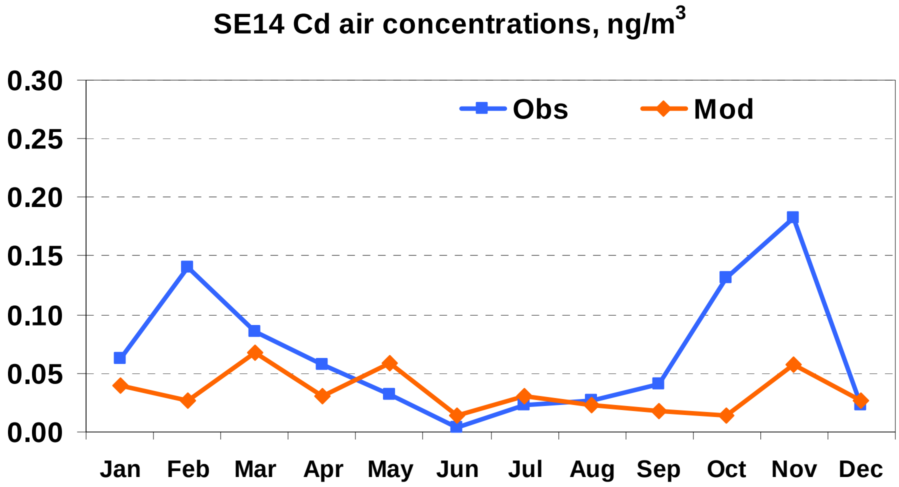


Figure 5.40. Comparison of calculated mean monthly cadmium concentrations in air for 2012 with measurements of the station Räo (SE14). Units: ng / m^3.

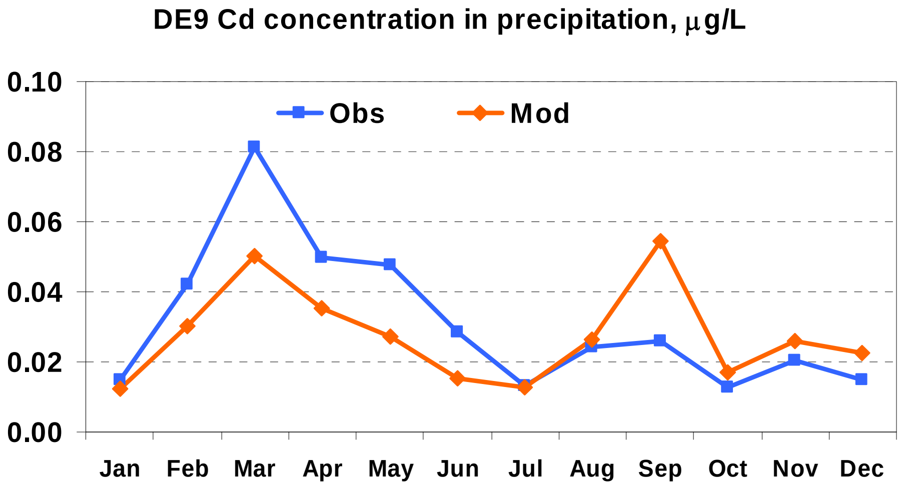


Figure 5.41. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Zingst (DE9). Units: µg / L.

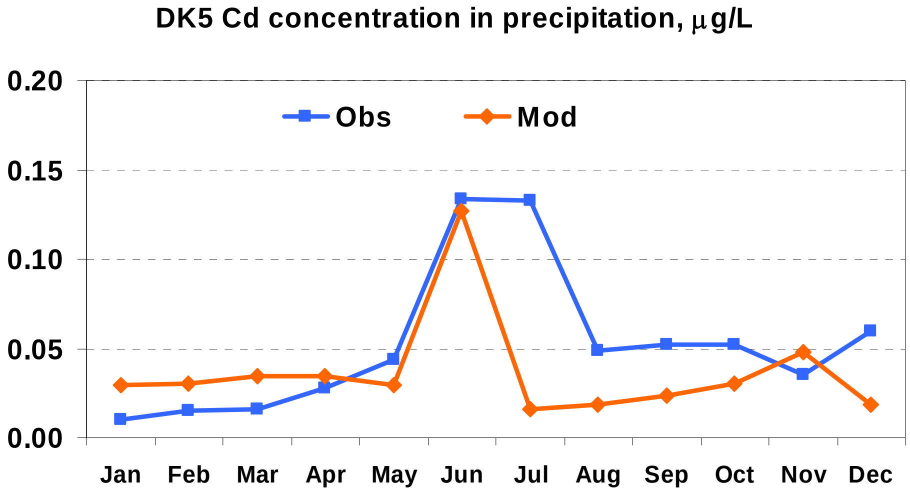


Figure 5.42. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Keldsnor (DK5). Units: µg / L.

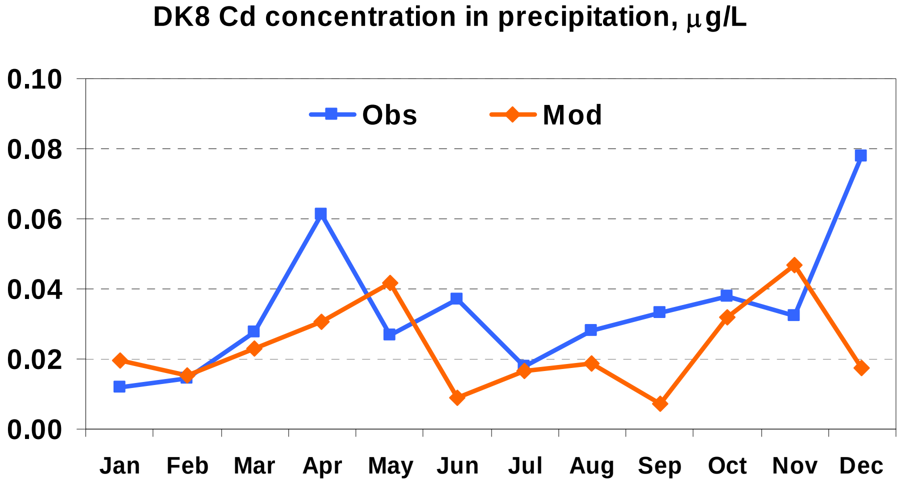


Figure 5.43. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Anholt (DK8). Units: µg / L.

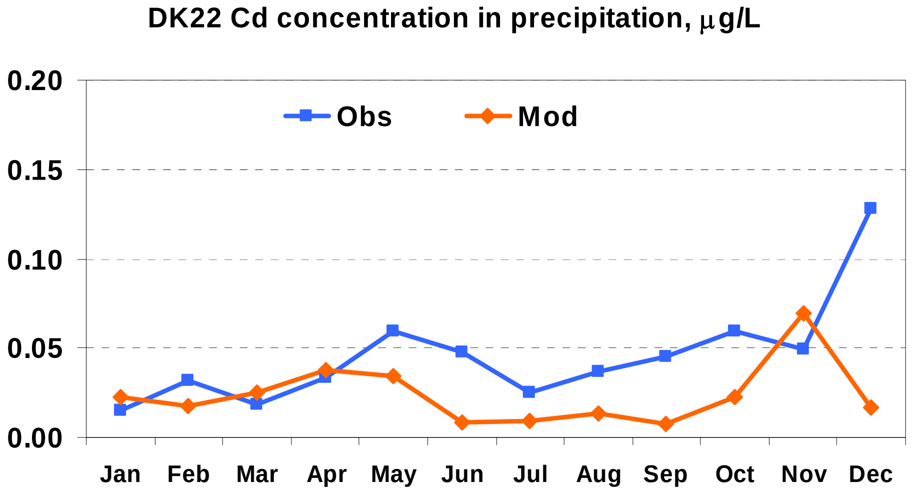


Figure 5.44. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Storebaelt (DK22). Units: µg / L.

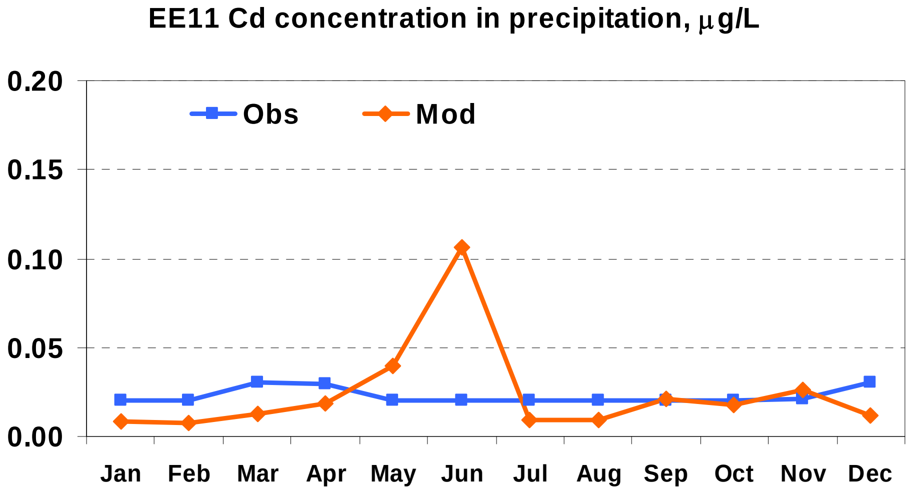


Figure 5.45. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Vilsandi (EE11). Units: µg / L.

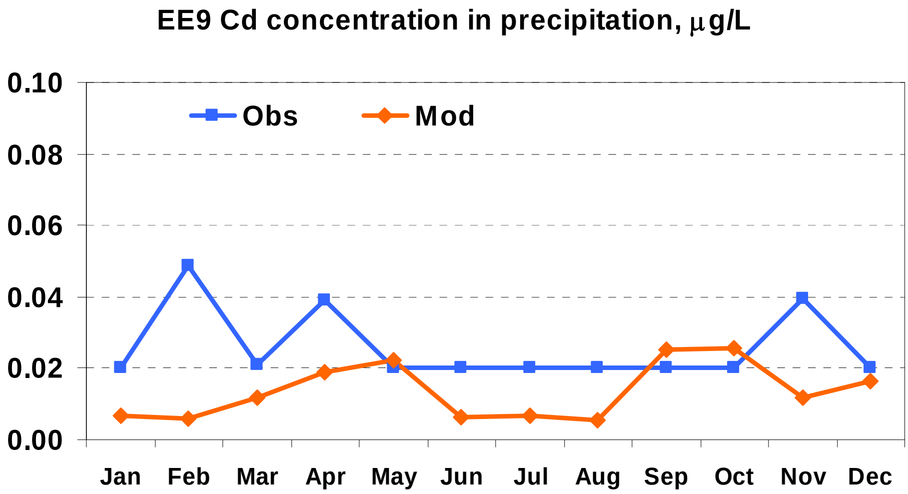


Figure 5.46. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Vilsandi (EE9). Units: μg / L.

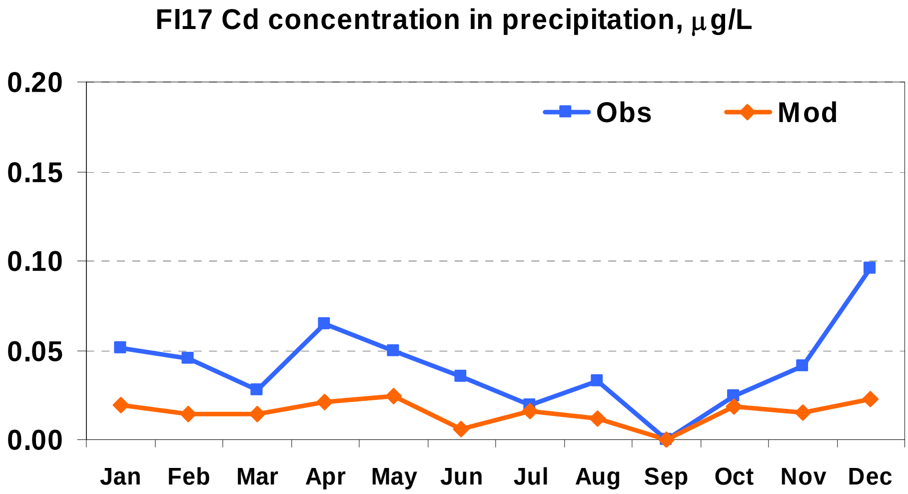


Figure 5.47. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Virolahty II (FI17). Units: μg / L.

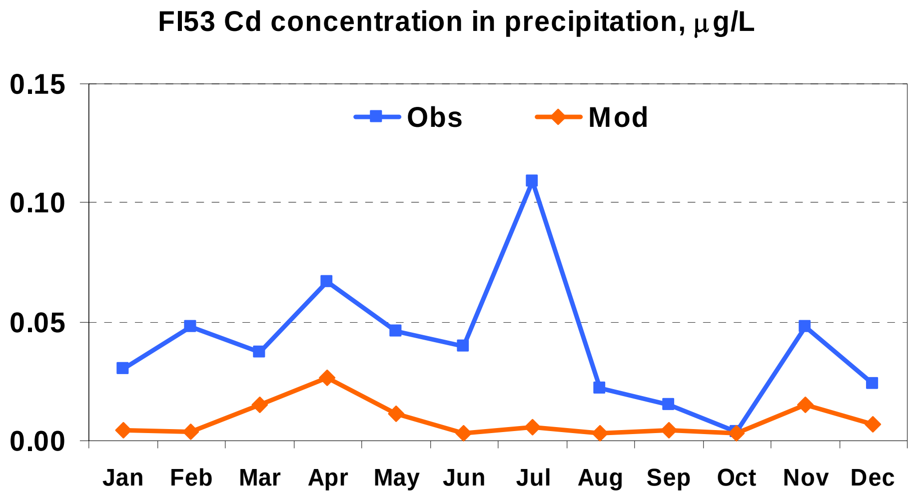


Figure 5.48. Comparison of calculated mean monthly cadmium concentrations in precipitation 2012 with measurements of the station Hailuoto (FI53). Units: µg / L.

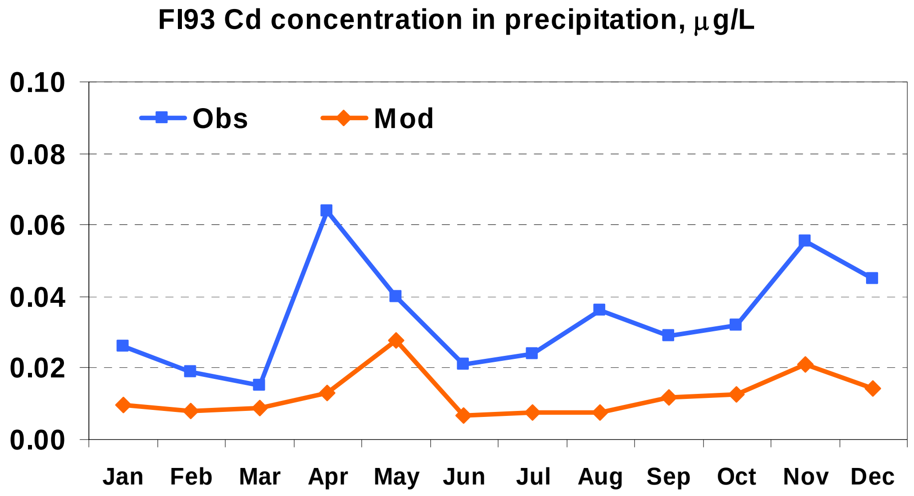


Figure 5.49. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Kotinen (FI93). Units: µg / L.

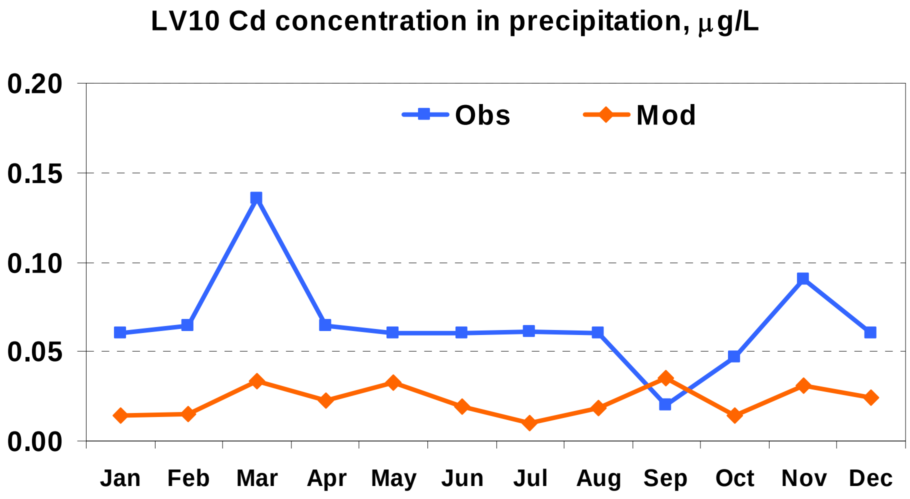


Figure 5.50. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Rucava (LV10). Units: µg / L.

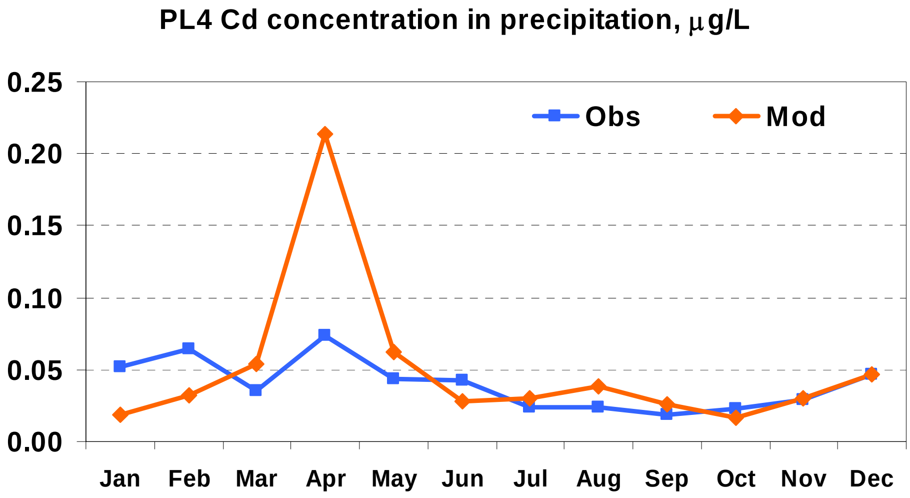


Figure 5.51. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Leba (PL4). Units: µg / L.

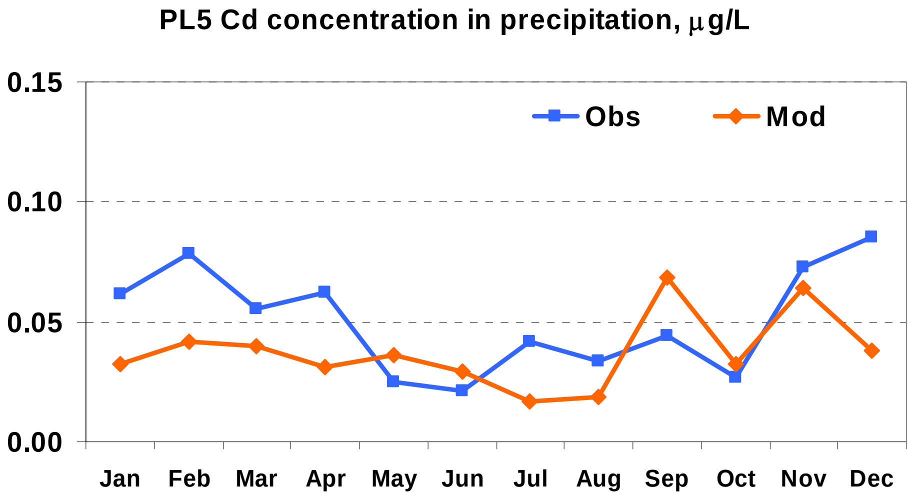


Figure 5.52. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Diabla Gora (PL5). Units: µg / L.

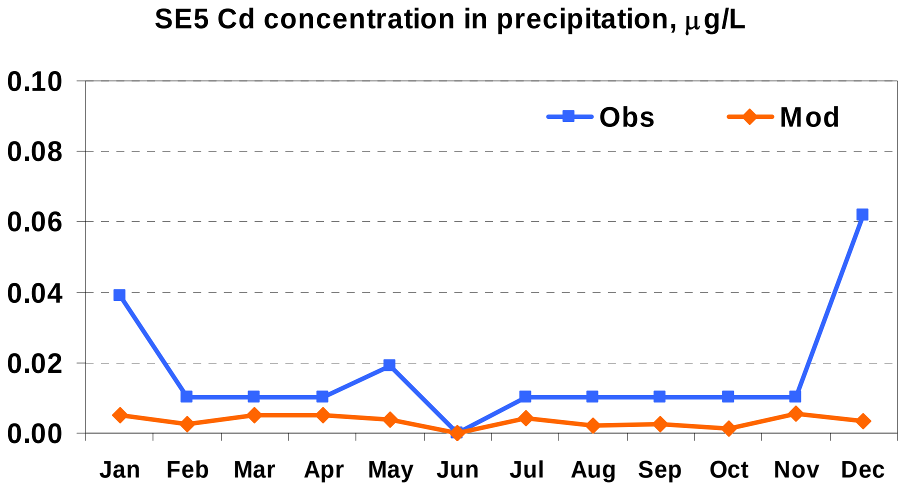


Figure 5.53. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Bredkälen (SE5). Units: µg / L.

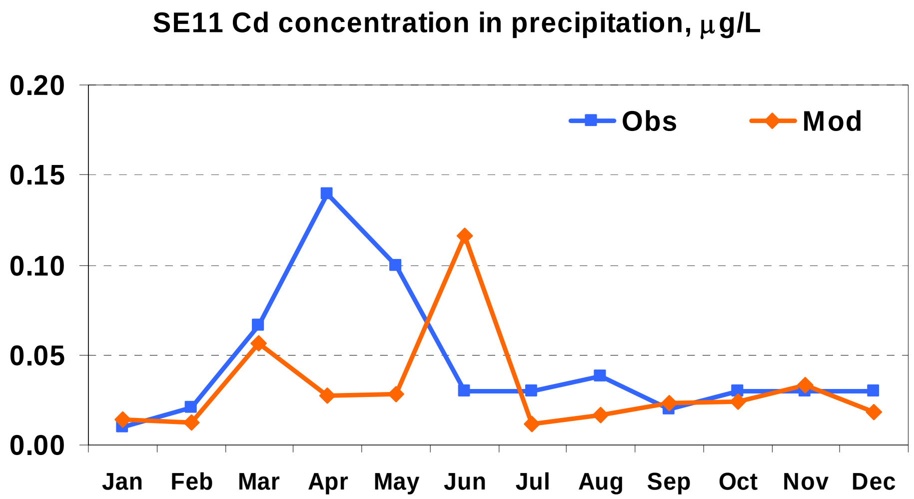


Figure 5.54. Comparison of calculated mean monthly cadmium concentrations in precipitation for 2012 with measurements of the station Vavihill (SE11). Units: μg / L.

Reasonable level of agreement between the computed concentrations of cadmium in air and in precipitation is obtained for the selected monitoring sites around the Baltic Sea. Comparing to lead more significant deviations between simulated and observed monthly mean concentrations of cadmium are found. The reason of deviations is connected with the uncertainties in seasonal variation of cadmium emission, differences between measured precipitation amount and the one used in the model, and difficulties in measurements of heavy metals.

5.6 Concluding remarks

- Emissions of cadmium from HELCOM countries have decreased from 1990 to 2012 by 58%. From 2011 to 2012 cadmium emissions have slightly increased by 1.6%.

- Annual deposition of cadmium to the Baltic Sea has decreased from 1990 to 2012 by 53%. Level of cadmium deposition has slightly decreased from 2011 to 2012 by 0.6%.

- The contribution of anthropogenic sources of HELCOM countries to total cadmium deposition over the Baltic Sea for 2012 was estimated to approximately 51%. Essential contribution belongs to the anthropogenic sources of other EMEP countries (7%) and natural sources and wind re-suspension (42%).

- Among the HELCOM countries the most significant contribution to cadmium deposition over the Baltic Sea was made by Poland (35%) and Germany (4%).

- Modelling results for cadmium were on average within a factor of two in comparison with measurements made around the Baltic Sea in 2012.

6. Atmospheric Supply of Mercury to the Baltic Sea in 2012

In this chapter the results of model evaluation of mercury atmospheric input to the Baltic Sea and its sub-basins for 2012 is presented. Modelling of mercury atmospheric transport and deposition was carried out using MSC-E Eulerian Heavy Metal transport model MSCE-HM (*Travnikov and Ilyin*, 2005). Latest available official information on mercury emission from HELCOM countries and other European countries was used in computations. Based on these data annual and monthly levels of mercury deposition to the Baltic Sea region have been obtained and contributions of HELCOM countries emission sources to the deposition over the Baltic Sea are estimated. Model results were compared with observed levels of mercury concentrations in air and precipitation measured at monitoring sites around the Baltic Sea in 2012.

6.1 Mercury emissions

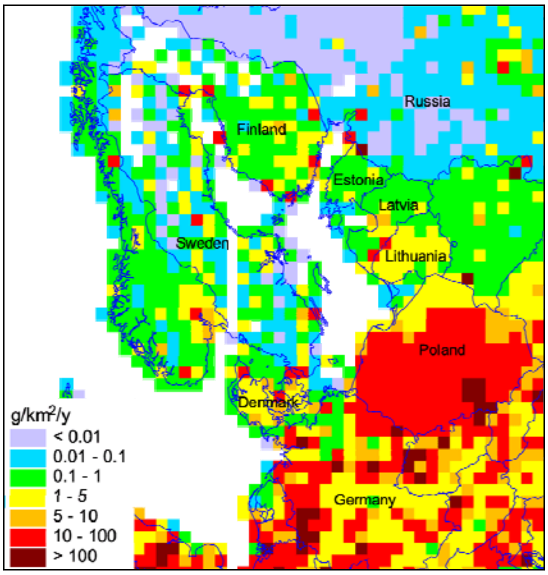


Figure 6.1. Annual total anthropogenic emissions of mercury in the Baltic Sea region for 2012, g/km^2/year.

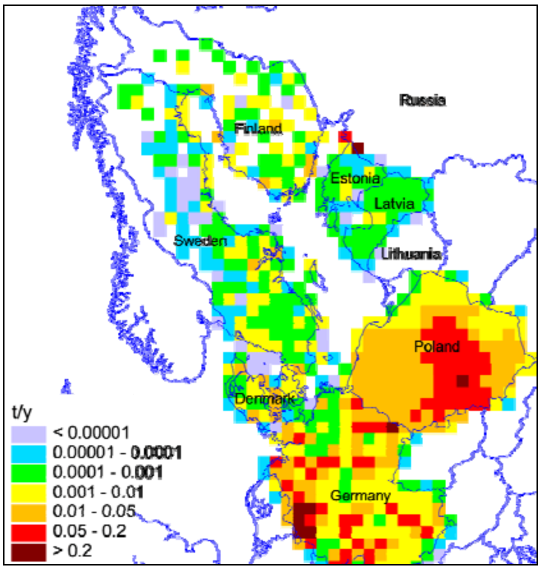


Figure 6.2. Annual mercury emission from Public Power sector for 2012, t/grid cell/y (white color means no information).

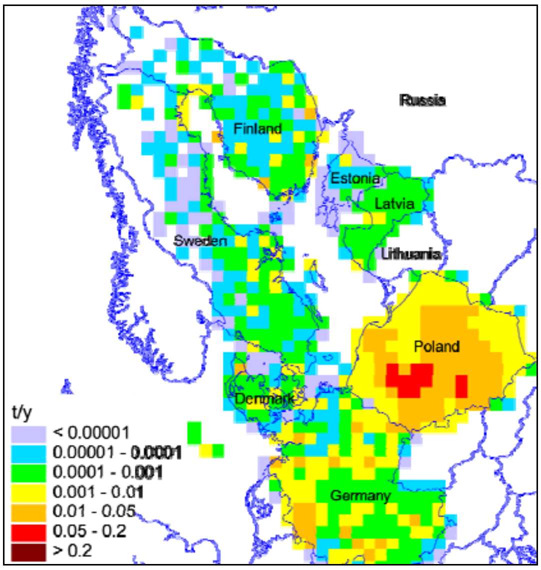


Figure 6.3. Annual mercury emission from Industrial Combustion sector for 2012, t/grid cell/y (white color means no information).

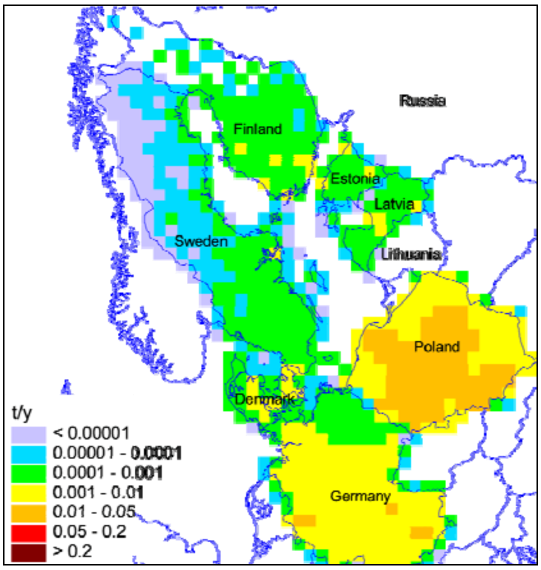


Figure 6.4. Annual mercury emission from Small Combustion sector for 2012, t/grid cell/y (white color means no information).

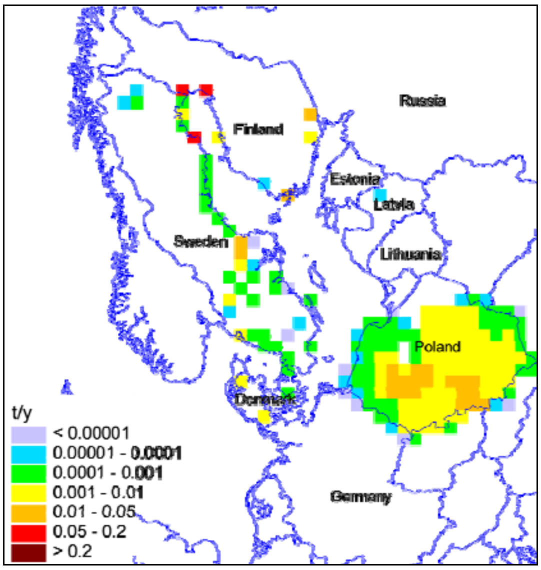


Figure 6.5. Annual mercury emission from Industrial Processes sector for 2012, t/grid cell/y (white color means no information).

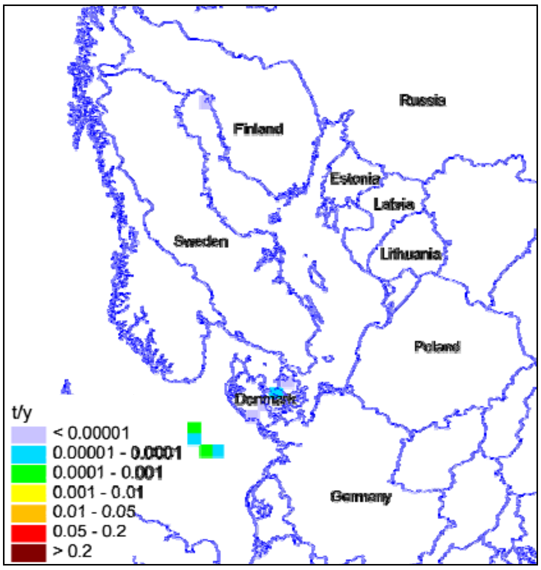


Figure 6.6. Annual mercury emission from Fugitive Emissions sector for 2012, t/grid cell/y (white color means no information).

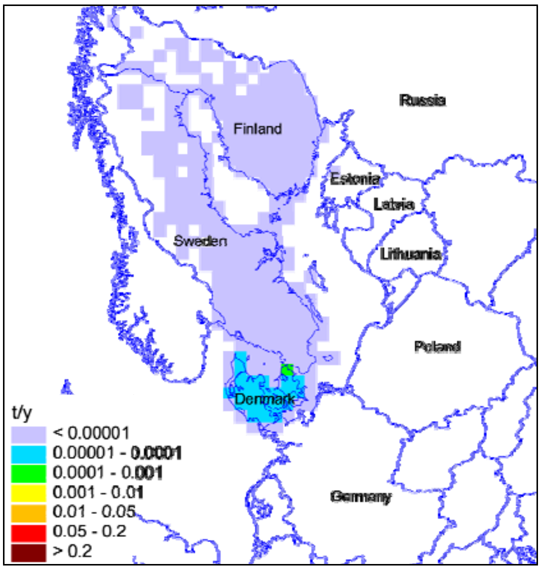


Figure 6.7. Annual mercury emission from Solvents sector for 2012, t/grid cell/y (white color means no information).

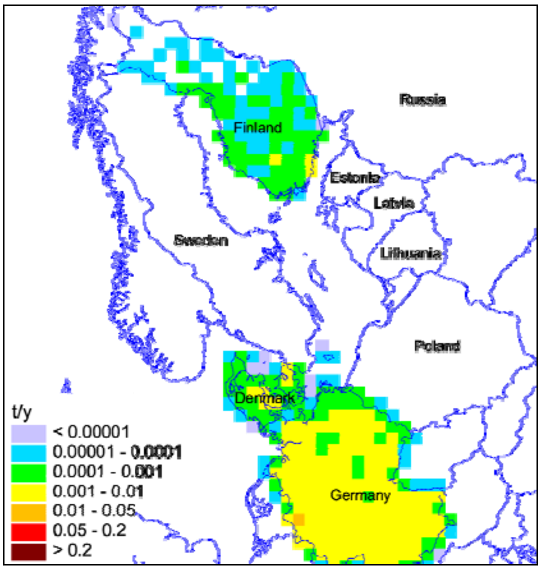


Figure 6.8. Annual mercury emission from Road Rail sector for 2012, t/grid cell/y (white color means no information).

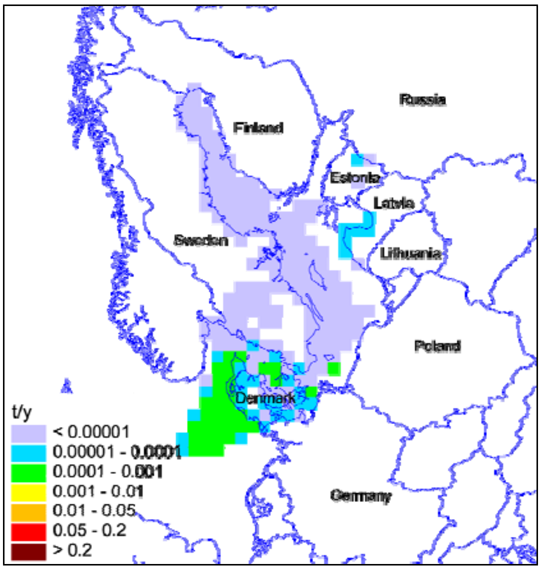


Figure 6.9. Annual mercury emission from Shipping Emissions sector for 2012, t/grid cell/y (white color means no information).

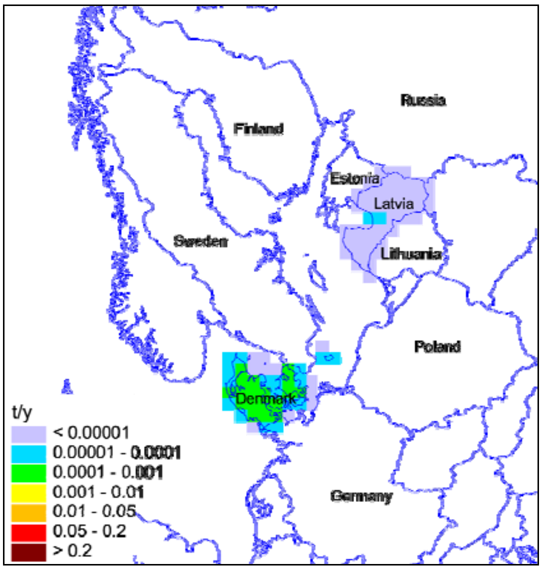


Figure 6.10. Annual mercury emission from Off Road Mobility sector for 2012, t/grid cell/y (white color means no information).

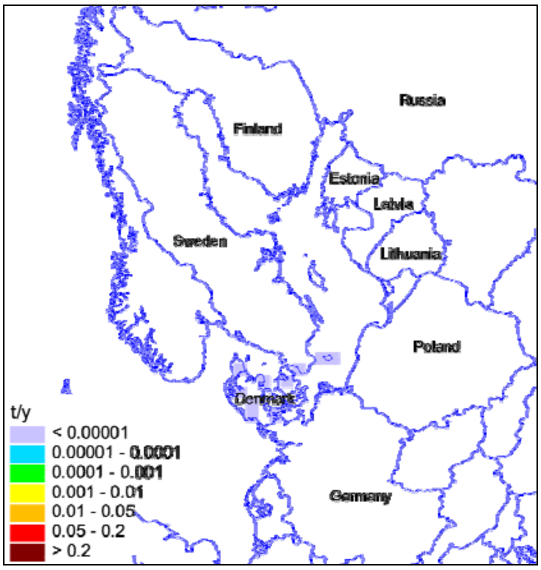


Figure 6.11. Annual mercury emission from Civil Aviation sector for 2012, t/grid cell/y (white color means no information).

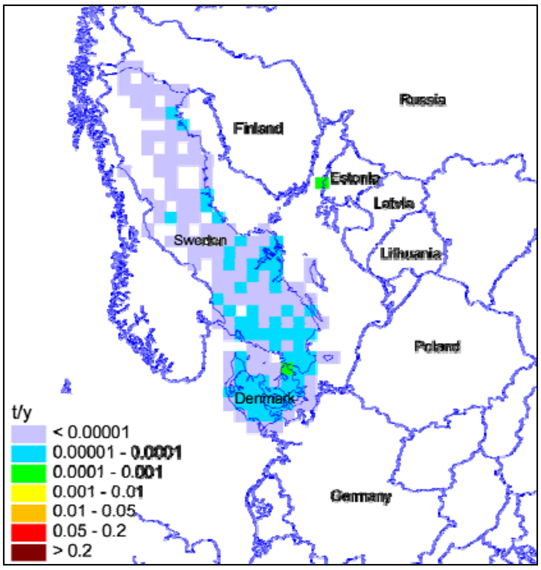


Figure 6.12. Annual mercury emission from Other Waste Displacement sector for 2012, t/grid cell/y (white color means no information).

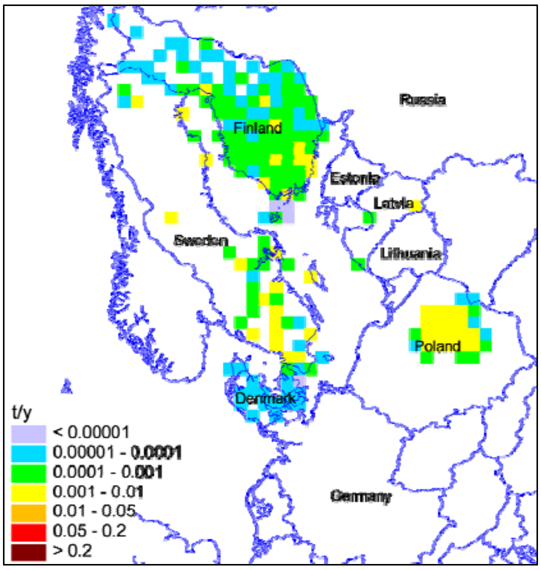


Figure 6.13. Annual mercury emission from Waste Incineration sector for 2012, t/grid cell/y (white color means no information).

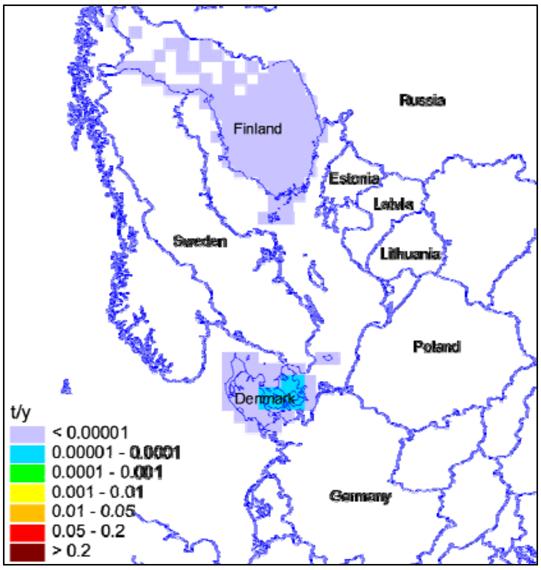
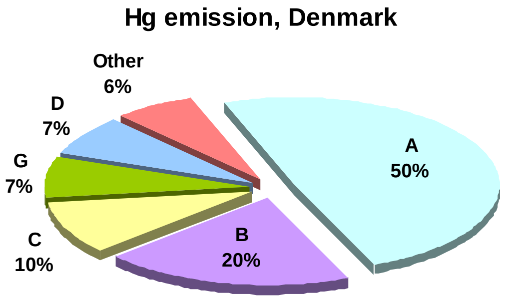


Figure 6.14. Annual mercury emission from Agricultural Wastes sector for 2012, t/grid cell/y (white color means no information).

Table 6.1. Annual total mercury anthropogenic emissions of HELCOM countries from different sectors for 2012, tonnes/year

GNFR emission sector	Sector name	Denmark	Estonia	Finland	Germany	Latvia	Lithuania	Poland	Russia	Sweden
A	Public Power	0.155	0.532	0.215	6.499	0.013	0.038	5.643		0.171
B	Industrial Combustion	0.062	0.004	0.157	0.729	0.034	0.048	2.408	0.672	0.064
C	Small Combustion	0.031	0.019	0.04	0.441	0.033	0.042	1.546		0.029
D	Industrial Processes	0.023		0.28	2.279	4.8E-05	1.0E-04	0.589	0.130	0.131
E	Fugitive Emissions	4.0E-04	1.7E-07	1.0E-05	NA					
F	Solvents	0.001	1.0E-07	1.1E-06	NA	NA		4.7E-06		4.2E-07
G	Road Rail	0.023		0.021	0.418			NA		
H	Shipping Emissions	0.012	5.7E-05		0.001	2.1E-04	1.4E-04	8.3E-05		1.5E-04
I	Off Road Mobility	0.005		NA		5.6E-05	1.2E-05	0.0		3.8E-07
J	Civil Aviation	1.3E-05	NA	NA	NE	NE	NE	NA		NE
L	Other Waste Displacement	9.1E-04	4.2E-04	NA	NA		NE	NA		0.001
N	Waste Incineration	4.6E-04		0.049	4.3E-04	2.9E-03	0.234	0.056		0.050
Q	Agricultural Waste	3.4E-04	NE	4.8E-05	NO	NA	NO	NA		NO
R	Other	NO	NO	NO	NO	NA	NO	NO	0.178	NO
Total		0.31	0.55	0.76	10.37	0.08	0.36	10.24	0.98	0.45

NO – not occurring, an activity or process does not exist within a country.
NA – not applicable, the process or activity exists but emissions are considered never to occur.
NE – not estimated, emissions occur but have not been estimated or reported in this submission.
IE – included elsewhere, emissions by sources of compounds are estimated but included elsewhere in the inventory.



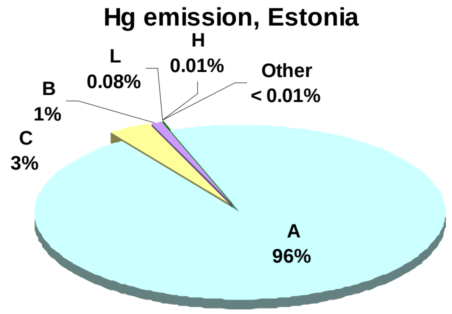
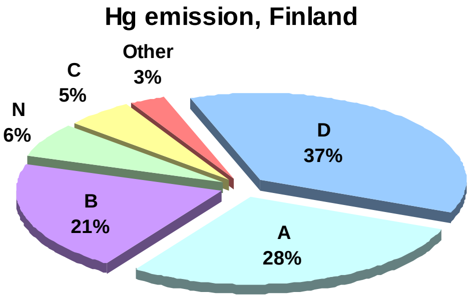


Figure 6.15. Contributions of different sectors to total annual mercury emission of Denmark in 2012

Figure 6.16. Contributions of different sectors to total annual mercury emission of Estonia in 2012



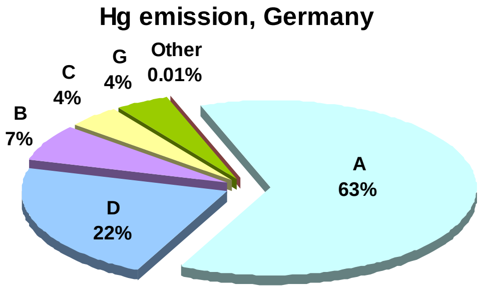


Figure 6.17. Contributions of different sectors to total annual mercury emission of Finland in 2012

Figure 6.18. Contributions of different sectors to total annual mercury emission of Germany in 2012

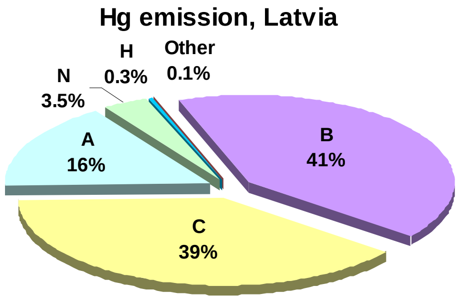


Figure 6.19. Contributions of different sectors to total annual mercury emission of Latvia in 2012

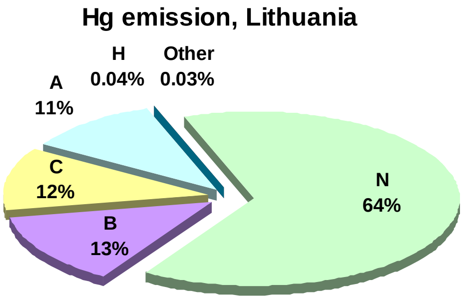


Figure 6.20. Contributions of different sectors to total annual mercury emission of Lithuania in 2012

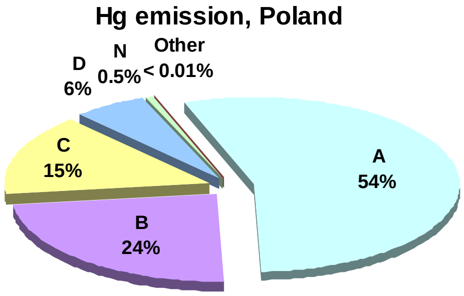


Figure 6.21. Contributions of different sectors to total annual mercury emission of Poland in 2012

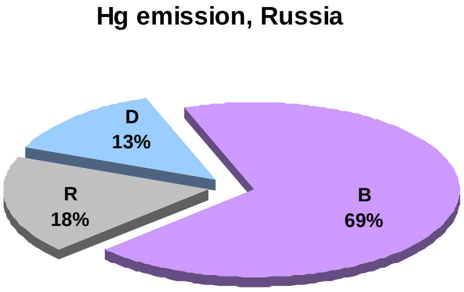


Figure 6.22. Contributions of different sectors to total annual mercury emission of Russia in 2012

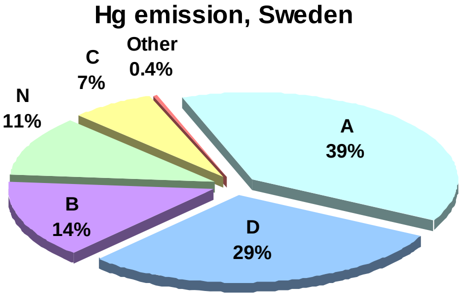
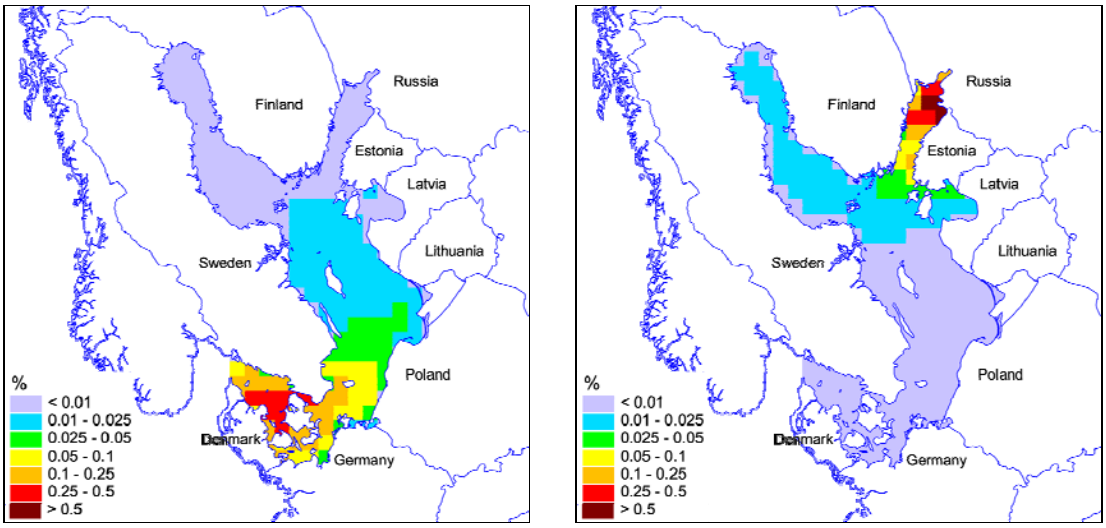
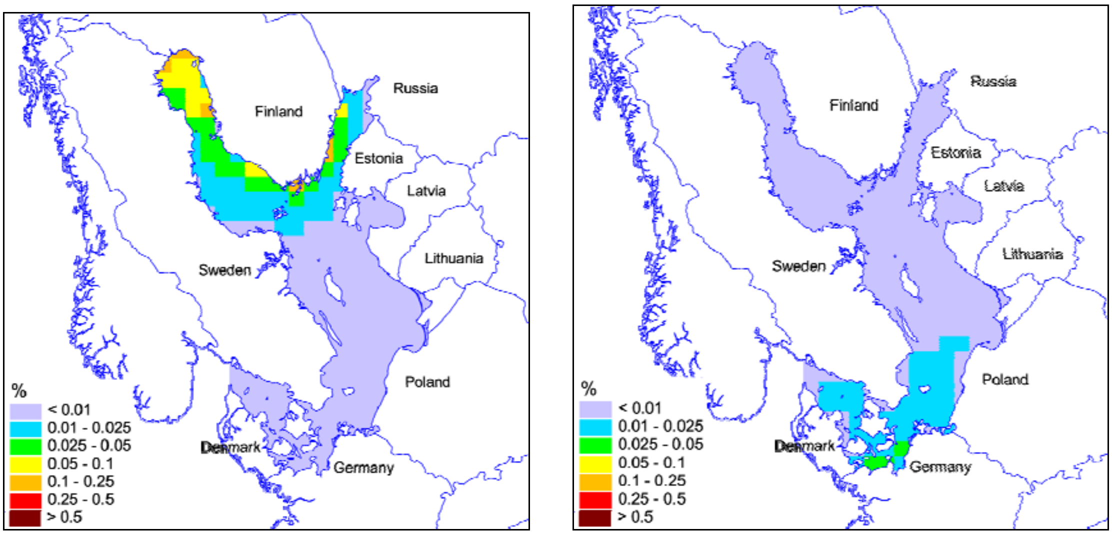


Figure 6.23. Contributions of different sectors to total annual mercury emission of Sweden in 2012

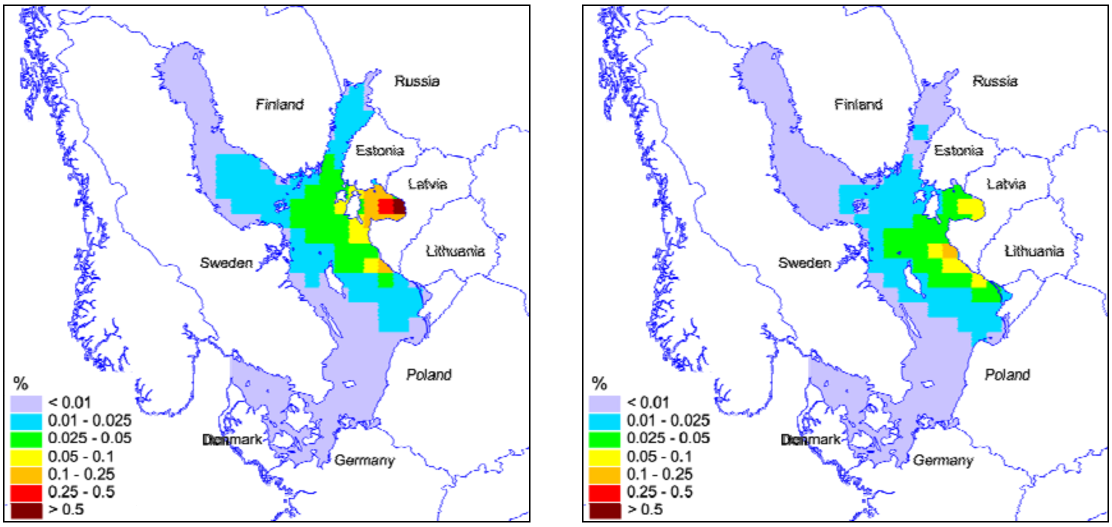


Denmark Estonia

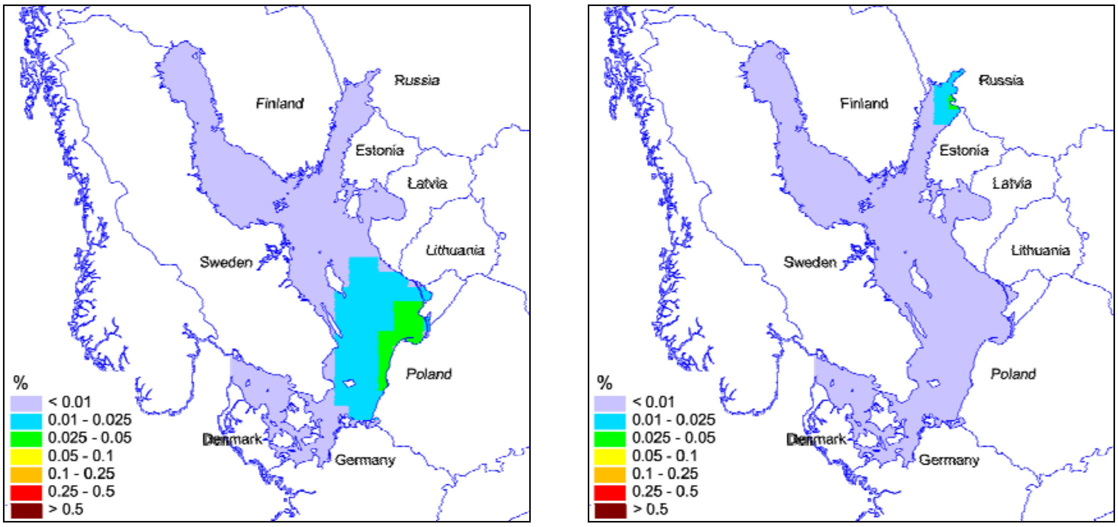


Finland Germany

Figure 6.24. Fractions of annual anthropogenic mercury emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

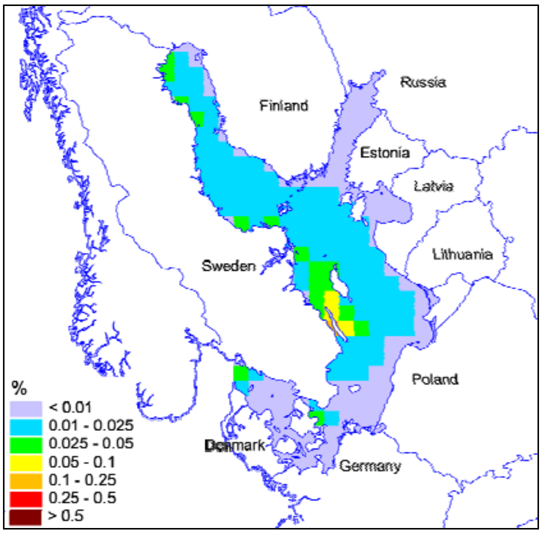


Latvia Lithuania



Poland Russia

Figure 6.24. (cont.) Fractions of annual anthropogenic mercury emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).



Sweden

Figure 6.24. (cont.) Fractions of annual anthropogenic mercury emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

Table 6.2. Annual total anthropogenic emissions of **mercury** of HELCOM countries and other EMEP countries in period 1990-2012, tonnes/year. (Expert estimates of emissions are shaded)

	1990	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**
DK	3,1	3,2	2,9	2,8	2,6	2,4	2,4	1,9	1,6	1,4	1	1	0,85	0,89	0,74	0,76	0,63	0,6	0,6	0,46	0,45	0,4	0,31
EE	1,1	1	0,83	0,64	0,64	0,6	0,6	0,6	0,53	0,5	0,51	0,5	0,5	0,58	0,54	0,52	0,52	0,65	0,57	0,44	0,63	0,63	0,55
FI	1,1	0,81	0,8	0,58	0,62	0,69	0,75	0,54	0,52	1,17	0,56	0,75	0,66	0,79	0,74	0,85	0,99	0,83	0,78	0,77	0,86	0,65	0,76
DE	32	26	22	19	17	17	17	17	17	16	16	16	15	15	14	13	13	12	10,8	9,8	10,2	10,1	10,4
LV	0,27	0,24	0,2	0,18	0,14	0,11	0,11	0,1	0,09	0,08	0,07	0,08	0,07	0,07	0,07	0,08	0,08	0,09	0,08	0,07	0,08	0,08	0,08
LT	1,1	1	0,4	0,4	0,3	0,5	0,3	0,4	0,3	0,18	0,19	0,25	0,29	0,32	0,33	0,49	0,39	0,3	0,16	0,14	0,19	0,32	0,36
PL	13	13	13	13	13	13	13	13	12	12	11	11	10	10	10	10	10	10	10	9,6	10	10	10
RU	16	13	11	12	10	10	10	9,6	9,4	9,9	10	10	10	11	12	14	14	9,7	5,3	1	1	1	1
SE	1,5	1,2	1,1	1	1	0,9	1	0,83	0,84	0,84	0,72	0,56	0,59	0,68	0,69	0,64	0,49	0,52	0,47	0,52	0,48	0,48	0,45
Total	**69**	**60**	**52**	**49**	**46**	**46**	**46**	**44**	**42**	**42**	**40**	**40**	**38**	**40**	**39**	**40**	**40**	**35**	**29**	**23**	**24**	**24**	**24**
AL	0,27	0,14	0,07	0,05	0,04	0,04	0,04	0,02	0,02	0,02	0,02	0,03	0,03	0,03	0,03	0,06	0,06	0,1	0,12	0,12	0,12	0,12	0,12
AM	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35	0,35
AT	2,1	2	1,6	1,4	1,2	1,2	1,2	1,1	0,95	0,94	0,89	0,96	0,92	0,96	0,93	1	1	1	1	0,9	1	1	1
AZ	0,98	0,98	0,98	0,98	0,98	0,98	0,98	0,98	0,98	0,98	0,98	1	1	1	1,1	1,1	1,1	1,1	1,1	1,2	1,2	1,2	1,2
BY	1,1	1,1	0,88	0,72	0,6	0,51	0,3	0,31	0,39	0,38	0,36	0,52	0,57	0,6	0,63	0,65	0,72	0,74	0,81	0,91	0,85	0,86	0,92
BE	11,7	5,5	5,5	2,7	4,1	3,1	3,3	3,6	2,6	3,1	3,3	2,9	4	3,8	3,7	2,5	2,3	3,4	3,9	2	2	2	1,7
BA	2	2	2	2	2	2	2	2	2	2	2	2	2	1,9	1,9	1,9	1,9	1,9	1,9	1,9	1,8	1,8	1,8
BG	2,4	2	1,8	1,8	1,9	1,9	1,9	2	1,8	1,6	1,5	1,4	1,3	2,2	2,1	1,6	1,7	1,5	1,4	1	0,87	0,94	0,78
HR	1,5	1,3	1,2	0,43	0,39	0,4	0,42	0,46	0,47	0,44	0,55	0,52	0,57	0,75	0,94	0,83	0,71	0,76	0,76	0,68	0,77	0,76	0,74
CY	0,17	0,16	0,17	0,19	0,2	0,19	0,2	0,2	0,19	0,19	0,19	0,19	0,2	0,19	0,2	0,2	0,2	0,2	0,2	0,17	0,14	0,14	0,14
CZ	7,5	7,4	7,3	7,5	7,2	7,4	5,9	5,5	5,2	3,7	3,8	3,3	2,8	1,8	2,1	3,8	3,8	3,9	4,1	4,3	3,5	3,2	3
FR	25	24	24	22	22	20	19	15	14	12	12	10,1	9,3	6,8	6,4	6,5	6,4	4,9	4,7	4,3	4,6	4,6	4,1
GE	0,25	0,25	0,25	0,25	0,25	0,25	0,25	0,25	0,25	0,25	0,25	0,26	0,26	0,27	0,27	0,28	0,28	0,29	0,29	0,3	0,31	0,31	0,32
GR	13	13	13	13	13	13	13	11	9,8	8,2	6,7	6,8	6,9	7	7,1	7,2	7,3	7,4	7,6	7,7	7,8	7,9	8
HU	6,3	5,8	5	5	4,7	4,2	4,7	4,5	4,3	3,6	3,6	3,5	3,2	3,1	3	3	3,2	2,8	3	2,8	0,78	0,82	0,6
IS	0,05	0,05	0,06	0,07	0,07	0,08	0,08	0,09	0,1	0,1	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11	0,11
IE	0,87	0,93	0,78	0,77	0,76	0,77	0,73	0,83	0,59	0,56	0,72	0,86	0,78	0,84	0,81	0,88	0,82	0,82	0,76	0,49	0,41	0,38	0,42
IT	12	11	11	10	10	10	9,8	10	9,5	8,8	9,3	9,4	9,3	9,1	9,9	10	10	11	10	8,5	8,7	8,9	8,4
KZ	10	10	10	10	10	10	10	10	10	10	10	11	11	11	11	11	11	11	12	12	12	12	12
LI	0,0001	0,0001	0,0001	0,0001	0,0001	0,0001	0,0001	0,0001	0,0002	0,0002	0,0002	0,0001	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002	0,0002
LU	0,3	0,28	0,25	0,23	0,2	0,1	0,1	0,1	0,1	0,29	0,27	0,29	0,29	0,29	0,29	0,29	0,29	0,29	0,29	0,29	0,29	0,29	0,29
MT	0,49	0,49	0,49	0,49	0,49	0,49	0,49	0,49	0,49	0,49	0,49	0,55	0,54	0,59	0,59	0,61	0,61	0,63	0,57	0,57	0,01	0,005	0,005
MC	0,11	0,11	0,12	0,13	0,07	0,07	0,07	0,08	0,08	0,08	0,08	0,09	0,08	0,06	0,06	0,06	0,04	0,06	0,05	0,05	0,05	0,05	0,05
NL	3,5	2,9	2,5	2,1	1,6	1,4	1,2	1	0,79	0,89	1	0,83	0,72	0,67	0,84	0,86	0,79	0,74	0,63	0,58	0,52	0,63	0,55
NO	1,4	1,3	1,2	0,89	1	0,79	0,82	0,84	0,79	0,83	0,68	0,63	0,6	0,6	0,62	0,61	0,56	0,55	0,52	0,4	0,46	0,41	0,39
PT	3,4	3,5	3,9	3,5	3,3	3,6	3,2	3,4	3,6	3,7	3,3	3,1	3,3	2,6	2,6	2,8	2,4	2,2	2,1	2	1,7	1,6	1,6
MD	3,4	3,8	3,3	1,8	1,3	0,89	0,95	0,57	0,41	0,18	0,26	0,23	0,39	0,34	0,32	0,24	0,22	0,19	0,24	0,17	0,12	0,2	0,2
RO	7,5	7,5	7,4	7,4	7,3	7,3	7,2	7,2	7,2	6,3	6,7	7,3	7,3	7,4	7,4	7,4	7,2	10	8	5	5,1	5	4,3
ME	0,07	0,08	0,06	0,06	0,05	0,02	0,07	0,06	0,08	0,08	0,07	0,06	0,09	0,08	0,08	0,06	0,08	0,06	0,09	0,05	0,09	0,09	0,09
RS	2,9	2,6	2,7	1,8	1,8	1,9	2,2	2,4	2,5	1,6	1,8	1,8	1,7	1,8	1,7	1,8	1,9	1,9	2	1,9	1,8	1,9	1,7
SK	13	9,5	6,2	5	3,9	4,3	3,4	3,6	3,9	3,6	6,1	4	3,2	2,9	3,2	3	3,5	2,6	2,8	1,1	1,4	1,2	1,3
SI	0,6	0,5	0,5	0,5	0,44	0,46	0,44	0,5	0,5	0,5	0,54	0,51	0,5	0,5	0,5	0,48	0,52	0,53	0,53	0,44	0,47	0,44	0,38
ES	14	15	16	15	15	16	15	13	14	14	13	12	12	11	11	11	10	9,1	8,2	7	6,7	6,4	6,3
CH	6,6	6,1	5,7	5,3	4,9	4	3,7	3,4	3,2	2,3	2,1	1,7	1,3	0,9	1	0,9	1	1	1	0,8	0,9	0,9	0,9
MK	0,94	0,57	0,53	0,47	0,42	0,55	0,63	0,54	0,54	0,51	0,55	0,32	0,3	0,27	0,24	0,28	0,28	0,24	0,39	0,3	0,39	0,41	0,3
TR	18	18	18	18	18	18	18	18	18	18	18	19	19	19	20	20	21	21	22	22	22	23	23
UA	36	35	34	33	32	31	30	29	28	27	26	25	5,9	30	6,6	6	16	7,6	6,8	5,6	6,8	6,8	6,8
UK	38	38	36	22	21	20	15	12	11	8,4	8,4	8,2	7,2	7,8	6,8	7,4	7,6	7,1	6,9	6,6	6,5	5,9	5,8
EMEP	**317**	**295**	**278**	**247**	**238**	**234**	**223**	**209**	**200**	**189**	**186**	**179**	**157**	**179**	**156**	**158**	**167**	**155**	**146**	**127**	**127**	**126**	**124**

Expert estimates:

- *Denier van der Gon, H.A.C., M. van het Bolscher A.J.H. Visschedijk P.Y.J. Zandveld [2006] Study to the effectiveness of the UNECE Persistent Organic Pollutants Protocol and costs of possible additional measures Phase I: Estimation of emission reduction resulting from the implementation of the POP Protocol, TNO report B&O-A R 2005/194*
- *Berdowski J.J.M., Baas J., Bloos J.P.J., Visschedijk A.J.H., Zandveld P.Y.J. [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990. TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03*

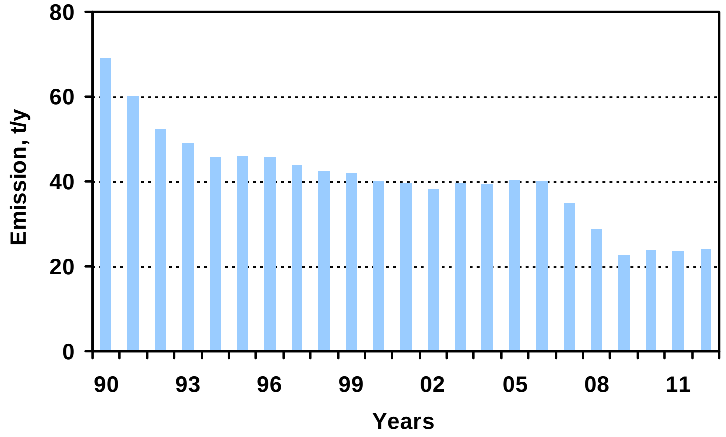


Figure 6.25. Time-series of total annual **mercury** emissions of HELCOM countries in 1990-2012, tonnes/year.

6.2 Annual total deposition of mercury

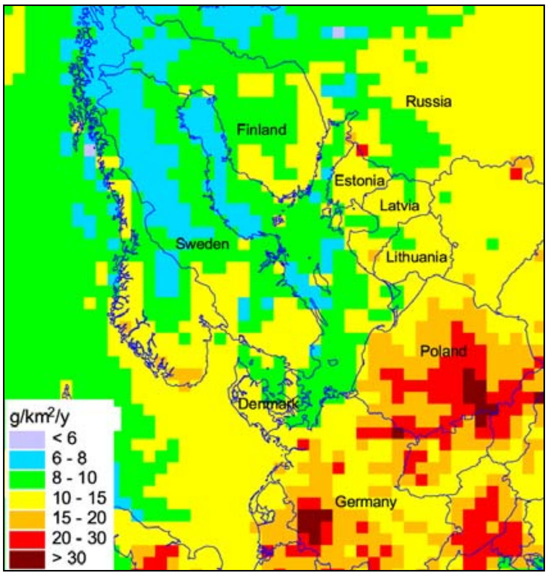


Figure 6.26. Annual total deposition fluxes of **mercury** over the Baltic Sea region for 2012, g/km^2/year.

6.3 Monthly total deposition of mercury

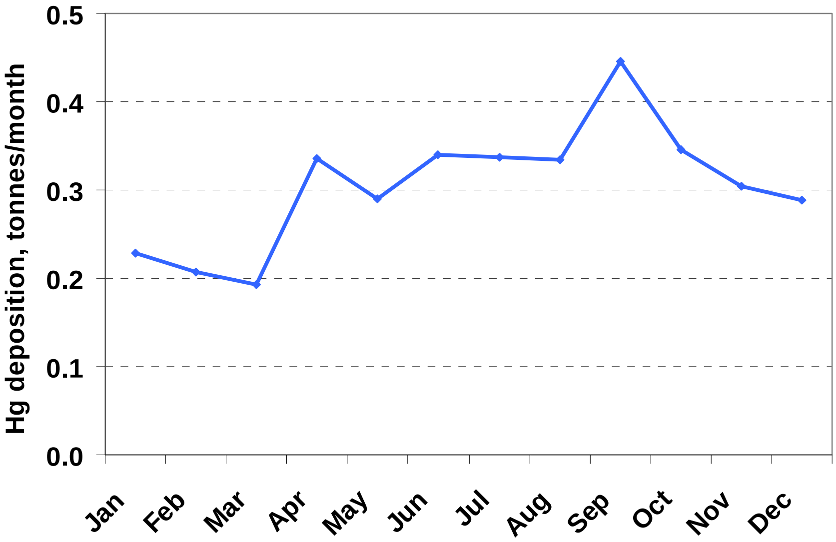


Figure 6.27. Monthly total deposition of **mercury** to the Baltic Sea for 2012, tonnes/month.

Table 6.3. Monthly total deposition of **mercury** to the Baltic Sea for 2012, tonnes/month.

Month	**Hg**
Jan	0.23
Feb	0.21
Mar	0.19
Apr	0.34
May	0.29
Jun	0.34
Jul	0.34
Aug	0.33
Sep	0.45
Oct	0.35
Nov	0.30
Dec	0.29

6.4 Source allocation of mercury deposition

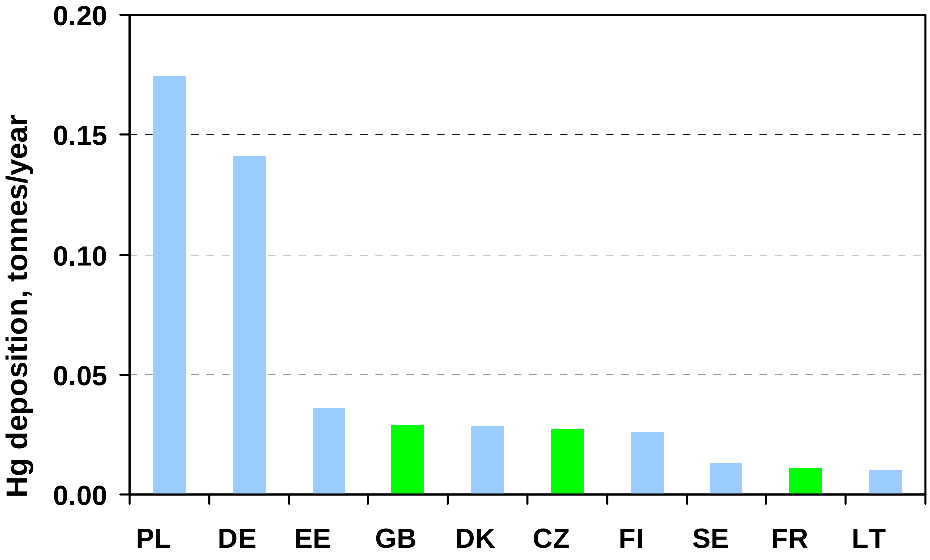


Figure 6.28. Top ten countries with the highest contribution to annual deposition of **mercury** over the Baltic Sea for 2012, tonnes/year.

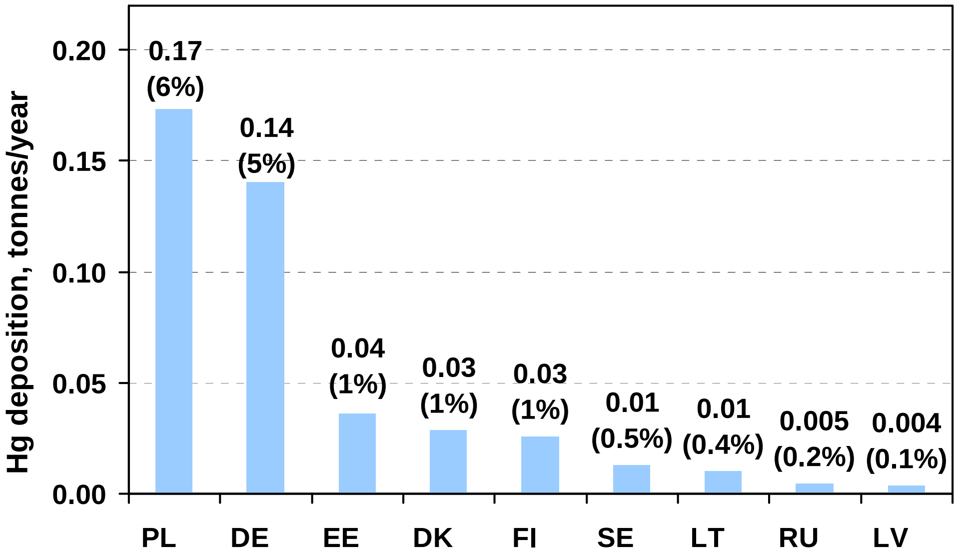


Figure 6.29. Sorted contributions (in %) of HELCOM countries to total deposition of **mercury** over the Baltic Sea for 2012. HELCOM countries emissions of mercury contributed 16% to the total annual **mercury** deposition over the Baltic Sea. Contribution of other EMEP countries accounted for 5%. Significant contribution was made by other emission sources, in particular, remote emissions sources, natural emissions and re-emission of **mercury** (79%).

Table 6.4. Two most significant contributors to the annual total deposition of **mercury** to the nine Baltic Sea sub-basins for 2012.

Sub-basin	Country(1)	%	Country(2)	%	*, %
ARC	Poland	4	Germany	3	84
BOB	Finland	6	Poland	2	86
BOS	Poland	3	Germany	2	87
BAP	Poland	9	Germany	6	77
GUF	Estonia	13	Poland	3	75
GUR	Poland	5	Germany	3	81
KAT	Germany	8	Denmark	5	77
SOU	Denmark	10	Germany	10	68
WEB	Germany	18	Denmark	5	67
BAS	Poland	6	Germany	5	78

* - contribution of re-emission, natural and remote sources.

6.5 Comparison of model results with measurements

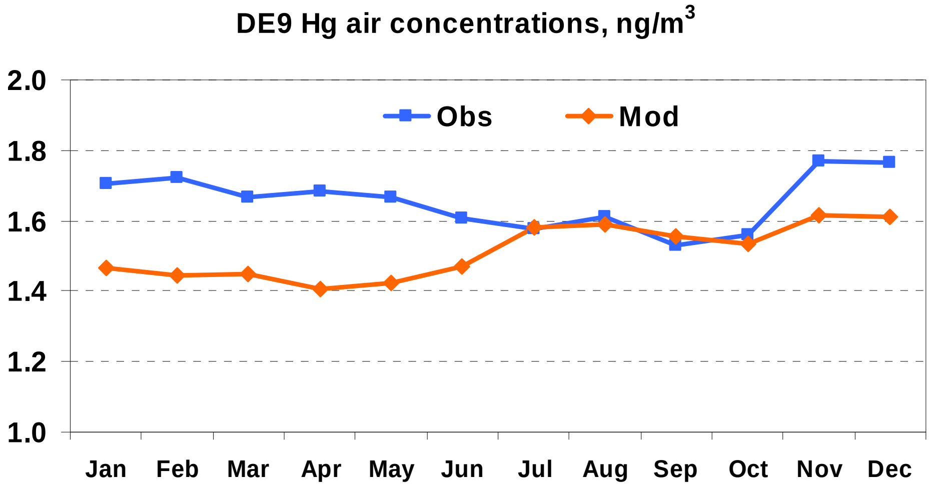


Figure 6.30. Comparison of calculated monthly mean Hg concentrations in air for 2012 with measurements of the station Zingst (DE9). Units: ng / m^3.

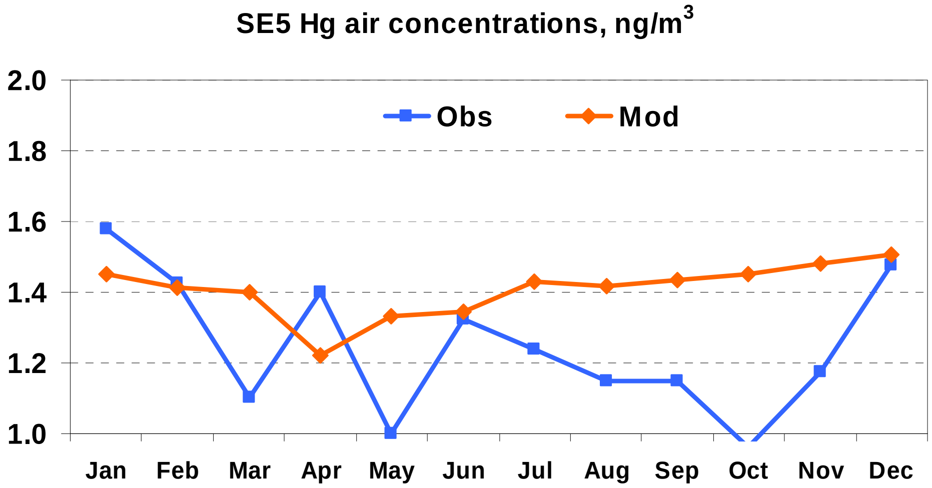


Figure 6.31. Comparison of calculated monthly mean Hg concentrations in air for 2012 with measurements of the station Bredkälen (SE5). Units: ng / m^3.

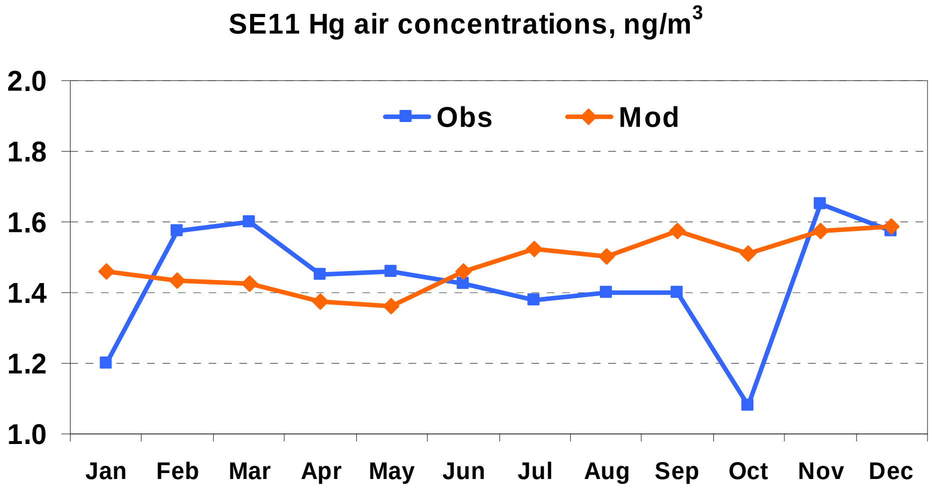


Figure 6.32. Comparison of calculated monthly mean Hg concentrations in air for 2012 with measurements of the station Vavihill (SE11). Units: ng / m^3.

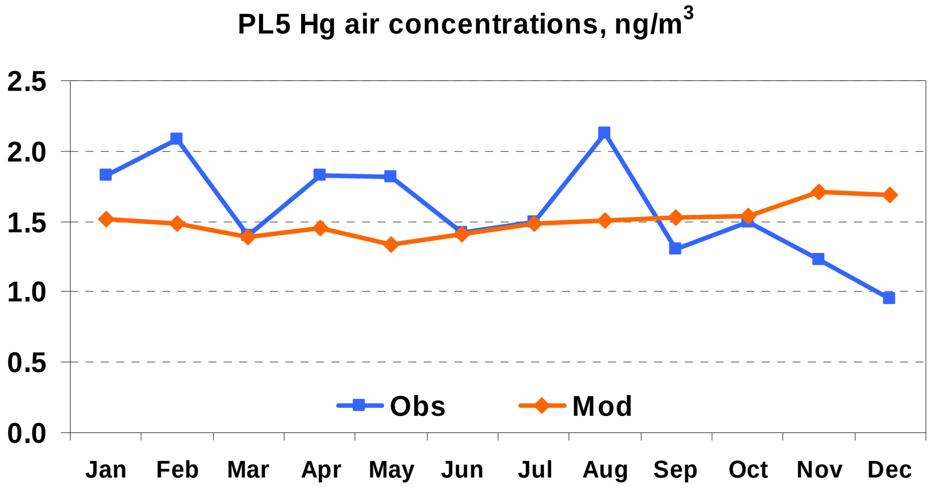


Figure 6.33. Comparison of calculated monthly mean Hg concentrations in air for 2012 with measurements of the station Diabla Gora (PL5). Units: ng / m^3.

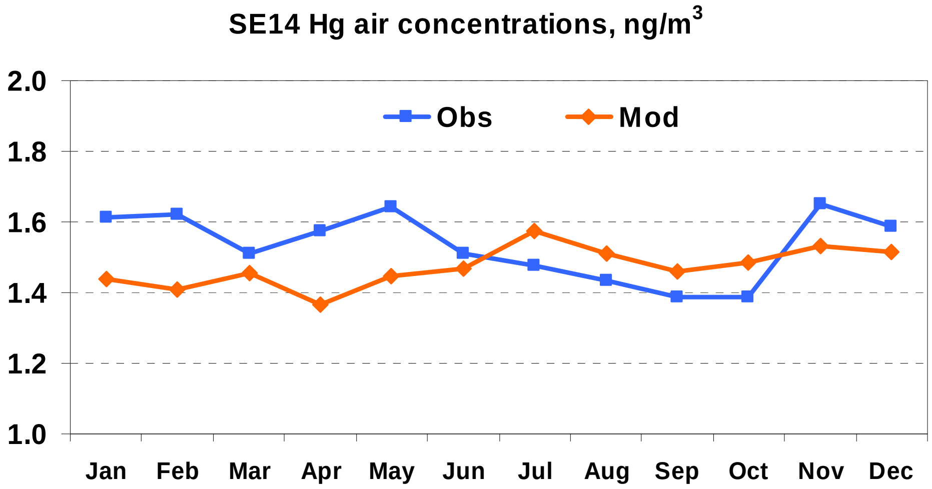


Figure 6.34. Comparison of calculated monthly mean Hg concentrations in air for 2012 with measurements of the station Råö (SE14). Units: ng / m^3.

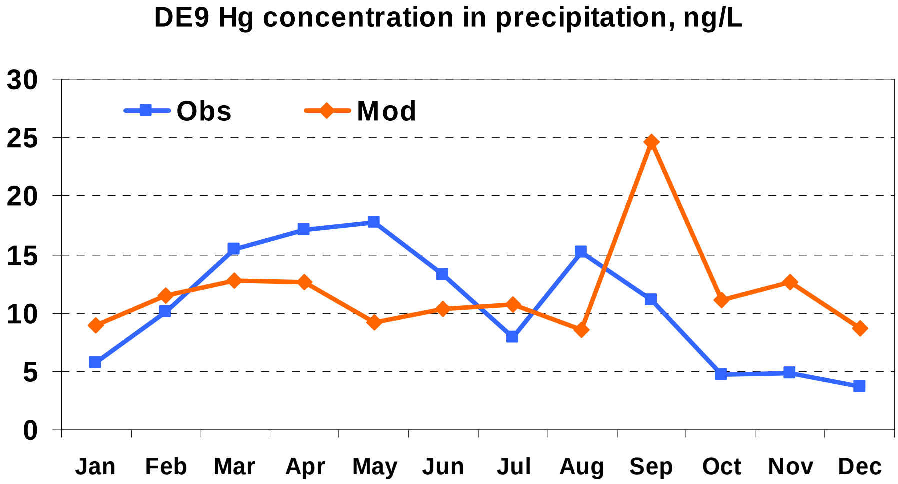


Figure 6.35. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Zingst (DE9). Units: ng/L.

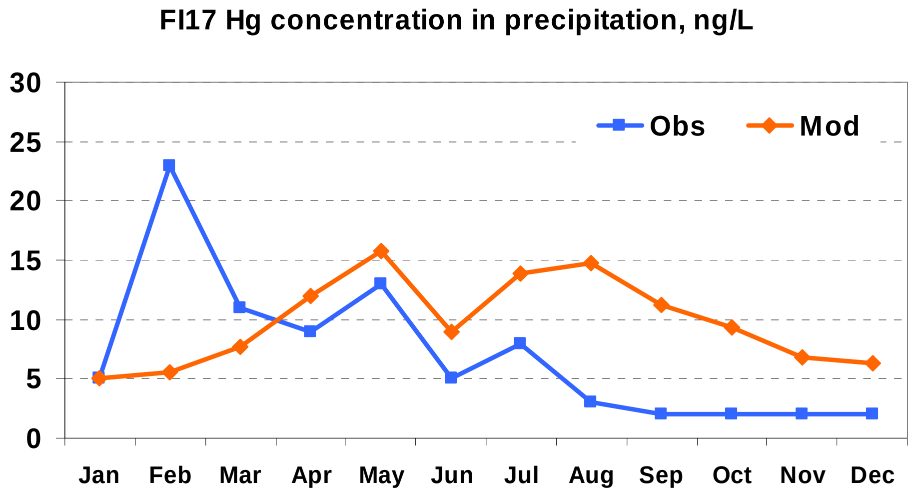


Figure 6.36. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Virolahty II (FI17). Units: ng/L.

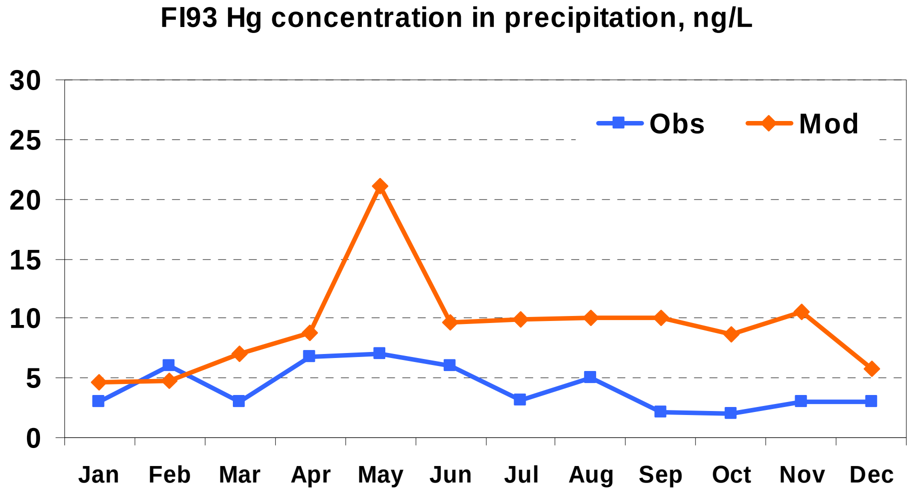


Figure 6.37. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Kotinen (FI93). Units: ng/L.

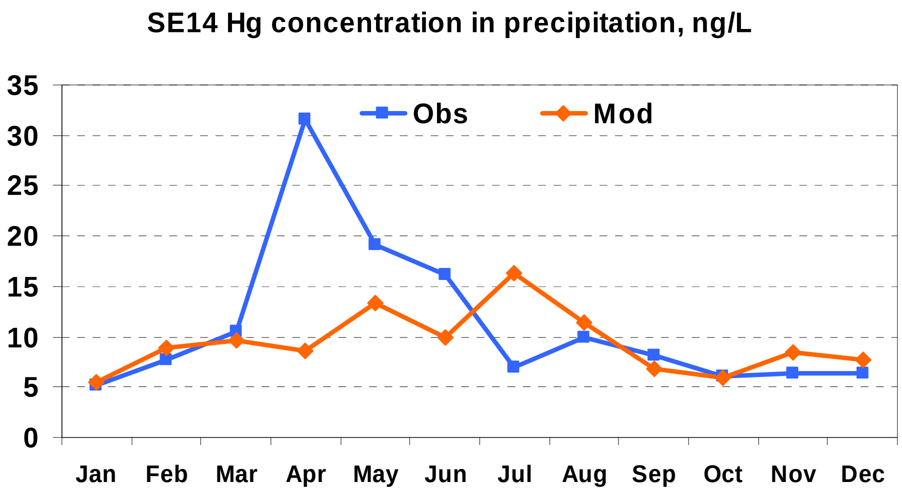


Figure 6.38. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Råö (SE14). Units: ng/L.

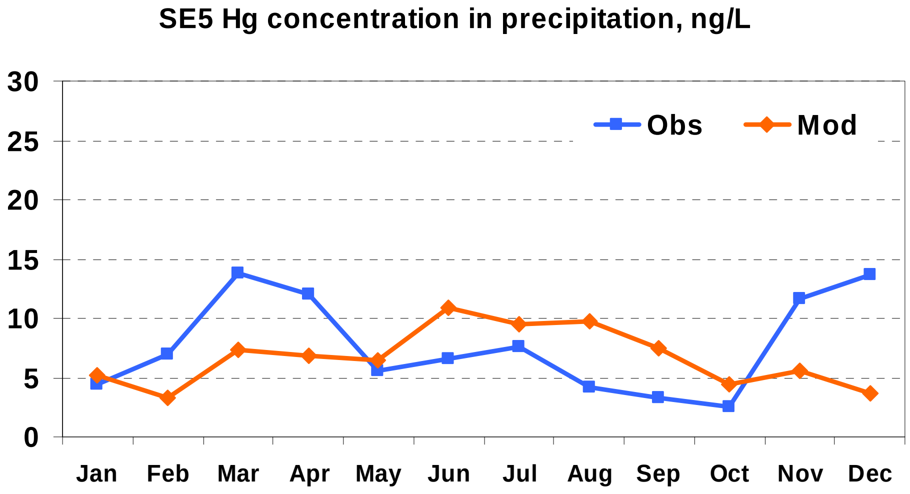


Figure 6.39. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Bredkälen (SE5). Units: ng/L.

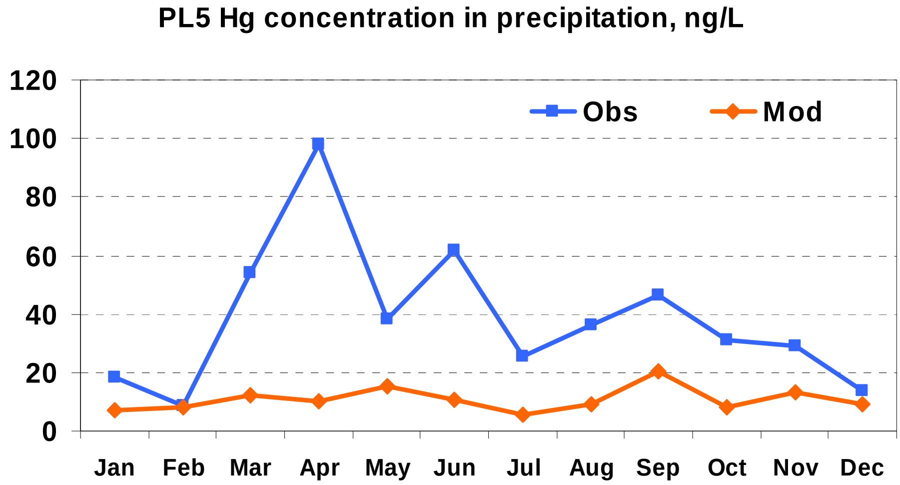


Figure 6.40. Comparison of calculated monthly mean Hg concentrations in precipitation for 2012 with measurements of the station Diabla Gora (PL5). Units: ng/L.

Modelled concentrations of mercury in air and in precipitation were compared with the measurement data of 6 monitoring sites around the Baltic Sea. It can be seen that that the model values generally agree with the measured concentrations. Some deviations between simulated and observed monthly mean concentrations of mercury can be explained by the uncertainties in seasonal variation of mercury emission used in modeling (anthropogenic and natural), differences between measured precipitation amount and the one used in the model, and difficulties in measurements of mercury.

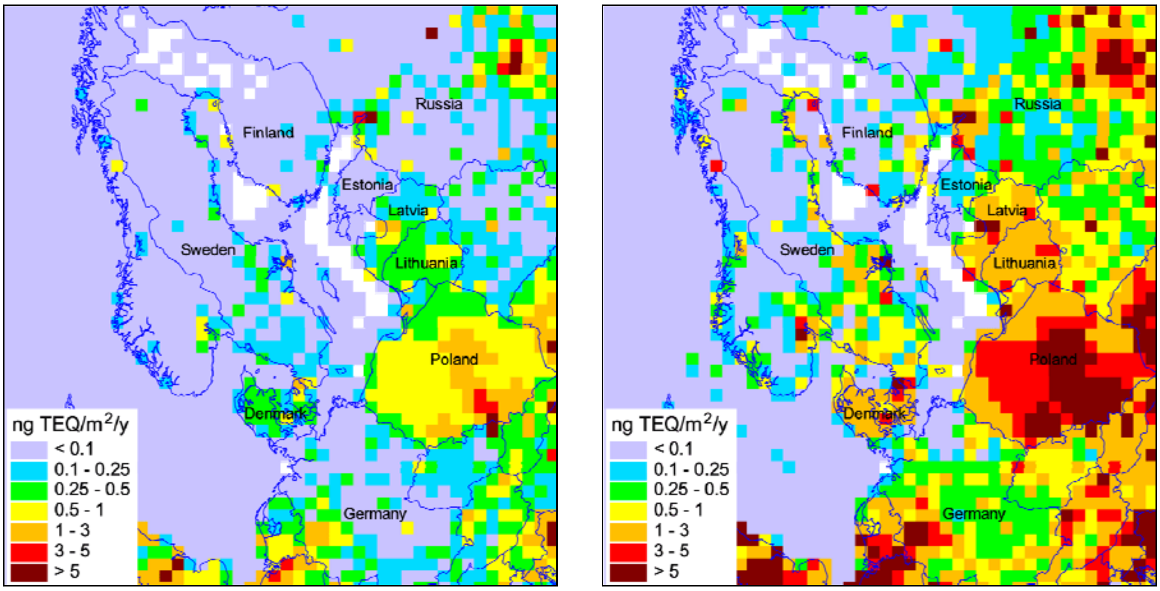
6.6 Concluding remarks

- Mercury emissions from HELCOM countries have decreased from 1990 to 2012 by 65%, whereas from 2011 to 2012 emissions have slightly increased by almost 2%.
- Annual deposition of mercury to the Baltic Sea has decreased from 1990 to 2012 by 31%. Mercury deposition in 2012 was higher comparing to 2011 by 10%.
- The contribution of anthropogenic sources of HELCOM countries to total mercury deposition over the Baltic Sea was estimated to 16%. Essential contribution belongs to the global and natural sources and re-emission (79%) and anthropogenic sources of other EMEP countries (5%).
- The most significant contribution to mercury deposition over the Baltic Sea was made by Poland (6%) and Germany (5%).
- Modelling results for mercury were generally within an accuracy of 25% in comparison to measured concentrations obtained around the Baltic Sea in 2012.

7. Atmospheric Supply of PCDD/Fs to the Baltic Sea in 2012

In this chapter the results of model evaluation of dioxins and furans (PCDD/Fs) atmospheric input to the Baltic Sea and its sub-basins for 2012 is presented. Modelling of PCDD/F atmospheric transport and deposition was carried out using MSC-E Eulerian Persistent Organic Pollutant transport model MSCE-POP (*Gusev et al.*, 2005). Latest available official information on PCDD/F emission from HELCOM countries and other European countries was used in model computations. Evaluation of PCDD/F contamination of the EMEP and the Baltic Sea regions is performed using two scenarios of emission data, namely, officially submitted PCDD/F emissions and scenario of adjusted PCDD/F emissions prepared by EMEP/MSC-E. Model simulations using official emission data underestimate observed levels of PCDD/F concentrations. The use of scenario with adjusted emissions obtained on the basis of developing inverse modelling approach and available measurements permit to obtain reasonable agreement of modelling results with observed PCDD/F pollution levels. Description of this approach and prepared scenario of PCDD/F emissions for the EMEP domain can be found in the EMEP Status Reports (*Shatalov et al.*, 2012; *Gusev et al.*, 2013). Based on these modelling results annual and monthly levels of PCDD/F deposition to the Baltic Sea have been obtained and contributions of HELCOM countries emission sources to the deposition over the Baltic Sea are estimated.

7.1 PCDD/Fs emissions



a) b)

Figure 7.1. Annual total anthropogenic emissions of **PCDD/F** in the Baltic Sea region for 2012 according to officially reported information by EMEP countries (a) and scenario of PCDD/F emissions prepared by EMEP/MSC-E (b), ng TEQ/m^2/y.

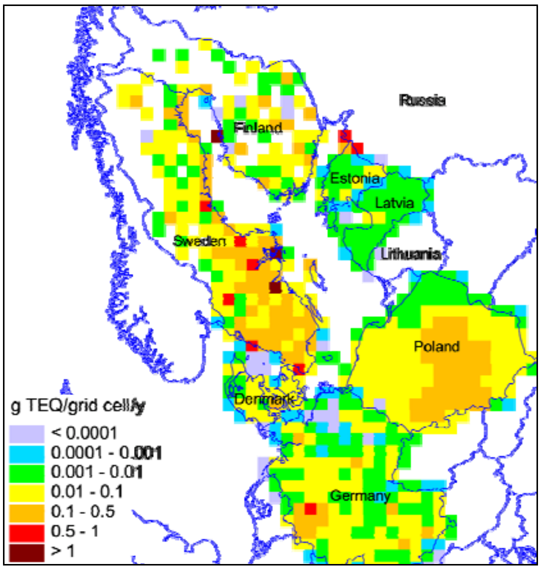


Figure 7.2. Annual PCDD/F emission from Public Power sector for 2012, g TEQ/grid cell/y (white color means no information).

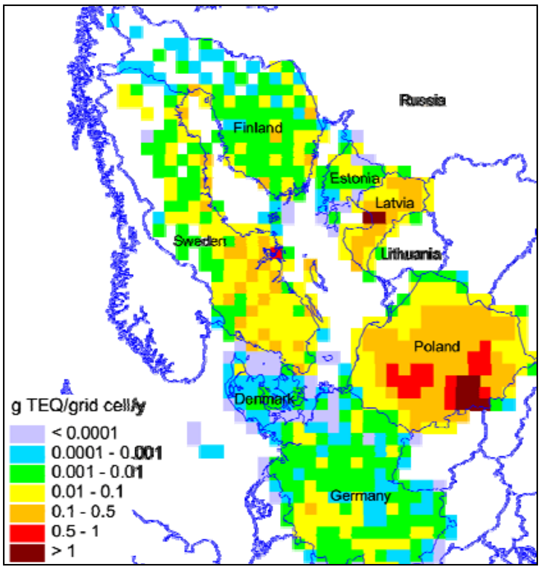


Figure 7.3. Annual PCDD/F emission from Industrial Combustion sector for 2012, g TEQ/grid cell/y (white color means no information).

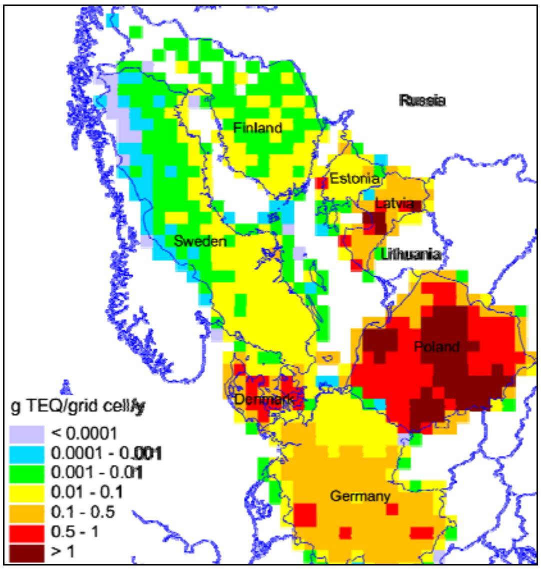


Figure 7.4. Annual PCDD/F emission from Small Combustion sector for 2012, g TEQ/grid cell/y (white color means no information).

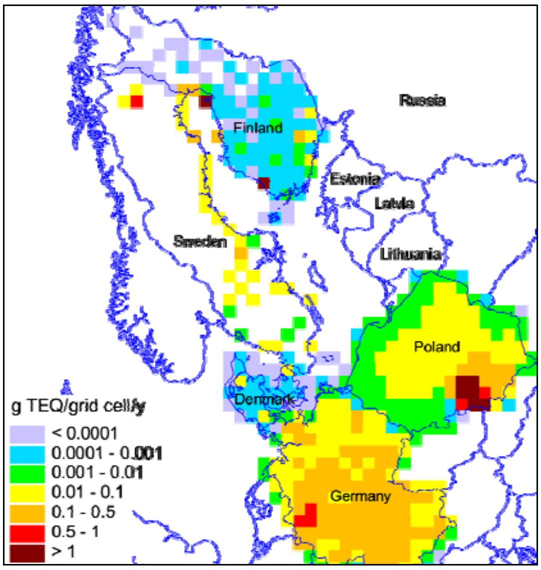


Figure 7.5. Annual PCDD/F emission from Industrial Processes sector for 2012, g TEQ/grid cell/y (white color means no information).

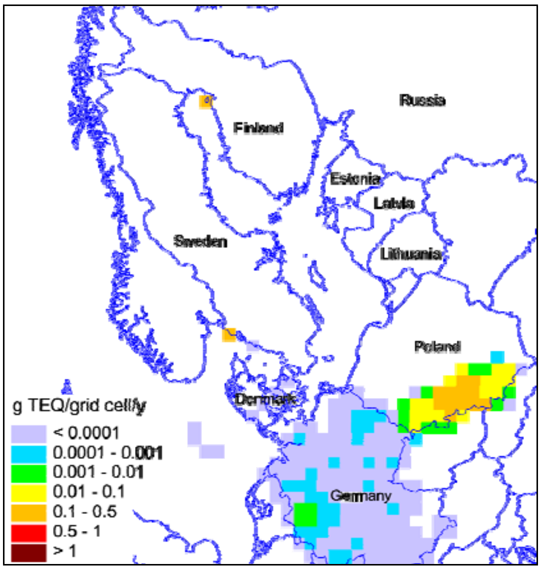


Figure 7.6. Annual PCDD/F emission from Fugitive Emissions sector for 2012, g TEQ/grid cell/y (white color means no information).

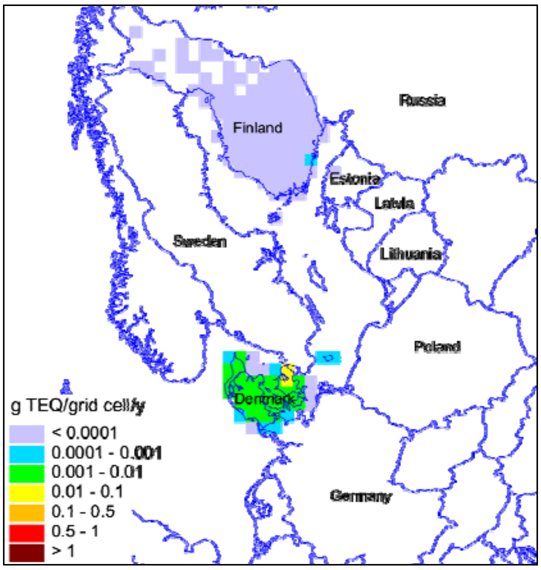


Figure 7.7. Annual PCDD/F emission from Solvents sector for 2012, g TEQ/grid cell/y (white color means no information).

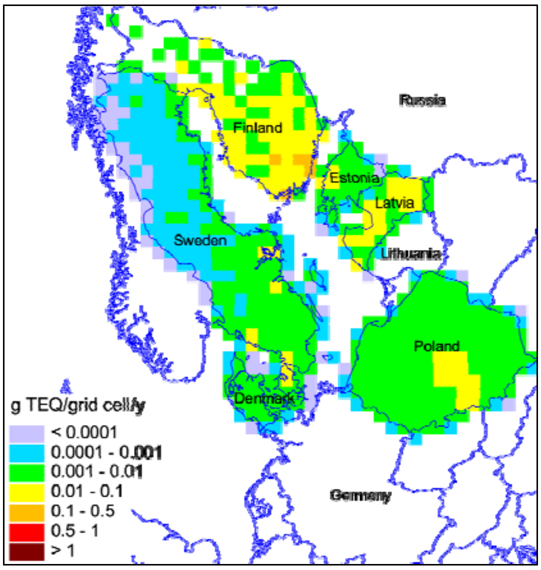


Figure 7.8. Annual PCDD/F emission from Road Rail sector for 2012, g TEQ/grid cell/y (white color means no information).

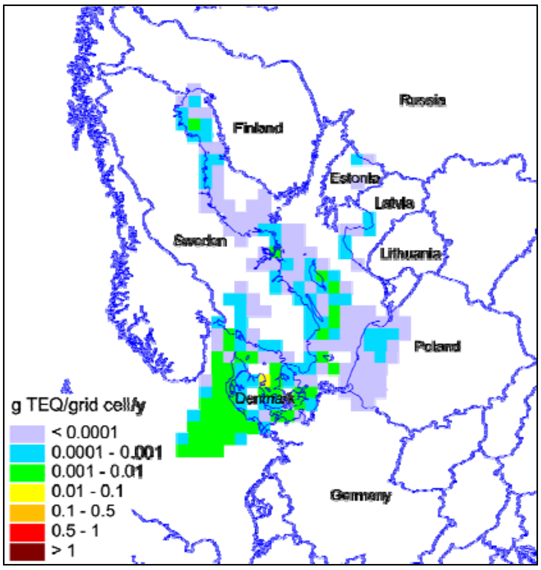


Figure 7.9. Annual PCDD/F emission from Shipping Emissions sector for 2012, g TEQ/grid cell/y (white color means no information).

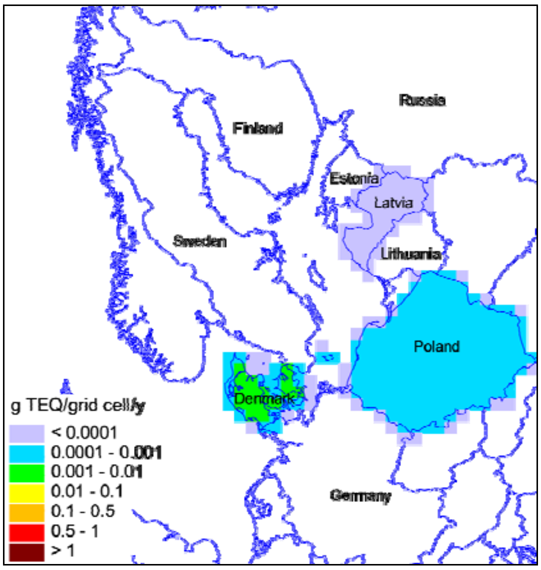


Figure 7.10. Annual PCDD/F emission from Off Road Mobility sector for 2012, g TEQ/grid cell/y (white color means no information).

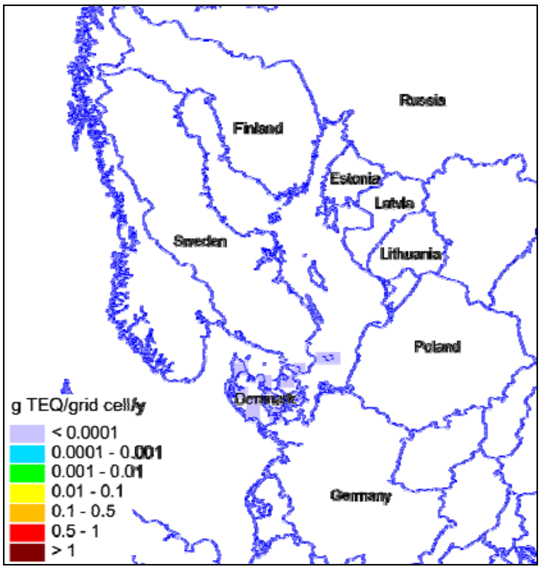


Figure 7.11. Annual PCDD/F emission from Civil Aviation sector for 2012, g TEQ/grid cell/y (white color means no information).

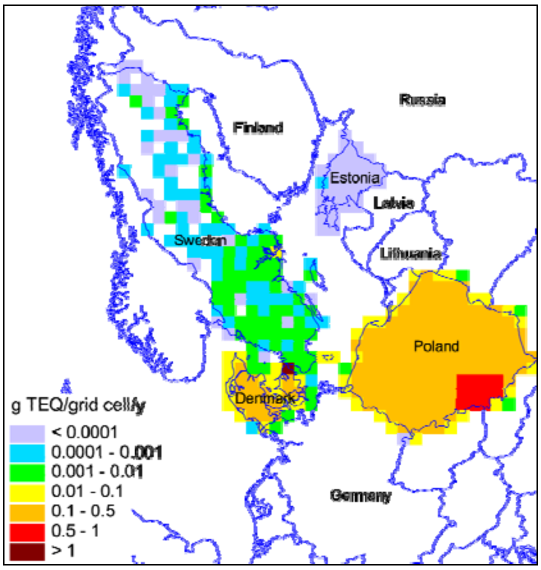


Figure 7.12. Annual PCDD/F emission from Other Waste Displacement sector for 2012, g TEQ/grid cell/y (white color means no information).

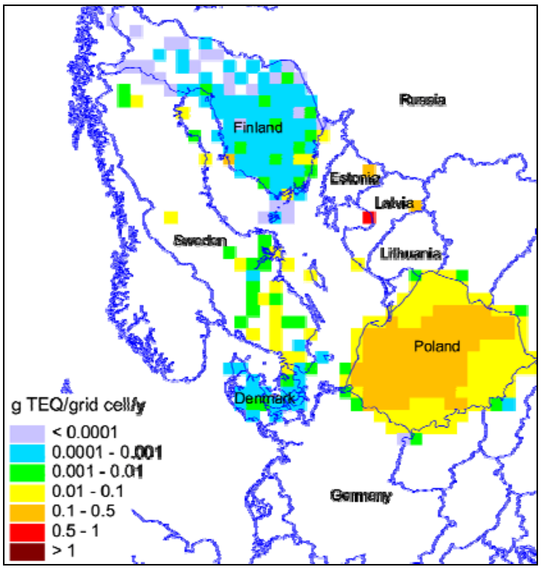


Figure 7.13. Annual PCDD/F emission from Waste Incineration sector for 2012, g TEQ/grid cell/y (white color means no information).

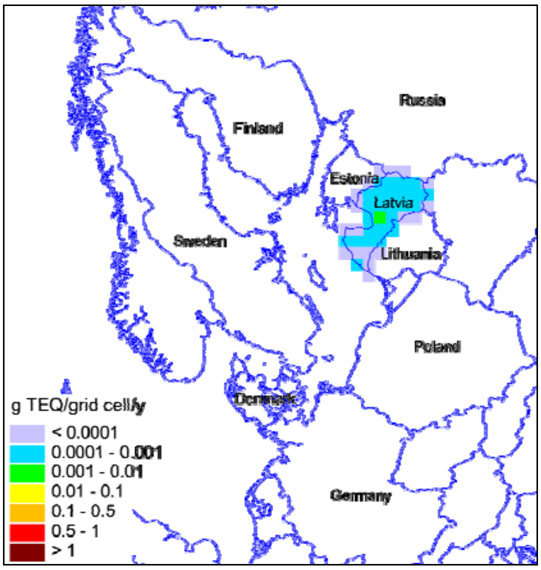


Figure 7.14. Annual PCDD/F emission from Agricultural Emissions sector for 2012, g TEQ/grid cell/y (white color means no information).

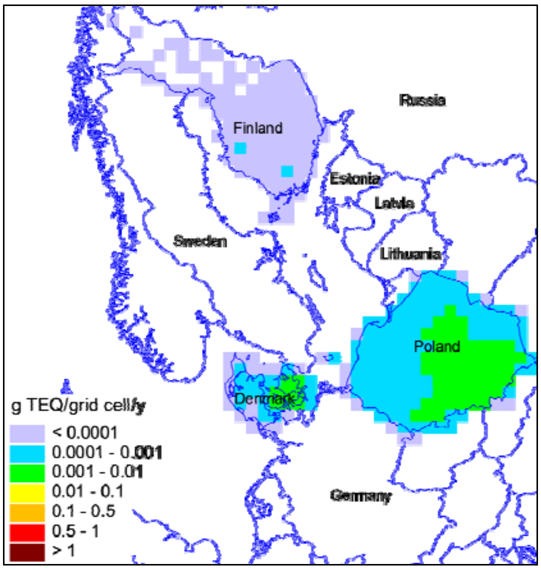


Figure 7.15. Annual PCDD/F emission from Agricultural Wastes sector for 2012, g TEQ/grid cell/y (white color means no information).

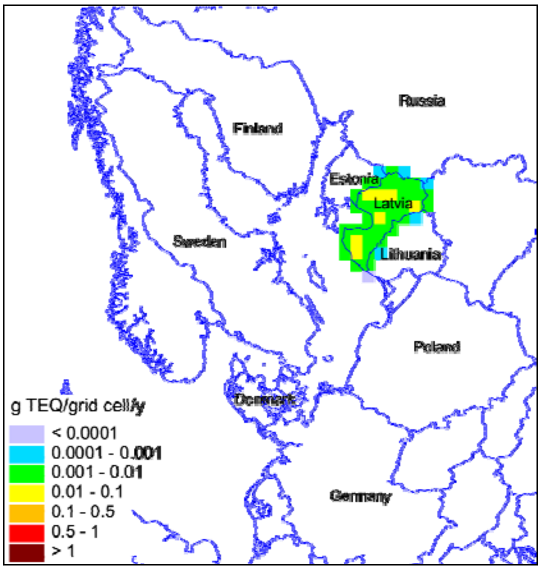


Figure 7.16. Annual PCDD/F emission from Other sector for 2012, g TEQ/grid cell/y (white color means no information).

Table 7.1. Annual total **PCDD/F** anthropogenic emissions of HELCOM countries from different sectors for 2012, in g TEQ/y

GNFR emission sector	**Sector name**	**DK**	**EE**	**FI**	**DE**	**LV**	**LT**	**PL**	**SE**
A	Public Power	1.25	1.70	3.89	5.84	0.31	0.75	13.0	21.4
B	Industrial Combustion	0.06	0.22	1.80	1.50	6.31	1.43	41.9	8.69
C	Small Combustion	15.3	1.69	1.44	28.3	23.4	21.6	138.6	3.94
D	Industrial Processes	0.05		3.42	25.7			14.8	2.05
E	Fugitive Emissions	9.4E-05		0.20	1.7E-02		0.0	2.67	0.25
F	Solvents	0.15	2.0E-06	1.3E-03	NA	NA		6.66	5.4E-06
G	Road Rail	0.11	0.14	2.69	2.46	0.47	0.15	0.75	0.5
H	Shipping emissions	0.15	2.5E-04		1.37	9.1E-04	6.2E-04	1.8E-03	3.2E-02
I	Off road mobility	4.3E-02		NA	0.41	2.4E-04	0.06	0.07	9.4E-04
J	Civil aviation	3.3E-04	NA	NA	NE	NA	NE	NA	NE
L	Other Waste Displacement	5.70	1.0E-03	NA	NA	NA		41.9	0.2
N	Waste Incineration	0.01	0.21	0.28	0.93	1.10	0.22	17.1	0.65
P	Agricultural emissions	NA	NA	NA	NA	1.1E-02		NA	
Q	Agricultural Wastes	2.5E-02	NE	0.003	NO	NA	NO	0.14	NO
R	Other	NO	NO	0.02	NO	0.27	NO	NA	NO
Total		22.9	4.0	13.8	66.6	31.9	24.2	277.6	37.8

NO – not occurring, an activity or process does not exist within a country.
NA – not applicable, the process or activity exists but emissions are considered never to occur.
NE – not estimated, emissions occur but have not been estimated or reported in this submission.
IE – included elsewhere, emissions by sources of compounds are estimated but included elsewhere in the inventory.

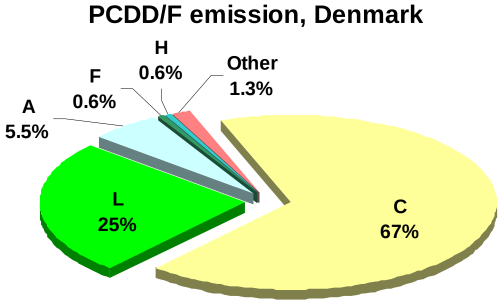


Figure 7.17. Contributions of different sectors to total annual PCDD/F emission of Denmark in 2012

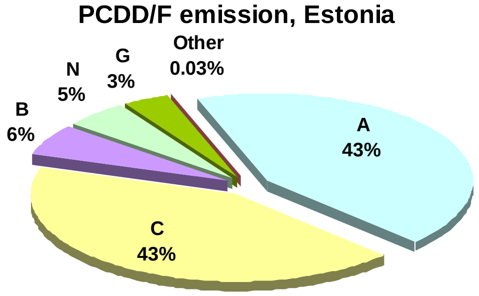


Figure 7.18. Contributions of different sectors to total annual PCDD/F emission of Estonia in 2012

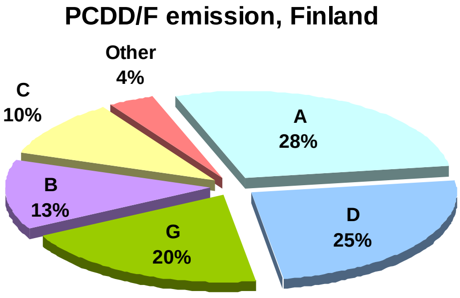


Figure 7.19. Contributions of different sectors to total annual PCDD/F emission of Finland in 2012

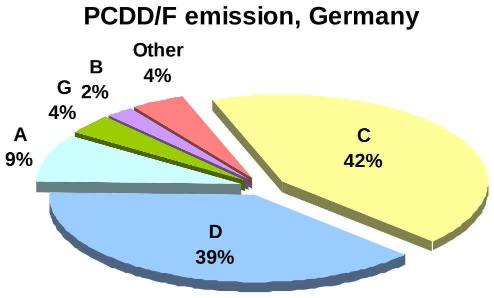


Figure 7.20. Contributions of different sectors to total annual PCDD/F emission of Germany in 2012

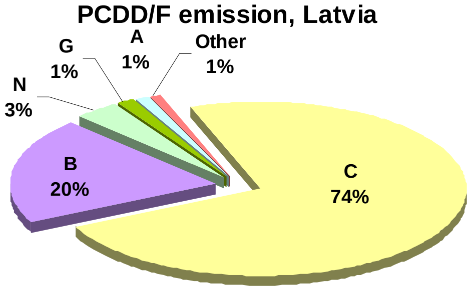


Figure 7.21. Contributions of different sectors to total annual PCDD/F emission of Latvia in 2012

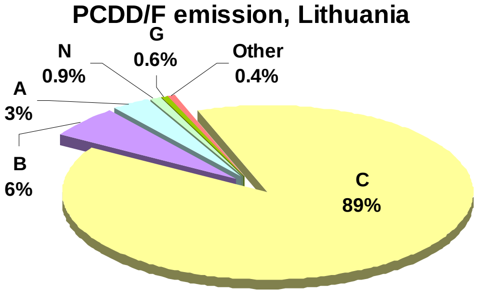
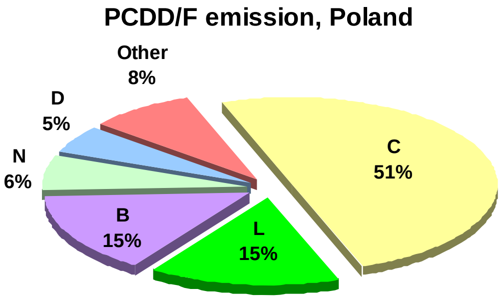


Figure 7.22. Contributions of different sectors to total annual PCDD/F emission of Lithuania in 2012



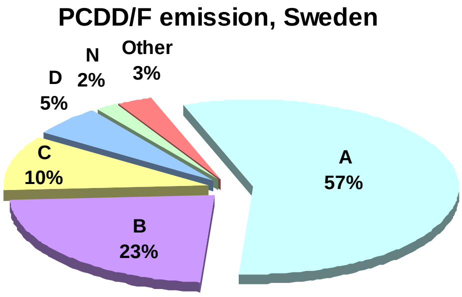
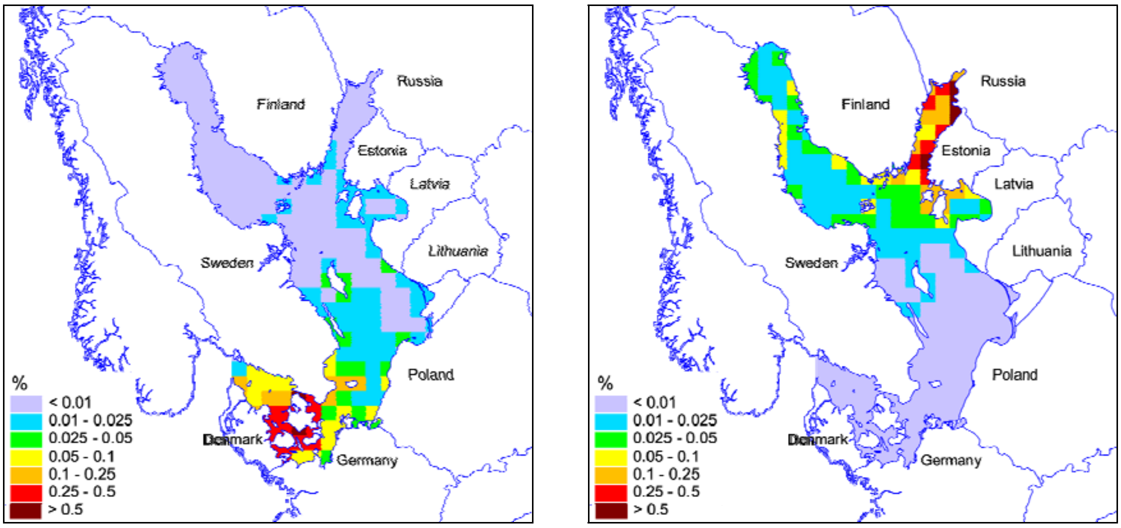
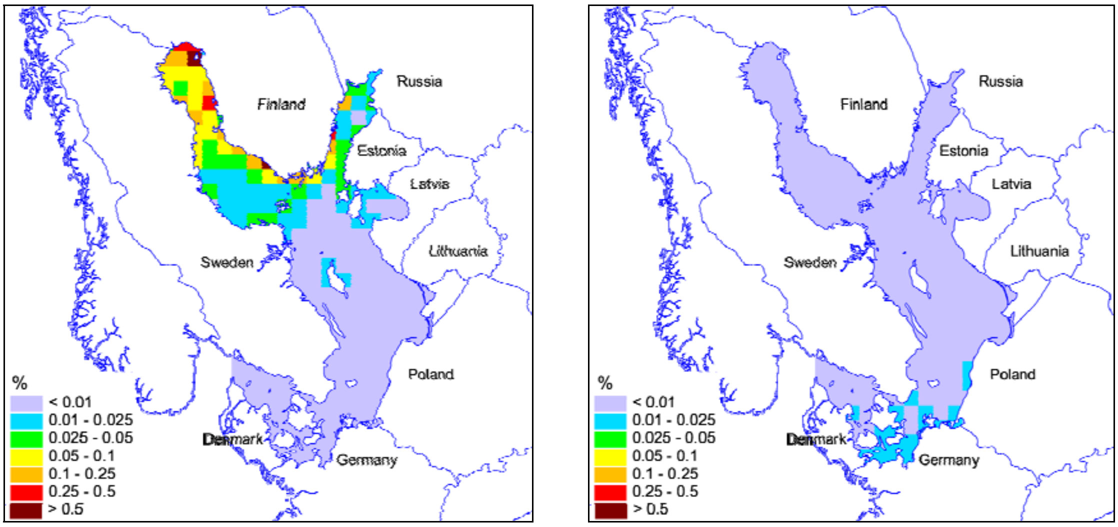


Figure 7.23. Contributions of different sectors to total annual PCDD/F emission of Poland in 2012

Figure 7.24. Contributions of different sectors to total annual PCDD/F emission of Sweden in 2012

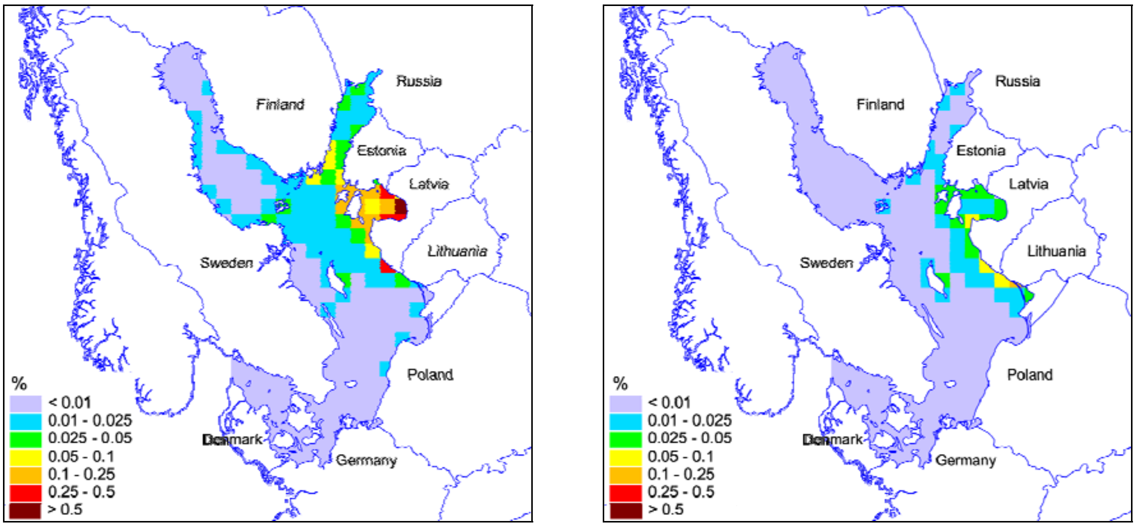


Denmark Estonia

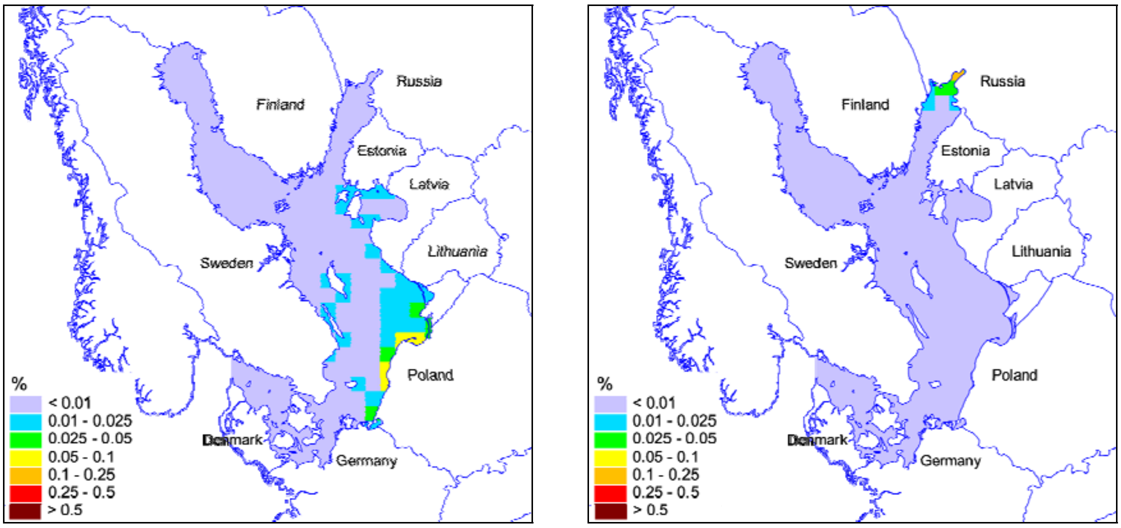


Finland Germany

Figure 7.25. Fractions of annual anthropogenic PCDD/F emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

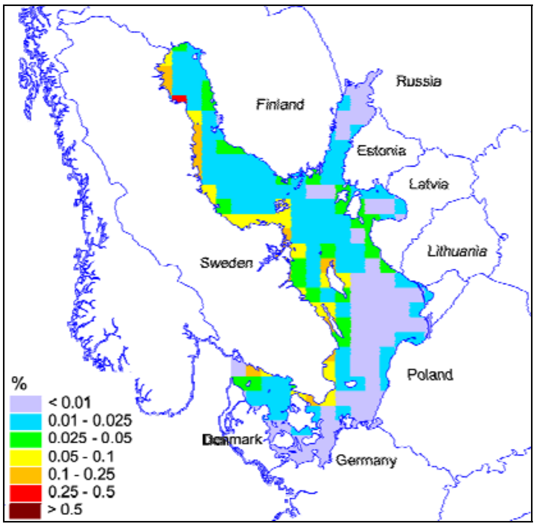


Latvia Lithuania



Poland Russia

Figure 7.25. (cont.) Fractions of annual anthropogenic PCDD/F emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).



Sweden

Figure 7.25. (cont.) Fractions of annual anthropogenic PCDD/F emissions of HELCOM Parties deposited to the Baltic Sea in 2012 (expressed as a percent of national anthropogenic emission deposited to the particular grid cells).

Table 7.2. Annual total anthropogenic emissions of **PCDD/Fs** of HELCOM countries and other EMEP countries in period 1990-2012, g TEQ/y (Expert estimates of emissions are shaded)

	1990	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**
DK	67	65	60	54	50	49	46	41	32	29	32	31	27	31	25	25	26	29	28	26	26	24	23
EE	5,7	5,4	4,3	3,6	3,8	4,5	4,9	4,8	3,8	3,5	3,4	3,5	3,8	4,1	3,8	3,4	2,8	4,9	5,2	4,9	5,5	5,4	4
FI	37	34	30	31	37	37	33	33	33	34	34	32	33	31	30	12	12	12	14	11	15	13	14
DE	747	622	493	364	244	230	206	197	184	166	152	131	110	93	69	69	71	71	72	62	70	64	67
LV	27	31	28	30	29	29	30	29	28	28	28	33	32	33	34	32	32	31	29	31	31	32	32
LT	24	26	14	18	17	17	19	18	19	19	20	20	21	22	22	22	24	22	24	24	25	24	24
PL	371	371	371	371	371	371	365	350	306	300	278	279	277	273	266	281	279	273	274	261	280	276	278
RU	991	947	901	878	825	769	637	614	606	625	631	643	655	686	716	747	778	808	839	869	900	900	900
SE	60	53	50	47	44	40	39	37	35	35	33	34	34	34	37	39	38	36	38	37	42	39	38
Total	**2 330**	**2 154**	**1 951**	**1 796**	**1 621**	**1 546**	**1 380**	**1 324**	**1 248**	**1 239**	**1 211**	**1 206**	**1 192**	**1 206**	**1 203**	**1 231**	**1 262**	**1 287**	**1 323**	**1 326**	**1 394**	**1 376**	**1 379**
AL	26	28	19	18	22	19	19	19	19	18	19	19	20	24	32	30	25	28	30	32	32	32	32
AM	2,1	2,1	2,1	2,1	2,1	2,1	2,1	2,1	2,1	1,8	28,4	5,5	5	4,6	4,2	3,7	3,3	2,8	2,8	2,8	2,8	2,8	2,8
AT	161	135	77	67	56	59	60	59	56	54	52	53	40	40	40	43	40	38	39	36	40	35	38
AZ	98	98	98	98	98	98	98	98	98	98	98	99	100	101	102	102	103	104	105	106	107	107	108
BY	22	21	19	16	14	13	14	17	18	16	24	30	23	28	25	24	27	27	34	37	36	36	34
BE	542	506	463	391	413	362	323	280	227	178	113	75	64	64	65	57	53	54	66	47	52	45	52
BA	67	67	67	67	67	67	67	67	67	67	67	65	63	61	59	57	56	54	52	50	48	46	44
BG	74	64	84	86	96	97	99	98	104	89	112	91	95	113	124	112	149	93	76	47	45	53	59
HR	155	99	85	81	79	83	106	103	94	87	99	73	80	101	95	91	92	79	83	88	98	117	123
CY	1,8	1,8	1,9	2	2	2	2	2,1	2,1	2,1	2,1	2,2	2,2	0,932	0,543	0,579	0,602	0,624	0,61	0,588	0,562	0,567	0,573
CZ	1252	1220	1220	1140	1135	1135	922	830	767	643	744	620	177	114	187	179	175	169	150	141	134	104	45
FR	1746	1795	1823	1882	1885	1684	1469	1037	934	608	515	383	355	234	314	193	120	117	103	88	99	92	81
GE	122	122	122	122	122	122	122	122	122	122	122	122	111	98	85	85	85	85	85	85	85	85	85
GR	279	279	279	279	279	279	279	279	279	279	279	255	231	207	183	159	135	111	87	63	40	16	16
HU	182	175	132	129	124	116	113	109	98	99	97	98	98	99	97	95	93	85	88	75	44	48	42
IS	13	12	12	11	11	9,3	8,3	7,7	6,4	5,2	4,7	4	3,6	2,9	2,3	1,6	1,7	1,7	2,4	2,2	2,1	1,3	1,3
IE	26	26	25	25	25	25	26	24	21	22	23	24	30	35	27	23	23	16	16	16	16	15	15
IT	458	474	457	432	424	442	406	411	400	378	362	284	275	273	277	283	291	304	294	220	228	235	222
KZ	40	40	40	40	40	40	40	40	40	40	40	40	41	41	41	42	42	42	43	43	43	44	44
LI	0,015	0,014	0,013	0,012	0,011	0,011	0,01	0,01	0,01	0,009	0,009	0,009	0,009	0,009	0,009	0,009	0,009	0,009	0,009	0,01	0,01	0,01	0,01
LU	46	46	46	47	39	31	24	16	8,4	7	5,7	4,4	3,1	1,8	1,7	1,6	1,5	1,4	1,3	1,2	1,1	1,1	1,1
MT	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	8,6	0,971	1
MC	2,4	2,4	2,6	2,9	3,1	3,1	3,3	3,8	3,5	3,6	3,7	3,9	3,5	2,9	2,6	2,6	1,9	2,6	2,5	2,3	2,2	2,4	2,4
NL	743	641	498	355	212	66	58	52	45	37	30	30	29	28	28	28	27	27	30	29	30	30	23
NO	126	95	93	93	91	68	47	38	33	37	33	32	31	28	31	23	23	23	22	21	25	23	22
PT	54	52	52	51	50	51	52	52	49	48	46	43	41	40	39	34	33	32	31	31	29	30	29
MD	14	11	6,9	5,5	5,1	3	3,4	2,9	6,4	2,4	2,4	2,2	2,5	3,9	5,2	5,5	5,5	6	5,8	5,2	3,9	5,2	5,2
RO	1537	1253	1202	1152	1101	1051	965	879	793	707	622	409	355	300	245	174	120	131	152	142	157	163	169
RS	27	22	29	23	23	20	24	20	25	19	23	16	21	22	21	24	24	25	25	20	20	21	17
ME	2,6	2,4	2,3	2,1	1,5	1,7	1,9	2,1	1,8	1,7	1,7	1,7	2	2,4	2,5	2,6	2,7	2,8	2,9	2,6	2,8	2,8	2,8
SK	169	165	161	157	153	150	142	134	167	132	128	86	72	56	58	59	53	50	46	32	46	43	49
SI	16	16	15	13	12	12	12	12	11	11	11	10	10	10	10	10	10	10	10	10	10	11	11
ES	181	185	193	191	184	157	155	119	117	121	126	116	116	119	120	117	121	126	121	114	119	122	120
CH	201	185	173	159	143	126	117	106	98	79	68	56	41	28	27	26	25	23	22	20	20	18	18
MK	15	15	14	14	14	14	13	13	13	13	13	12	12	12	12	11	11	11	11	11	10	10	9,9
TR	1 012	1 012	1 012	1 012	1 012	1 012	1 012	1 012	1 012	1 012	1 012	1 018	1 024	1 029	1 035	1 041	1 047	1 053	1 058	1 064	1 070	1 076	1 082
UA	1 022	1 022	1 022	1 022	1 022	1 022	1 022	1 022	1 022	1 022	1 022	1 024	1 026	1 027	1 029	1 030	1 032	1 033	1 035	1 037	1 038	1 040	1 041
UK	1 304	1 276	1 227	1 035	815	842	570	522	425	388	315	306	277	271	283	247	225	209	216	203	205	205	212
EMEP	**14 077**	**13 331**	**12 735**	**12 026**	**11 405**	**10 839**	**9 784**	**8 945**	**8 444**	**7 697**	**7 481**	**6 730**	**6 082**	**5 841**	**5 922**	**5 658**	**5 548**	**5 473**	**5 480**	**5 258**	**5 344**	**5 296**	**5 238**

Expert estimates:

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- *Berdowski J,J,M,, Baas J,, Bloos J,P,J,, Visschedijk A,J,H,, Zandveld P,Y,J, [1997] The European Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990, TNO Institute of Environmental Sciences, Energy Research and Process Innovation, UBA-FB report 104 02 672/03*

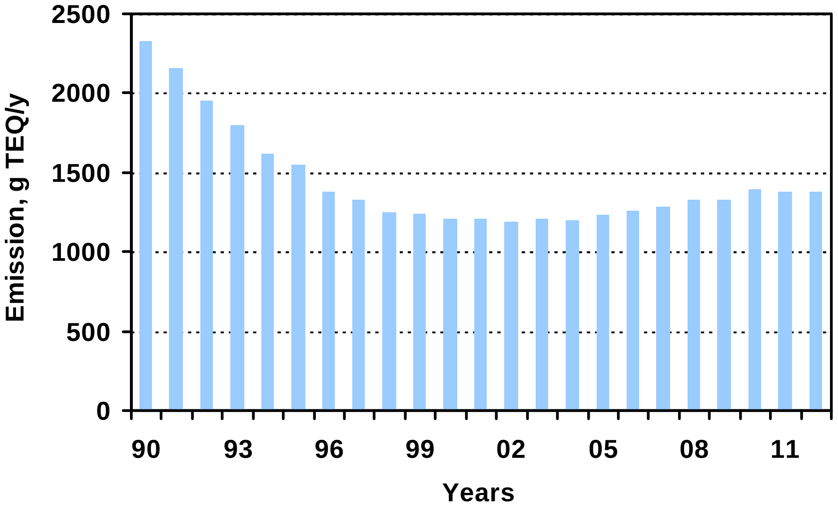
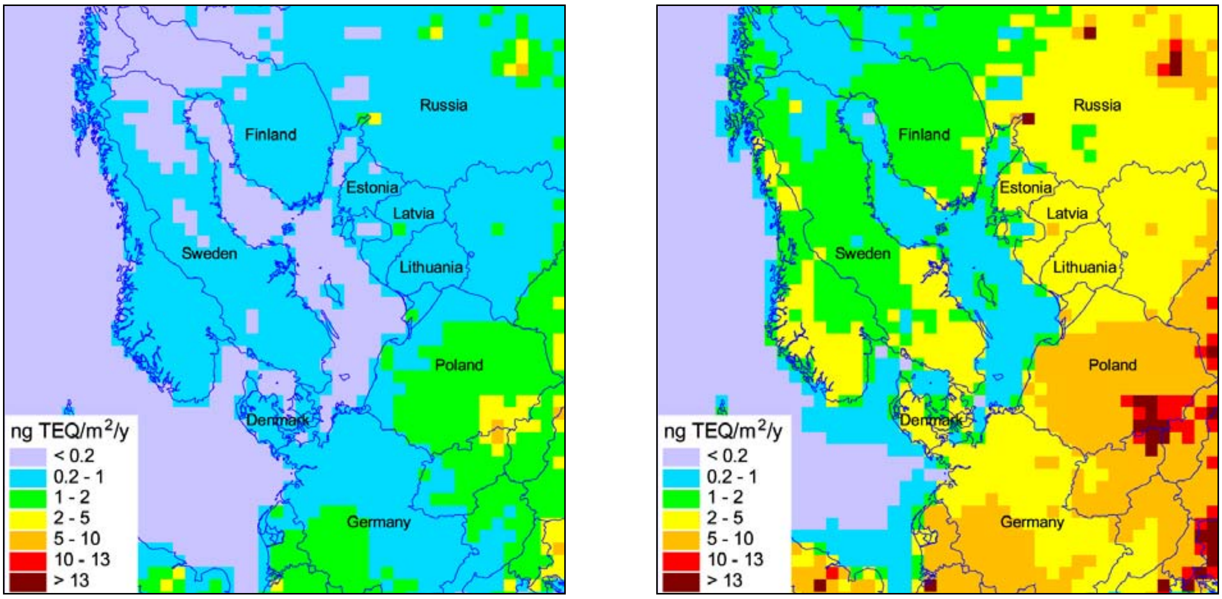


Figure 7,26, Time-series of total annual **PCDD/F** emissions of HELCOM countries for 1990-2012 based on officially reported emission data, g TEQ/year,

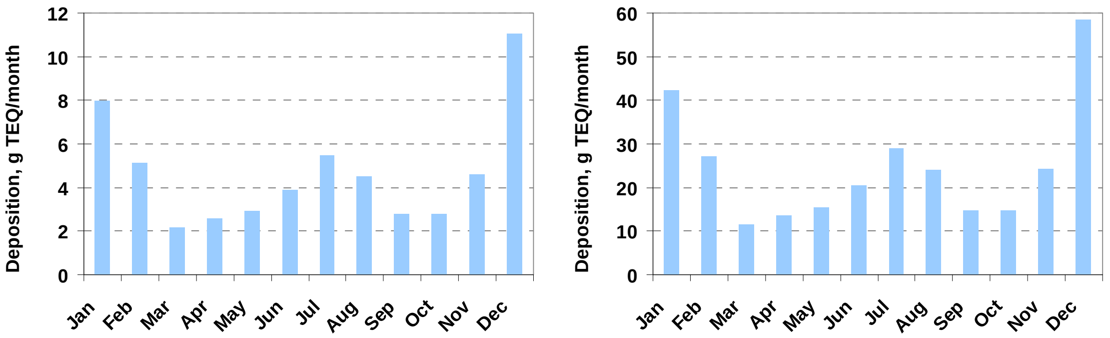
7.2 Annual total deposition of PCDD/F



a) b)

Figure 7,27, Annual total deposition fluxes of **PCDD/Fs** over the Baltic Sea region for 2012 calculated on the basis of official emissions (a) and scenario emissions (b), ng TEQ/m^2/y,

7,3 Monthly total deposition of PCDD/Fs



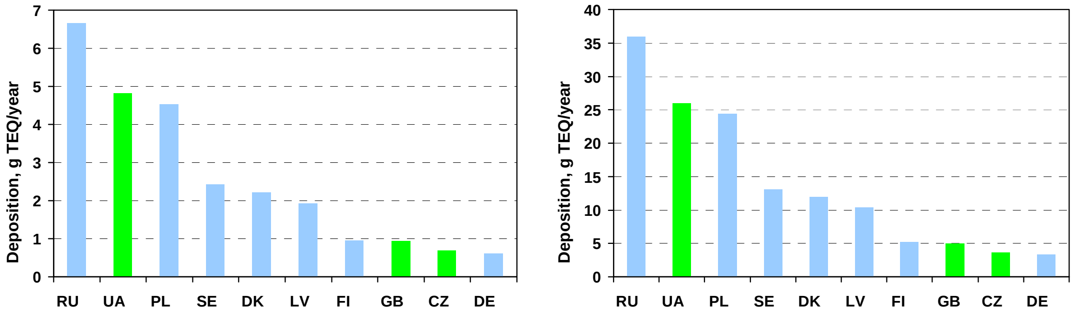
a) b)

Figure 7,28, Monthly total deposition of PCDD/Fs over the Baltic Sea for 2012 calculated on the basis of official emissions (a) and scenario emissions (b), g TEQ/month,

Table 7,3, Monthly total deposition of PCDD/Fs over the Baltic Sea for 2012 calculated on the basis of official emissions and scenario emissions, g TEQ/month,

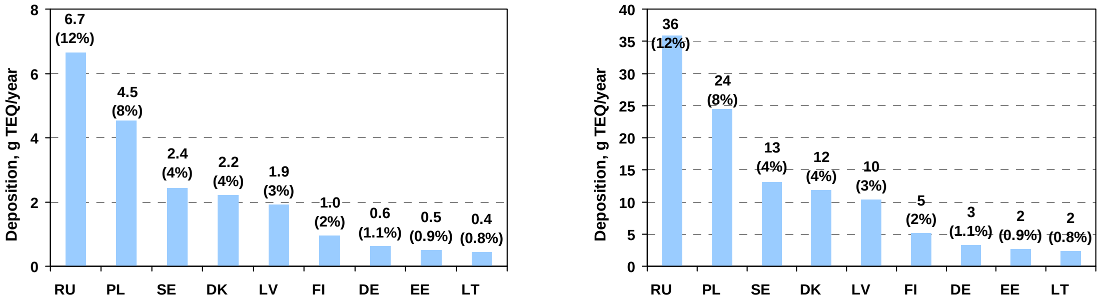
Month	**PCDD/F deposition (Official Emissions)**	**PCDD/F deposition (Scenario Emissions)**
Jan	8,0	42,2
Feb	5,1	27,2
Mar	2,2	11,5
Apr	2,6	13,6
May	2,9	15,4
Jun	3,9	20,6
Jul	5,5	29,1
Aug	4,5	23,9
Sep	2,8	14,7
Oct	2,8	14,9
Nov	4,6	24,3
Dec	11,0	58,5

7,4 Source allocation of PCDD/F deposition



a) b)

Figure 7,29, Top ten countries with the highest contribution to annual total deposition of PCDD/Fs over the Baltic Sea for 2012 calculated on the basis of official emissions (a) and scenario emissions (b), g TEQ/y,



a) b)

Figure 7,30, Contributions (in g TEQ/y and in %) of HELCOM countries to annual total PCDD/F deposition to the Baltic Sea for 2012 calculated on the basis of official emissions (a) and scenario emissions (b), HELCOM countries emissions of PCDD/Fs contributed 36% to total PCDD/F deposition over the Baltic Sea in 2012, Contribution of other EMEP countries accounted for 20%, Significant contribution was made by other emission sources, in particular, global emissions sources and re-emission of PCDD/Fs (44%),

Table 7,4, Two most significant contributors to annual total deposition of PCDD/Fs to the nine Baltic Sea sub-basins for 2012,

Sub-basin	Country (1)	%	Country (2)	%	*, %
ARC	Ukraine	15	Russia	12	39
BOB	Russia	16	Finland	12	34
BOS	Ukraine	14	Russia	12	37
BAP	Poland	14	Ukraine	9	46
GUF	Russia	39	Ukraine	8	32
GUR	Latvia	19	Ukraine	12	41
KAT	Denmark	15	Sweden	6	59
SOU	Denmark	21	Sweden	9	59
WEB	Denmark	16	Poland	3	66
BAS	Russia	12	Ukraine	9	44

* - contribution of re-emission and remote sources,

7,5 Comparison of model results with measurements

PCDD/Fs are not regularly measured by the EMEP monitoring network, Evaluation of modelling results on PCDD/Fs against measurements was performed in framework of the studies of EMEP region pollution by dioxins and furans (*Shatalov et al*,, 2012; *Gusev et al*,, 2013), For this purpose available measurements made by various national and international campaigns reported in literature were used, It was found that the agreement between calculated and measured total PCDD/F toxicities was within a factor of two for more than 50% of available measurements at background locations, More detailed information on the comparison of model estimates and observed PCDD/F concentrations can be found in the EMEP Status Reports (*Shatalov et al*,, 2012; *Gusev et al*,, 2013),

7,6 Concluding remarks

- PCDD/F emissions from HELCOM countries have decreased from 1990 to 2012 by 41%, Emission of dioxins and furans emission from 2011 to 2012 has slightly increased,
- Annual PCDD/F deposition to the Baltic Sea has decreased from 1990 to 2012 by 60%, Level of PCDD/F deposition in 2012 has increased comparing to 2011 by about 9%,
- The contribution of anthropogenic sources of HELCOM countries to total PCDD/F deposition over the Baltic Sea was estimated to approximately 36%, Essential contribution belongs to the anthropogenic sources of other EMEP countries (20%) and other sources of emission including re-emission and global sources (44%),
- The most significant contribution to dioxins and furans deposition over the Baltic Sea in 2012 was made by Russia (12%) and Ukraine (9%),

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Appendix A: Tables with measurements available at HELCOM stations for 2012

Table A.1 Monthly and annual mean concentrations of nitrogen components in air.

Site	Component	Matrix	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
DE0009R	nitrogen_dioxide	air	µg N /m3	2.19	2.13	3.15	2.08	2.10	1.29	1.57	1.55	1.58	1.84	2.85	3.00	2.12
DK0005R	nitrogen_dioxide	air	µg N /m3	2.16	2.03	2.85	2.61	2.72	1.64	1.93	2.19	1.59	2.46	3.92	3.09	2.44
DK0008R	nitrogen_dioxide	air	µg N /m3	-	-	2.58	1.59	2.08	1.18	1.15	1.24	0.83	1.70	2.20	1.56	1.55
DK0012R	nitrogen_dioxide	air	µg N /m3	2.99	3.41	2.89	2.49	1.93	1.65	1.55	2.14	2.02	3.45	4.32	4.40	2.80
EE0009R	nitrogen_dioxide	air	µg N /m3	1.48	1.67	1.15	0.89	0.56	0.53	0.44	0.42	0.38	0.47	0.68	1.94	0.89
EE0011R	nitrogen_dioxide	air	µg N /m3	0.75	1.23	1.40	0.73	0.84	0.59	0.48	0.43	0.45	0.58	0.73	1.26	0.78
FI0009R	nitrogen_dioxide	air	µg N /m3	0.79	1.05	1.03	1.22	1.30	1.06	0.94	0.56	0.57	0.50	0.87	1.43	0.94
FI0017R	nitrogen_dioxide	air	µg N /m3	2.89	3.19	1.57	1.32	1.28	1.06	1.06	0.71	0.69	0.99	0.97	3.55	1.59
FI0037R	nitrogen_dioxide	air	µg N /m3	1.80	1.54	0.62	0.52	0.40	0.23	0.21	0.20	0.34	0.47	0.64	1.89	0.74
LT0015R	nitrogen_dioxide	air	µg N /m3	1.32	1.65	1.40	1.16	0.94	0.63	0.83	0.71	0.62	0.85	1.56	1.56	1.08
LV0010R	nitrogen_dioxide	air	µg N /m3	1.06	1.26	0.96	0.60	0.56	0.46	0.53	0.29	0.54	0.77	0.99	1.50	0.79
PL0004R	nitrogen_dioxide	air	µg N /m3	1.96	2.07	1.57	1.29	1.39	1.12	0.90	0.95	0.91	1.43	2.10	2.58	1.54
SE0005R	nitrogen_dioxide	air	µg N /m3	0.26	0.25	0.08	0.07	0.06	0.09	0.06	0.07	0.05	0.07	0.14	0.24	0.12
SE0011R	nitrogen_dioxide	air	µg N /m3	1.40	1.90	2.05	1.01	0.90	0.84	0.76	1.04	1.18	1.45	2.51	1.27	1.37
SE0012R	nitrogen_dioxide	air	µg N /m3	0.86	1.14	0.51	0.50	0.45	0.33	0.35	0.33	0.39	0.53	0.63	1.22	0.60
SE0014R	nitrogen_dioxide	air	µg N /m3	1.19	1.80	1.82	1.10	1.38	0.94	0.81	0.87	0.68	1.31	1.56	1.62	1.26
DE0009R	nitrate	pm25	µg N /m3	0.40	0.89	1.86	0.82	0.34	0.08	0.04	0.11	0.08	0.33	1.06	0.68	0.55
FI0009R	nitrate	pm25	µg N /m3	0.04	0.05	0.20	0.13	0.12	0.10	0.05	0.04	0.07	0.08	0.11	0.08	0.09
FI0017R	nitrate	pm25	µg N /m3	0.09	0.07	0.05	0.04	0.02	0.03	0.02	0.01	0.02	0.03	0.03	0.04	0.04
LV0010R	nitrate	pm25	µg N /m3	0.04	-	-	-	-	-	0.03	0.02	0.03	0.03	0.06	0.73	(0.14)
PL0004R	nitrate	aerosol	µg N /m3	0.38	0.51	1.01	0.51	0.32	0.26	0.24	0.25	0.25	0.39	0.75	0.69	0.46
DK0003R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.53	0.64	1.06	0.85	0.69	0.48	0.44	0.52	0.36	0.44	0.99	0.74	0.66
DK0008R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.38	0.51	1.04	0.76	0.70	0.59	0.62	0.70	0.45	0.58	0.97	0.47	0.65
DK0012R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.57	0.52	1.55	1.11	0.82	0.65	0.67	0.83	0.75	0.74	1.15	0.86	0.86
FI0009R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.24	0.32	0.30	0.35	0.35	0.34	0.35	0.28	0.30	0.30	0.33	0.39	0.32
FI0017R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.31	0.38	0.20	0.22	0.17	0.14	0.25	0.15	0.17	0.16	0.18	0.32	0.22
FI0037R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.27	0.26	0.11	0.13	0.10	0.03	0.08	0.07	0.10	0.10	0.13	0.22	0.13
LT0015R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.59	0.90	1.07	0.52	0.37	0.43	0.38	0.44	0.53	0.58	0.98	1.10	0.63
PL0004R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.46	0.63	1.24	0.62	0.43	0.32	0.28	0.32	0.33	0.47	0.83	0.77	0.56
SE0005R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.04	0.04	0.04	0.04	0.04	0.03	0.03	0.02	0.02	0.01	0.02	0.04	0.03
SE0011R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.30	0.26	0.84	0.53	0.38	0.29	0.30	0.33	0.37	0.40	0.65	0.31	0.41
SE0012R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.15	0.17	0.28	0.24	0.22	0.18	0.20	0.14	0.15	0.15	0.22	0.14	0.19
SE0014R	sum_nitric_acid_and_nitrate	air+aerosol	µg N /m3	0.27	0.33	0.72	0.53	0.51	0.47	0.46	0.41	0.33	0.39	0.69	0.20	0.44
DK0003R	ammonia	air	µg N /m3	0.42	0.56	2.51	1.69	1.23	0.77	0.87	1.01	0.62	0.40	0.35	0.29	0.90
DK0008R	ammonia	air	µg N /m3	0.04	0.04	0.53	0.26	0.32	0.20	0.21	0.25	0.19	0.09	0.07	0.01	0.18
DK0012R	ammonia	air	µg N /m3	0.14	0.30	2.54	1.25	0.95	0.62	0.69	1.08	0.76	0.49	0.23	0.06	0.78
DE0009R	ammonium	pm25	µg N /m3	0.38	1.54	2.39	1.19	0.74	0.55	0.44	0.64	0.37	0.61	1.79	1.26	0.98
DK0003R	ammonium	aerosol	µg N /m3	0.74	0.98	1.39	1.11	0.97	0.77	0.74	0.72	0.41	0.64	1.29	1.08	0.92
DK0008R	ammonium	aerosol	µg N /m3	0.42	0.66	1.17	0.97	0.87	0.68	0.67	0.72	0.44	0.61	1.16	0.53	0.74
DK0012R	ammonium	aerosol	µg N /m3	0.75	1.07	1.92	1.35	0.99	0.83	0.82	1.02	0.74	0.78	1.45	1.13	1.07
FI0009R	ammonium	aerosol	µg N /m3	0.26	0.35	0.31	0.28	0.22	0.32	0.32	0.25	0.26	0.24	0.32	0.38	0.29
FI0009R	ammonium	pm25	µg N /m3	0.26	0.26	0.25	0.26	0.16	0.28	0.25	0.19	0.14	0.18	0.33	0.33	0.24
FI0017R	ammonium	aerosol	µg N /m3	0.48	0.63	0.24	0.24	0.16	0.17	0.28	0.19	0.16	0.20	0.25	0.48	0.29
FI0017R	ammonium	pm25	µg N /m3	0.33	0.29	0.18	0.18	0.10	0.11	0.15	0.12	0.10	0.16	0.21	0.40	0.19
FI0037R	ammonium	aerosol	µg N /m3	0.38	0.36	0.10	0.18	0.10	0.10	0.16	0.13	0.10	0.13	0.20	0.34	0.19
LV0010R	ammonium	pm25	µg N /m3	0.21	-	-	-	-	-	0.29	0.22	0.19	0.22	0.56	1.34	(0.45)
PL0004R	ammonium	aerosol	µg N /m3	0.66	1.03	1.55	0.82	0.89	0.77	0.69	1.01	0.65	0.83	1.29	1.35	0.96
DK0003R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	1.16	1.53	3.90	2.79	2.20	1.52	1.61	1.75	1.03	1.04	1.64	1.37	1.83
DK0008R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.46	0.69	1.71	1.23	1.20	0.88	0.88	0.96	0.62	0.70	1.22	0.54	0.92
DK0012R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.89	1.33	4.46	2.60	1.93	1.46	1.51	2.10	1.49	1.26	1.67	1.19	1.86
FI0009R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.27	0.36	0.34	0.36	0.31	0.36	0.45	0.38	0.38	0.31	0.38	0.40	0.36
FI0017R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.48	0.64	0.27	0.30	0.41	0.27	0.49	0.35	0.31	0.27	0.28	0.52	0.38
FI0037R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.39	0.39	0.17	0.27	0.27	0.14	0.29	0.23	0.19	0.17	0.22	0.35	0.26
LT0015R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.50	0.96	0.99	0.90	0.56	0.58	0.60	0.65	0.56	0.78	1.10	1.66	0.78
PL0004R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.88	1.35	1.91	1.40	1.51	1.73	1.30	1.51	1.62	1.25	1.59	1.52	1.47
SE0005R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.19	0.13	0.08	0.11	0.13	0.16	0.16	0.13	0.07	0.10	0.20	0.17	0.14
SE0011R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.40	0.43	1.55	0.99	1.01	0.97	0.85	0.87	0.74	0.58	0.93	0.47	0.82
SE0012R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.23	0.30	0.44	0.36	0.29	0.33	0.46	0.37	0.23	0.25	0.42	0.34	0.34
SE0014R	sum_ammonia_and_ammonium	air+aerosol	µg N /m3	0.37	0.40	1.05	0.82	0.84	0.60	0.62	0.66	0.36	0.39	0.71	0.37	0.60

Table A.2 Monthly and annual mean concentrations of heavy metals in air.

Site	Component	Matrix	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
DE0009R	cadmium	pm10	ng Cd/m3	0.130	0.107	0.090	0.066	0.059	0.025	0.031	0.041	0.054	0.104	0.162	0.191	0.088
DK0008R	cadmium	aerosol	ng Cd/m3	0.070	0.080	0.095	0.255	0.073	0.026	0.018	0.019	0.018	0.048	0.086	0.096	0.073
DK0012R	cadmium	aerosol	ng Cd/m3	0.094	0.110	0.093	0.085	0.082	0.015	0.008	0.056	0.066	0.063	0.109	0.121	0.073
EE0009R	cadmium	aerosol	ng Cd/m3	0.387	0.124	0.023	0.087	0.068	0.090	0.094	0.023	0.023	0.055	0.093	0.194	0.093
FI0017R	cadmium	pm10	ng Cd/m3	0.268	0.211	0.051	0.068	0.050	0.031	0.053	0.048	0.062	0.079	0.126	0.284	0.111
FI0037R	cadmium	pm10	ng Cd/m3	0.171	0.119	0.047	0.059	0.033	0.021	0.044	0.051	0.058	0.055	0.076	0.135	0.072
LV0010R	cadmium	pm10	ng Cd/m3	0.025	-	-	-	-	-	0.006	0.042	0.091	0.294	0.177	0.202	(0.143)
SE0005R	cadmium	aerosol	ng Cd/m3	0.022	0.008	0.007	0.008	0.010	0.001	0.004	0.004	0.008	0.003	0.008	0.021	0.009
SE0011R	cadmium	aerosol	ng Cd/m3	0.018	0.009	0.011	0.046	0.046	0.013	0.003	0.012	0.010	0.007	0.006	0.004	0.016
SE0012R	cadmium	aerosol	ng Cd/m3	0.060	0.050	0.044	0.053	0.044	0.014	0.023	0.027	0.026	0.025	0.074	0.096	0.044
SE0014R	cadmium	aerosol	ng Cd/m3	0.063	0.141	0.086	0.057	0.032	0.004	0.023	0.027	0.041	0.131	0.183	0.023	0.067
DE0009R	lead	pm10	ng Pb/m3	5.09	4.06	3.29	2.41	2.46	1.33	1.33	1.75	2.04	3.36	6.47	6.77	3.35
DK0008R	lead	aerosol	ng Pb/m3	2.79	2.67	1.06	1.75	2.38	0.93	1.38	1.13	0.71	1.62	3.45	3.20	1.91
DK0012R	lead	aerosol	ng Pb/m3	3.78	2.70	2.05	1.71	1.58	0.94	0.89	0.83	1.46	2.14	3.86	4.08	2.12
EE0009R	lead	aerosol	ng Pb/m3	5.62	5.87	1.53	1.94	1.29	0.62	1.07	0.56	0.79	1.20	1.74	5.38	2.29
FI0017R	lead	pm10	ng Pb/m3	8.48	7.42	1.58	2.26	1.62	1.00	1.77	1.18	1.35	1.74	2.29	9.20	3.29
FI0037R	lead	pm10	ng Pb/m3	5.35	4.10	1.36	1.48	0.91	0.49	0.81	0.68	0.80	0.96	1.45	5.22	1.94
LV0010R	lead	pm10	ng Pb/m3	1.80	-	-	-	-	-	2.13	1.44	2.86	5.19	5.94	8.36	(4.39)
SE0005R	lead	aerosol	ng Pb/m3	0.81	0.31	0.23	0.20	0.16	0.09	0.16	0.10	0.24	0.10	0.16	0.76	0.28
SE0011R	lead	aerosol	ng Pb/m3	0.49	0.37	0.50	1.59	1.50	1.07	0.14	0.37	0.29	0.23	0.14	0.11	0.58
SE0012R	lead	aerosol	ng Pb/m3	1.72	1.45	1.34	1.37	0.99	0.47	0.75	0.63	0.82	0.74	1.99	2.86	1.23
SE0014R	lead	aerosol	ng Pb/m3	1.97	4.22	2.76	2.15	1.63	1.06	1.26	1.22	1.57	3.87	4.89	0.95	2.28
DE0009R	mercury (TGM)	air	ng Hg/m3	1.71	1.72	1.67	1.68	1.67	1.61	1.58	1.61	1.53	1.56	1.77	1.76	1.66
SE0005R	mercury (TGM)	air+aerosol	ng Hg/m3	1.58	1.43	1.10	1.40	1.00	1.33	1.24	1.15	1.15	0.96	1.18	1.48	1.25
SE0011R	mercury (TGM)	air+aerosol	ng Hg/m3	1.20	1.58	1.60	1.45	1.46	1.43	1.38	1.40	1.40	1.08	1.65	1.58	1.42
SE0014R	mercury (TGM)	air+aerosol	ng Hg/m3	1.61	1.62	1.51	1.58	1.64	1.51	1.48	1.43	1.39	1.39	1.65	1.59	1.53
SE0014R	mercury (aerosol)	aerosol	pg Hg/m3	5.5	7.5	8.4	8.0	7.3	7.0	4.1	6.7	2.5	6.3	10.1	8.8	6.9

Table A.3 Monthly and annual mean concentrations of ammonium and nitrate in precipitation.

Site	Component	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
DE0009R	ammonium	mgN/l	0.22	0.83	2.91	1.70	1.11	0.75	0.41	0.84	0.99	0.26	0.53	0.25	0.61
DK0005R	ammonium	mgN/l	0.15	1.40	1.81	1.67	0.86	0.50	0.33	0.68	0.53	0.36	0.43	0.38	0.49
DK0008R	ammonium	mgN/l	0.11	0.26	0.97	1.13	0.54	0.25	0.34	0.42	0.59	0.26	0.33	0.31	0.40
EE0009R	ammonium	mgN/l	0.05	0.06	0.22	0.28	0.33	0.08	0.10	0.06	0.08	0.09	0.12	0.10	0.13
EE0011R	ammonium	mgN/l	0.09	0.16	0.81	0.81	0.49	0.10	0.41	0.56	0.64	0.04	0.49	0.17	0.32
FI0004R	ammonium	mgN/l	0.06	0.10	0.10	0.37	0.18	0.07	0.05	0.06	0.28	0.07	0.20	0.08	0.11
FI0017R	ammonium	mgN/l	0.20	0.31	0.23	0.43	0.33	0.18	0.22	0.18	0.20	0.18	0.27	0.30	0.23
FI0053R	ammonium	mgN/l	0.25	0.37	0.40	0.46	0.23	0.12	0.20	0.06	0.19	0.21	0.54	0.32	0.25
LT0015R	ammonium	mgN/l	0.24	0.38	2.02	0.34	1.14	0.29	0.37	0.69	0.34	0.16	0.34	0.51	0.37
LV0010R	ammonium	mgN/l	0.22	0.54	0.90	0.80	0.83	0.25	0.36	0.64	0.26	0.10	0.56	0.29	0.38
PL0004R	ammonium	mgN/l	0.26	0.31	1.19	0.48	0.54	0.56	0.32	0.64	0.28	0.27	0.46	0.28	0.40
SE0011R	ammonium	mgN/l	-	0.55	2.06	1.25	0.55	0.31	0.23	0.38	0.40	0.29	0.36	0.38	0.49
SE0012R	ammonium	mgN/l	0.08	0.08	0.17	0.42	0.44	0.11	0.12	0.45	0.33	0.26	-	-	0.26
SE0014R	ammonium	mgN/l	0.12	0.28	1.44	1.10	0.75	0.34	0.28	0.46	0.27	0.26	0.33	0.15	0.39
SE0053R	ammonium	mgN/l	0.18	0.14	0.15	0.26	0.15	0.30	0.24	0.07	0.10	0.10	0.24	0.14	0.16
DE0009R	nitrate	mgN/l	0.25	0.64	1.28	0.81	0.55	0.40	0.27	0.33	0.53	0.33	0.43	0.34	0.40
DK0005R	nitrate	mgN/l	0.22	1.91	1.84	0.87	0.39	0.39	0.33	0.44	0.40	0.39	0.54	0.63	0.43
DK0008R	nitrate	mgN/l	0.19	0.39	0.65	0.73	0.58	0.25	0.42	0.40	0.51	0.46	0.60	0.58	0.44
EE0009R	nitrate	mgN/l	0.22	0.34	0.25	0.32	0.20	0.12	0.13	0.27	0.15	0.18	0.24	0.47	0.22
EE0011R	nitrate	mgN/l	0.32	0.21	0.72	0.71	0.52	0.04	0.22	0.38	0.20	0.20	0.63	0.42	0.32
FI0004R	nitrate	mgN/l	0.22	0.29	0.15	0.38	0.18	0.15	0.09	0.10	0.36	0.14	0.38	0.32	0.20
FI0017R	nitrate	mgN/l	0.38	0.60	0.36	0.43	0.34	0.21	0.23	0.17	0.24	0.21	0.35	0.49	0.29
FI0053R	nitrate	mgN/l	0.32	0.42	0.33	0.35	0.21	0.14	0.20	0.06	0.16	0.19	0.55	0.39	0.24
LT0015R	nitrate	mgN/l	0.61	0.77	2.28	0.32	0.73	0.33	0.34	0.46	0.48	0.28	0.47	0.93	0.43
LV0010R	nitrate	mgN/l	0.52	0.84	0.92	0.42	0.48	0.28	0.33	0.46	0.45	0.22	0.90	0.63	0.48
PL0004R	nitrate	mgN/l	0.34	0.44	0.73	0.42	0.35	0.40	0.28	0.40	0.37	0.28	0.50	0.54	0.38
SE0011R	nitrate	mgN/l	-	0.55	1.34	0.73	0.42	0.28	0.32	0.39	0.55	0.37	0.51	0.62	0.50
SE0012R	nitrate	mgN/l	0.26	0.33	0.21	0.37	0.39	0.15	0.26	0.38	0.37	0.39	-	-	0.32
SE0014R	nitrate	mgN/l	0.23	0.41	1.12	0.67	0.60	0.30	0.28	0.37	0.33	0.46	0.55	0.37	0.40
SE0053R	nitrate	mgN/l	0.31	0.27	0.25	0.24	0.15	0.18	0.18	0.16	0.13	0.17	0.41	0.30	0.21

Table A.4 Monthly and annual mean concentrations of heavy metals in precipitation.

Site	Component	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
DE0009R	cadmium	µgL	0.015	0.042	0.081	0.068	0.048	0.028	0.013	0.024	0.026	0.013	0.02	0.015	0.023
DK0005R	cadmium	µgL	0.01	0.015	0.016	0.028	0.044	0.134	0.133	0.049	0.052	0.052	0.035	0.06	0.064
DK0008R	cadmium	µgL	0.012	0.014	0.028	0.061	0.027	0.037	0.018	0.028	0.033	0.038	0.032	0.078	0.035
EE0009R	cadmium	µgL	0.01	0.048	0.011	0.039	0.01	0.01	0.01	0.01	0.01	0.01	0.039	0.01	0.015
EE0011R	cadmium	µgL	*0.02*	*0.01*	*0.03*	*0.029*	*0.07*	*0.687*	*0.02*	*0.01*	*0.01*	*0.01*	*0.021*	*0.03*	*0.093*
FI0017R	cadmium	µgL	0.051	0.045	0.028	0.065	0.05	0.035	0.019	0.033	0.099	0.024	0.041	0.096	0.05
FI0053R	cadmium	µgL	0.03	0.048	0.037	0.067	0.046	0.04	0.109	0.022	0.015	0.004	0.048	0.024	0.041
LV0010R	cadmium	µgL	*0.031*	*0.035*	*0.135*	*0.037*	*0.03*	*0.03*	*0.033*	*0.03*	*0.01*	*0.03*	*0.077*	*0.042*	*0.04*
PL0004R	cadmium	µgL	0.052	0.064	0.035	0.074	0.044	0.043	0.024	0.024	0.019	0.023	0.029	0.047	0.034
SE0005R	cadmium	µgL	0.039	0.005	0.005	0.01	0.025	0.09	0.009	0.005	0.009	0.01	0.01	0.062	0.023
SE0011R	cadmium	µgL	0.01	0.021	0.066	0.139	0.162	0.561	0.03	0.038	0.025	0.158	0.03	0.03	0.126
SE0014R	cadmium	µgL	0.005	0.011	0.04	0.089	0.04	0.04	0.011	0.013	0.05	0.02	0.03	0.02	0.03
DE0009R	lead	µgL	0.54	1.43	2.40	1.99	1.57	0.86	0.50	0.83	0.99	0.41	0.79	0.54	0.79
DK0005R	lead	µgL	2.79	5.35	0.60	1.75	1.25	8.50	8.43	1.49	8.07	7.98	2.32	-	5.17
DK0008R	lead	µgL	0.53	0.45	0.52	1.72	1.32	1.86	0.94	1.59	1.17	1.39	1.18	2.09	1.38
EE0009R	lead	µgL	0.15	0.60	0.24	0.15	0.25	0.10	0.12	0.06	0.30	0.12	0.43	0.34	0.21
EE0011R	lead	µgL	0.16	0.16	0.24	0.10	0.20	1.28	0.10	0.10	0.10	0.17	3.83	0.62	0.53
FI0017R	lead	µgL	1.64	1.73	1.16	1.39	1.42	0.38	0.44	0.27	0.53	0.50	1.01	2.76	0.89
FI0053R	lead	µgL	0.83	1.68	1.31	0.72	0.76	0.30	0.42	0.14	0.18	0.06	0.94	0.93	0.55
LV0010R	lead	µgL	0.40	0.30	0.33	0.50	0.30	0.30	0.41	0.30	0.32	0.21	1.01	0.74	0.44
PL0004R	lead	µgL	0.66	0.96	0.45	0.76	0.48	0.50	0.37	0.44	0.43	0.39	0.76	0.83	0.54
SE0005R	lead	µgL	1.31	0.14	0.18	0.19	0.19	0.14	0.19	0.10	1.56	0.42	0.15	1.65	0.45
SE0011R	lead	µgL	0.27	0.57	1.09	0.51	1.02	0.75	0.34	1.01	0.69	0.67	0.79	0.63	0.65
SE0014R	lead	µgL	0.15	0.42	0.92	1.13	0.48	0.30	0.36	0.63	0.28	0.29	0.63	0.42	0.46
DE0009R	mercury	ng/L	5.8	10.1	15.5	17.1	17.7	13.3	7.9	15.2	11.1	4.7	4.9	3.7	8.7
FI0017R	mercury	ng/L	5	23	11	9	13	5	8	3	1	1	1	1	3.8
LV0010R	mercury	ng/L	*30*	*30*	*30*	*30*	*30*	*30*	*30*	*30*	*30*	*30*	*30*	*31*	*30*
SE0005R	mercury	ng/L	4.4	7.0	13.8	12.1	5.6	6.6	7.6	4.2	3.3	2.5	11.6	13.6	5.9
SE0011R	mercury	ng/L	4.1	7.8	19.5	15.3	22.4	8.3	13.3	15.9	8.7	17.5	8.7	11.0	11.0
SE0014R	mercury	ng/L	5.1	7.7	10.6	31.6	19.1	16.1	7.0	9.9	8.1	6.0	6.4	6.4	11.4
DE0009R	precipitation_amount	mm'	53	23	5	28	16	54	104	24	36	60	39	60	502
DE0009R	precipitation_amount (Hg)	mm'	56	23	6	28	20	49	105	23	40	59	42	60	510
DK0005R	precipitation_amount	mm'	68	13	7	20	38	4	123	35	3	78	38	54	482
DK0008R	precipitation_amount	mm'	65	20	21	41	26	126	38	104	85	72	43	58	698
EE0009R	precipitation_amount	mm'	65	36	47	62	83	75	103	70	109	134	56	53	894
EE0011R	precipitation_amount	mm'	75	96	41	39	38	98	60	68	95	120	48	113	891
FI0017R	precipitation_amount	mm'	66	18	25	26	32	68	63	63	126	134	73	48	743
FI0017R	precipitation_amount (Hg)	mm'	31	4	22	19	28	62	54	66	131	115	47	27	604
FI0053R	precipitation_amount	mm'	24	10	11	46	64	42	35	52	65	21	31	25	425
LV0010R	precipitation_amount	mm'	86	50	32	30	51	42	97	47	82	141	103	67	829
PL0004R	precipitation_amount	mm'	79	42	16	37	24	58	188	96	120	77	81	50	868
SE0005R	precipitation_amount	mm'	20	17	11	17	66	61	93	89	61	62	33	26	555
SE0005R	precipitation_amount (Hg)	mm'	14	11	5	9	48	73	107	89	67	43	9	11	484
SE0011R	precipitation_amount	mm'	65	31	16	38	20	76	38	34	87	91	51	52	599
SE0011R	precipitation_amount (Hg)	mm'	64	31	9	19	18	99	51	49	104	64	50	46	604
SE0014R	precipitation_amount	mm'	48	25	12	55	39	91	88	95	99	63	48	79	743
SE0014R	precipitation_amount (Hg)	mm'	32	14	7	42	30	87	105	198	37	32	19	19	622

Data in italic indicates data with more than 75% of data below detection limit

Table A.5 Monthly and annual deposition of ammonium and nitrate in precipitation.

Site	Component	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year	Total N
DE0009R	ammonium	mg N m/2	14	21	20	51	19	44	45	23	40	17	23	16	337	
DE0009R	nitrate	mg N m/2	16	16	9	24	9	23	30	9	21	22	19	22	222	559
DE0009R	precipitation_amount	mm	65	26	7	30	17	59	111	27	40	66	43	65	554	
DK0005R	ammonium	mg N m/2	9	17	8	42	32	35	21	26	23	15	18	21	246	
DK0005R	nitrate	mg N m/2	14	24	8	22	15	28	21	17	17	16	23	34	217	462
DK0005R	precipitation_amount	mm	63	12	4	25	38	71	65	39	43	41	42	55	500	
DK0008R	ammonium	mg N m/2	5	5	19	49	13	28	11	44	18	20	14	21	248	
DK0008R	nitrate	mg N m/2	9	8	13	32	13	29	13	42	16	34	26	39	275	523
DK0008R	precipitation_amount	mm	50	21	20	44	23	114	32	104	31	75	42	68	625	
EE0009R	ammonium	mg N m/2	3	2	10	17	28	6	10	5	9	13	10	6	119	
EE0009R	nitrate	mg N m/2	14	12	12	20	17	9	13	26	16	25	19	26	209	327
EE0009R	precipitation_amount	mm	65	35	47	61	83	74	104	98	108	137	81	55	948	
EE0011R	ammonium	mg N m/2	7	15	33	32	17	9	30	33	65	5	20	21	287	
EE0011R	nitrate	mg N m/2	24	20	29	28	19	4	16	22	20	25	25	52	284	570
EE0011R	precipitation_amount	mm	75	95	41	40	36	87	72	59	102	122	40	122	891	
FI0004R	ammonium	mg N m/2	3	4	3	14	11	3	8	5	14	8	8	4	85	
FI0004R	nitrate	mg N m/2	11	12	5	14	10	7	13	9	18	17	15	17	150	235
FI0004R	precipitation_amount	mm	50	41	32	38	59	46	153	87	52	118	39	54	767	
FI0017R	ammonium	mg N m/2	14	8	7	14	12	11	14	15	22	27	21	16	179	
FI0017R	nitrate	mg N m/2	26	15	11	14	12	13	15	14	27	31	27	26	230	409
FI0017R	precipitation_amount	mm	69	24	32	32	37	63	65	81	109	150	76	54	790	
FI0053R	ammonium	mg N m/2	6	4	6	21	15	4	7	3	13	17	21	7	125	
FI0053R	nitrate	mg N m/2	8	5	5	16	14	5	7	3	11	15	22	9	119	244
FI0053R	precipitation_amount	mm	25	12	14	46	65	37	35	51	67	81	40	22	494	
LT0015R	ammonium	mg N m/2	8	7	3	10	31	13	32	33	24	23	27	9	221	
LT0015R	nitrate	mg N m/2	20	15	4	10	20	15	29	22	34	40	37	16	262	482
LT0015R	precipitation_amount	mm	33	19	2	30	27	45	87	47	71	145	80	18	604	
LV0010R	ammonium	mg N m/2	19	27	29	24	42	10	35	30	21	15	58	19	316	
LV0010R	nitrate	mg N m/2	45	42	30	13	25	12	32	22	36	31	92	42	396	712
LV0010R	precipitation_amount	mm	86	50	32	30	51	42	97	47	82	141	103	67	829	
PL0004R	ammonium	mg N m/2	21	13	19	16	12	31	60	61	33	21	37	14	338	
PL0004R	nitrate	mg N m/2	27	18	11	14	8	22	52	38	44	22	41	27	324	661
PL0004R	precipitation_amount	mm	79	42	16	34	22	55	186	96	120	77	81	50	855	
SE0011R	ammonium	mg N m/2	0	97	48	54	21	27	12	19	38	34	20	23	394	
SE0011R	nitrate	mg N m/2	0	99	31	32	16	25	17	20	53	44	29	37	401	795
SE0011R	precipitation_amount	mm	0	178	23	43	38	89	53	51	96	117	57	60	805	
SE0012R	ammonium	mg N m/2	4	3	2	23	14	5	5	25	23	12	0	0	116	
SE0012R	nitrate	mg N m/2	14	13	2	20	12	7	10	21	26	18	0	0	144	260
SE0012R	precipitation_amount	mm	54	39	11	54	32	46	39	55	70	46	0	0	448	
SE0014R	ammonium	mg N m/2	8	8	17	66	40	36	31	43	31	21	19	7	326	
SE0014R	nitrate	mg N m/2	15	12	13	40	32	32	31	34	38	38	32	19	335	661
SE0014R	precipitation_amount	mm	62	30	12	60	53	106	110	93	115	83	59	51	833	
SE0053R	ammonium	mg N m/2	12	4	3	21	9	13	9	4	7	14	10	6	112	
SE0053R	nitrate	mg N m/2	20	8	4	19	9	8	7	10	9	23	18	12	147	259
SE0053R	precipitation_amount	mm	65	31	17	81	61	43	37	60	74	135	43	40	686	

Table A.6 Monthly and annual deposition of heavy metals in precipitation.

Site	Component	Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
DE0009R	cadmium	µg Cd /m2	0.8	1.0	0.4	1.9	0.7	1.5	1.4	0.6	0.9	0.8	0.8	0.9	11.7
DK0005R	cadmium	µg Cd /m2	0.7	0.2	0.1	0.6	1.7	0.5	16.4	1.7	0.1	4.0	1.3	3.3	30.6
DK0008R	cadmium	µg Cd /m2	0.8	0.3	0.6	2.5	0.7	4.6	0.7	2.9	2.8	2.7	1.4	4.5	24.5
EE0009R	cadmium	µg Cd /m2	*0.7*	*1.8*	*0.5*	*2.4*	*0.8*	*0.7*	*1.0*	*0.7*	*1.1*	*1.4*	*2.2*	*0.5*	*13.9*
EE0011R	cadmium	µg Cd /m2	1.5	1.0	1.2	1.2	2.7	67.4	1.2	0.7	1.0	1.2	1.0	3.4	83.3
FI0017R	cadmium	µg Cd /m2	3.4	0.8	0.7	1.7	1.6	2.4	1.2	2.1	12.4	3.2	3.0	4.6	37.1
FI0053R	cadmium	µg Cd /m2	0.7	0.5	0.4	3.1	2.9	1.7	3.8	1.1	1.0	0.1	1.5	0.6	17.4
LV0010R	cadmium	µg Cd /m2	2.7	1.8	4.3	1.1	1.5	1.3	3.2	1.4	0.8	4.3	8.0	2.8	33.3
PL0004R	cadmium	µg Cd /m2	4.1	2.7	0.5	2.7	1.0	2.5	4.5	2.3	2.3	1.8	2.4	2.4	29.2
SE0005R	cadmium	µg Cd /m2	0.8	0.1	0.1	0.2	1.7	5.5	0.8	0.4	0.6	0.6	0.3	1.6	12.6
SE0011R	cadmium	µg Cd /m2	0.7	0.6	1.1	5.3	3.2	42.7	1.2	1.3	2.2	14.4	1.5	1.6	75.6
SE0014R	cadmium	µg Cd /m2	0.2	0.3	0.5	4.9	1.6	3.7	1.0	1.2	5.0	1.3	1.4	1.6	22.6
DE0009R	lead	µg Pb /m2	29	33	13	55	24	47	52	20	36	25	31	32	396
DK0005R	lead	µg Pb /m2	191	70	4	35	47	35	1037	52	21	624	88	-	2489
DK0008R	lead	µg Pb /m2	35	9	11	71	34	234	35	166	99	100	51	121	965
EE0009R	lead	µg Pb /m2	10	22	11	9	20	8	12	4	33	17	24	18	188
EE0011R	lead	µg Pb /m2	12	15	10	4	8	125	6	7	10	20	183	70	470
FI0017R	lead	µg Pb /m2	109	31	29	36	45	26	28	17	67	67	74	134	661
FI0053R	lead	µg Pb /m2	20	16	14	33	48	13	15	7	12	1	29	24	232
LV0010R	lead	µg Pb /m2	35	15	11	15	15	13	39	14	26	29	104	50	368
PL0004R	lead	µg Pb /m2	52	40	7	28	11	29	70	42	52	30	62	42	465
SE0005R	lead	µg Pb /m2	26	2	2	3	13	9	18	9	96	26	5	42	250
SE0011R	lead	µg Pb /m2	17	17	17	19	20	57	13	35	60	61	40	33	390
SE0014R	lead	µg Pb /m2	7	10	11	62	19	27	32	60	28	18	30	33	338
DE0009R	mercury	ng Hg /m2	325	236	92	475	356	650	827	353	443	278	204	222	4460
FI0017R	mercury	ng Hg /m2	153	94	242	167	359	312	431	197	131	115	47	27	2274
LV0010R	mercury	ng Hg /m2	*2591*	*1508*	*960*	*913*	*1538*	*1251*	*2916*	*1416*	*2447*	*4240*	*3087*	*2055*	*24922*
SE0005R	mercury	ng Hg /m2	59	75	73	104	266	484	819	372	222	106	107	150	2837
SE0011R	mercury	ng Hg /m2	258	239	178	295	414	816	677	781	905	1123	432	507	6627
SE0014R	mercury	ng Hg /m2	165	107	72	1321	572	1403	734	1959	303	192	121	121	7069

Data in italic indicates data with more than 75% of data below detection limit

Appendix B: Monitoring methods

The monitoring regime for nitrogen compounds and metals are summarised in tables B.1 to B.3:

Table B.1. General information about sampling and analysis of nitrogen compounds in precipitation in 2012.

Country		Sampling period	Sampler		Analytical methods
			Wet only	Bulk	
Denmark	Nitrate ammonium	Biweekly	x		IC Spect. (CFA)
Estonia	Nitrate Ammonium	Weekly		X	IC Spect (indophenol)
Finland	Nitrate Ammonium	Weekly		X	IC IC
Germany	Nitrate Ammonium	Weekly	X		IC IC
Latvia	Nitrate Ammonium	Weekly	X		IC Spect (indophenol)
Lithuania	Nitrate Ammonium	Daily	X		IC Spect (indophenol)
Poland	Nitrate Ammonium	Daily		x	IC Spect (chloramin T)
Sweden	Nitrate Ammonium	Daily: SE05, 14 monthly: SE11, 12	X		IC Spect (FIA)

IC: Ion chromatograpy
Spect: Spectrofotometric detection
CFA: continuously flow analysis
FIA: Flow injection analysis

Table B.2. General information about sampling and analysis of nitrogen compounds in air in 2012.

Country		**Sampl period**	**Sampler**	**Analytical methods**
Denmark	NO_2	Hourly	Chemiluminisence	
	Sum of nitric acid and nitrate	Daily	Millipore RAWP, 1.2 µm + KOH-impregnated Whatman 41, 58 m^3/day	IC
	Sum of ammonia and ammonium	Daily	Millipore RAWP, 1.2 µm + Oxalic acid impregnated Whatman 41, 58 m^3/day	Spect (CFA)
Estonia	NO_2	Hourly	Chemiluminisence	
Finland 1)daily at FI09,FI17 Weekly at FI37	*(*NO_2	Hourly	Chemiluminisence	
	Sum of nitric acid and nitrate	Daily1)	Teflon filter + NaOH impregnated Whatman 40 filter, 24 m^3/day	IC
	Sum of ammonia and ammonium	Daily1)	Teflon filter +Oxalic acid impregnated Whatman 40 filter, 24 m^3/day	IC
Germany	NO_2	Daily	NaI imp. Glass filters, 0.7m^3/day	FIA
	NH3	Weekly	Low cost denuder	FIA
	NH4	Daily	Filterpack, Teflon filter (jan-july)	IC
			Low vol sampl., $PM_{2.5}$ quartz filter (july -dec)	
	NO3	Daily	Filterpack, Teflon filter (jan-july)	IC
			Low vol sampl., $PM_{2.5}$ quartz filter (july -dec)	
Lithuania	NO_2,	Daily	KI imp glass filters 0.4-0.7 m^3/day	Spect. Griess
	Sum of nitric acid and nitrate	Daily	Aerosol filter (Whatman 40) + KOH impregnated filter, 20 m^3/day	IC
	Sum of ammonia and ammonium	Daily	Aerosol filter (Whatman 40) + oxalic acid impregnated filter, 20 m^3/day	Spect (indophenol)
Poland	NO_2	Daily	Abs.sol. TGS 0.7 m^3/day	Spect. Griess
	Sum of nitric acid and nitrate	Daily	Aerosol filter (Whatman 40) + NaF impregnated Whatman 40 filter, 3.5-4 m^3/day	Spect. Griess
	Sum of ammonia and ammonium	Daily	Aerosol filter (Whatman 40) + Oxalic acid impregnated Whatman 40 filter, 3.5-4 m^3/day	Spect. Chloramin T)
Sweden	NO_2	Daily	NaI imp. glass sinters 0.7 m^3/day	Spect, FIA
	Sum of nitric acid and nitrate		Teflon filter, Mitex membrane + KOH-impregnated Whatman 40 filter, 20 m^3/day	IC
	Sum of ammonia and ammonium		Teflon filter, Mitex membrane + Oxalic acid impregnated Whatman 40 filter, 20 m^3/day	FIA

IC: Ion chromatograpy
Spect Spectrofotometric detection
FIA: Flow injection analysis

***Table B.3**. General information about sampling and analysis of heavy metals in 2012.*

Country	**Precipitation**		**Air and aerosols**		**Laboratory method**
	Field method	**Frequency**	**Field method**	**Frequency**	
Denmark	Bulk	Monthly	Low volume sampler, Millipore RAWP 1.2 mm, 58 m^3/day TGM: monitor (Tekran)	daily continuously	Precip: GF-AAS Aerosols: ICP-MS
Estonia	Bulk	EE08 daily EE11 weekly		weekly	GF-AAS
Finland	Bulk	Monthly	PM$_{10}$, Teflon, Millipore Fluoropore 3 µm, 20 l/min	FI17: 2+2+3 days, FI36+FI37: weekly	ICP-MS
Germany Hg	wet only wet only	Weekly Weekly	Low volume sampler TGM:Tekran Monitor	weekly hourly	ICP-MS
Latvia	Wet only	Weekly	PM$_{10}$, low volume sampler. 2.3 m^3/h	Weekly	ICP-MS
Poland	Wet-only	biweekly			GF-AAS
Sweden Hg	Bulk Bulk (Hg)	Monthly Monthly	Low volume sampler, teflon filter Hg: gold traps (TGM) Hg: mini traps (TPM)	monthly 2 X 24 h a week 1 X 24 h a week	ICP-MS CV-AFS CV-AFS

GF-AAS: Graphic Furnace Atomic Absorption Spectroscopy
ICP-MS: Inductively Coupled Plasma - Mass Spectrometry
CV-AFS: Cold Vapour Atomic Fluorescence Spectroscopy

Appendix C: Indicator Fact Sheets on nitrogen emissions

Here we give the links to Indicator Fact Sheets available on HELCOM web pages:

1. Nitrogen emissions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/eutrophication/nitrogen-emissions-to-the-air-in-the-baltic-sea-area/

2. Nitrogen depositions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/eutrophication/nitrogen-atmospheric-deposition-to-the-baltic-sea/

3. Heavy metals emissions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/hazardous-substances/atmospheric-emissions-of-heavy-metals-in-the-baltic-sea-region/

4. Heavy metals depositions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/hazardous-substances/atmospheric-deposition-of-heavy-metals-on-the-baltic-sea/

5. PCDD/Fs emissions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/hazardous-substances/atmospheric-emissions-of-pcdd-fs-in-the-baltic-sea-region/

6. PCDD/Fs depositions:
http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/hazardous-substances/atmospheric-deposition-of-pcdd-fs-on-the-baltic-sea/

Appendix D: Calculation of normalised deposition to the Baltic Sea Basin

D.1 Introduction

In the frame of co-operation between HELCOM and EMEP, estimation of atmospheric nitrogen deposition has been carried out for each year of the period 1995-2011. Annual depositions, monthly depositions, as well as annual source-allocation budgets for nitrogen deposition have been calculated using the EMEP MSC-W model. The main purpose of this appendix is a description and explanation how nitrogen deposition, source-allocation budgets and especially normalised nitrogen deposition to the Baltic Sea are calculated. We focus on nitrogen here, but normalised depositions of heavy metals and persistent organic pollutants are calculated in very similar way.

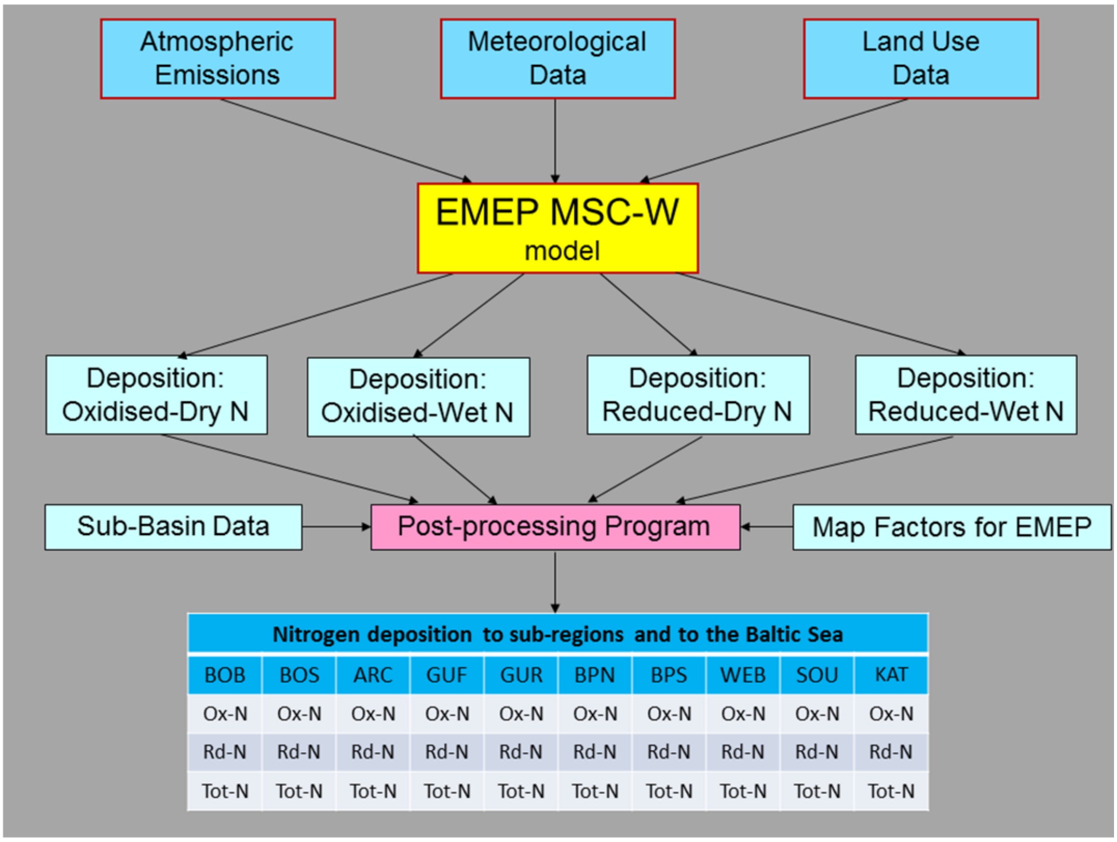
D.2 Annual deposition

The routine runs of the EMEP MSC-W model are performed every ear with updated input data for the purpose of LRTAP Convention and in the frame of co-operation between HELCOM and EMEP. The input data necessary for routine runs of the EMP model are: emissions, meteorological data and land use data. Emissions and meteorological fields must be updated each year for routine runs. The land used data are updated each time when better information about the land use is available.

Both anthropogenic and biogenic emissions are required for the EMEP MSC-W model runs. Concerning anthropogenic emissions, as much as possible, data officially reported by EMEP Contracting Parties are used for the purpose of modelling. Annual national totals for each country should be reported every year to EMEP and they are distributed to each grid cell of the model. In addition, approximately every five years, the distribution of national emissions in the EMEP gird is updated by the Contracting Parties. The main conditions for using official data are availability and quality good enough. When the officially reported data is not available or the data quality is not good enough, the expert estimates are used instead for the model runs. The procedures used for collecting anthropogenic emissions, filling-in gaps, and for spatial distribution can be found in Vestreng (2003). Emissions of eight species are necessary for routine runs of the EMEP model: SO_2, NO_x NH_3, CO, NMVOC, primary $PM_{2.5}$ and PM_{10}. These emission fields must be available and updated in the model grid for routine annual runs.

Meteorological data include both, three dimensional fields and two dimensional fields on the surface layer. Meteorological fields available in 3-D are the following: velocity, pressure, temperature and humidity. Precipitation is one example of 2-D meteorological data. The land use data include matrices with different types of land cover which are variable in space in time, especially for different seasons of the year.

Computational diagram for calculating atmospheric oxidised, reduced and total nitrogen deposition to sub-basins of the Baltic Sea and to the entire Baltic Sea Basin using the EMEP MSC-W model is illustrated in Fig. D1.



Nitrogen deposition to sub-regions and to the Baltic Sea									
BOB	BOS	ARC	GUF	GUR	BPN	BPS	WEB	SOU	KAT
Ox-N	Ox-N	Ox-N	Ox-N	Ox-N	Ox-N	Ox-N	Ox-N	Ox-N	Ox-N
Rd-N	Rd-N	Rd-N	Rd-N	Rd-N	Rd-N	Rd-N	Rd-N	Rd-N	Rd-N
Tot-N	Tot-N	Tot-N	Tot-N	Tot-N	Tot-N	Tot-N	Tot-N	Tot-N	Tot-N

Fig. D1. Computational diagram for calculating oxidised, reduced and total nitrogen deposition to sub-basins of the Baltic Sea and to the entire Baltic Sea Basin using the EMEP MSC-W model.

Using input data with updated emissions, land use and meteorology for the current year, the EMEP MSC-W model is run to calculate annual, monthly and daily values of oxidized-dry, oxidized-wet, reduced-dry and reduced-wet nitrogen deposition (in mg N m^{-2}) in each grid square of the EMEP grid systems. Calculated annual and monthly depositions are used for the purpose of HELCOM.

Four output files from the EMEP model run, with annual nitrogen depositions are then used as input for the post-processing program. The file defining the sub-basins of the Baltic Sea in the EMEP grid and the file with map factors for the EMEP grid system are also used by the post-processing program. The output from the post-processor program includes annual total depositions (in tonnes of N) of oxidised, reduced and total (oxidised + reduced) nitrogen to each of ten sub-basins of the Baltic Sea, as requested by HELCOM. Annual depositions of oxidised, reduced and total nitrogen the entire Baltic Sea basin are calculated as the sum of depositions to all sub-basins. The deposition files shown in Fig. 1 are also used for creating annual deposition maps for HELCOM, shown in Chapter 3.

D. 3 Contributions from individual sources

The procedure for calculating contribution of individual emission sources to nitrogen deposition is a bit complicated in that sense that nitrogen deposition depends not only on nitrogen emissions, but other emissions as well (EMEP Status Report, 2006). As emission sources we consider both country sources (emissions from individual EMEP contracting Parties) and other sources (international ship emissions, volcanoes etc.). There are altogether 55 country sources and other sources which are taken into account in the EMEP model calculations every year.

To calculate the contributions from individual sources to nitrogen deposition into the Baltic Sea and its sub-basins the model is run with complete emissions first. In the next step, four model runs are performed for each contributing source. In the first run emissions of nitrogen oxides from the source under consideration are reduced by 15%. In the second run, emissions of ammonia are reduced by 15%. In the third model run, VOC emissions are reduced by 15% and finally in the fourth run emissions of VOC are reduced by 15%. Atmospheric deposition of oxidised-dry, oxidized-wet, reduced-dry and reduced-wet nitrogen is calculated for each of the model runs. The contribution of country (or other source) ***n*** to oxidised nitrogen deposition to each grid of the model domain is calculated as:

$$\begin{aligned} d^{n}_{oxdry}(i,j) = {} & \Big[\Big(d^{tot}_{oxdry}(i,j) - d^{SOx-15\%}_{oxdry}(i,j)\Big) + \Big(d^{tot}_{oxdry}(i,j) - d^{NOx-15\%}_{oxdry}(i,j)\Big) + \\ & \Big(d^{tot}_{oxdry}(i,j) - d^{NO3-15\%}_{oxdry}(i,j)\Big) + \Big(d^{tot}_{oxdry}(i,j) - d^{VOC-15\%}_{oxdry}(i,j)\Big)\Big] \times \frac{100}{15} \end{aligned} \qquad \text{(D1)}$$

where:

$d^{n}_{oxdry}(i,j)$ - is the contribution of source ***n*** to oxidised nitrogen deposition in the model grid square ***(i,j)***,

$d^{tot}_{oxdry}(i,j)$ - is the oxidised nitrogen deposition in model grid ***(i,j)*** calculated with all emission sources,

$d^{SOx-15\%}_{oxdry}(i,j)$ - is the deposition calculated with 15% reduction of SO_x emissions in source ***n***,

$d^{NOx-15\%}_{oxdry}(i,j)$ - is the deposition calculated with 15% reduction of NO_x emissions in source ***n***,

$d^{NH3-15\%}_{oxdry}(i,j)$ - is the deposition calculated with 15% reduction of ammonia emissions in source ***n***,

$d^{VOC-15\%}_{oxdry}(i,j)$ - is the deposition calculated with 15% reduction of VOC emissions in source ***n***.

The same procedure is used to calculate contributions of source ***n*** to oxidized-wet $d^{n}_{oxwet}(i,j)$, reduced-dry $d^{n}_{rddry}(i,j)$ and reduced-wet $d^{n}_{rdwet}(i,j)$ nitrogen deposition to each grid of the EMEP model. The contribution of the source ***n*** to nitrogen deposition into the Baltic Sea is calculated as a sum of contributions from each model grid square belonging to the Baltic Sea basin. For example, contribution of source n to oxidised nitrogen deposition into the Baltic Sea is calculated in the following way:

$$D^{n}_{oxdry} = \sum_{(i,j)\in Baltic}\left(d^{n}_{oxdry}(i,j)\times S(i,j)\right) \qquad \text{(D2)}$$

Where D^{n}_{oxdry} is the contribution of source ***n*** to deposition of oxidised dry nitrogen into the Baltic Sea basin and $S(i,j)$ is the surface of the grid ***(i,j)*** belonging to the Baltic Sea basin. Similar calculations are made for contribution of source n to oxidized-wet - D^{n}_{oxwet}, reduced-dry - D^{n}_{rddry} and reduced-wet - D^{n}_{rdwet} nitrogen deposition. The most important for HELCOM are depositions of oxidised nitrogen - D^{n}_{ox}, reduced nitrogen – D^{n}_{rd} and total nitrogen - D^{n}_{tot} to the Baltic Sea basin. These depositions are defined as:

$$\begin{aligned} D^{n}_{ox} &= D^{n}_{oxdry} + D^{n}_{oxwet} \\ D^{n}_{rd} &= D^{n}_{rddry} + D^{n}_{rdwet} \\ D^{n}_{tot} &= D^{n}_{ox} + D^{n}_{rd} \end{aligned} \qquad \text{(D3)}$$

The calculations described by Equations (D1)-(D3) are performed for all emissions sources in the EMEP domain in order to calculate all contributions. The sum of these contributions is equal to total deposition of nitrogen to the Baltic Sea basin.

D.4 Source-receptor matrices

Assuming linearity, or at least local linearity, the source-receptor matrices describe the relation between emissions of nitrogen in the EMEP sources and nitrogen deposition to the Baltic Sea basin. With the simplified linearity assumption, the source-receptor matrices are defined in the following as:

$$A_{ij}(iy) = \frac{D_i(iy)}{E_j(iy)} \tag{D4}$$

where:

$E_j(iy)$ - is the annual emission from the source ***j*** in year ***iy***,

$D_i(iy)$ - is the annual deposition in the receptor ***i*** in year ***iy***,

$A_{ij}(iy)$ - is the source-receptor matrix for the year ***iy***.

The source-receptor matrix gives the amount of annual emission in the source ***j*** deposited in the receptor ***i*** for a given year. The dimension of the source-receptor matrix for a given year is $(ne \times ns)$, where ***ne*** is the number of receptors and ***ns*** is the number of emission sources. In our case, we are only interested in one receptor, namely the Baltic Sea basin and the index ***i*** can be omitted. In this case, the source-receptor matrices for oxidized and reduced nitrogen become vectors and are defined as:

$$\begin{aligned} A_i^{ox}(iy) &= \frac{D_i^{ox}(iy)}{E_i^{ox}(iy)} \\ A_i^{rd}(iy) &= \frac{D_i^{rd}(iy)}{E_i^{rd}(iy)} \end{aligned} \tag{D5}$$

where:

$E_i^{ox}(iy)$ - is the annual emission of nitrogen oxides from the source ***i*** in the year ***iy***,

$E_i^{rd}(iy)$ - is the annual emission of ammonia from the source ***i*** in the year ***iy***,

$D_i^{ox}(iy)$ - is the annual deposition of oxidised nitrogen from the source ***i*** in the year ***iy***,

$D_i^{rd}(iy)$ - is the annual deposition of reduced nitrogen from the source ***i*** in the year ***iy***,

$A_i^{ox}(iy)$ - is the source-receptor matrix (vector) for oxidized nitrogen the year ***iy***,

$A_i^{rd}(iy)$ - is the source-receptor matrix (vector) for reduced nitrogen for the year ***iy***.

The total nitrogen deposition to the Baltic Sea basin in the year ***iy*** can be calculated as:

$$D^{tot}(iy) = D^{ox}(iy) + D^{rd}(iy) = \sum_{i=1}^{ns1} A_i^{ox}(iy) \times E_i^{ox}(iy) + \sum_{i=1}^{ns2} A_i^{rd}(iy) \times E_i^{rd}(iy) \tag{D6}$$

where $D^{ox}(iy)$ and $D^{rd}(iy)$ is the annual total deposition of oxidized and reduced nitrogen, respectively, to the Baltic Sea in the year ***iy***. The numbers of emission sources contributing to oxidized nitrogen deposition (***ns1***) and reduced nitrogen (***ns2***) are different in general, because some sources (e.g. ship traffic on the Baltic Sea) emit only oxidized nitrogen.

D.5 Normalised depositions

The calculated nitrogen depositions to the Baltic Sea vary from one year to another, not only because of different emissions, but because of different meteorological conditions for each year. Some model runs with constant emissions and variable meteorology performed for 12 years period (Bartnicki et al. 2010) show that calculated annual nitrogen depositions can differ up to 60% for different years. Therefore, the best way to reduce the influence of meteorology on computed annual nitrogen depositions would be to run the EMEP model with the same emissions from one particular year, but with all available different meteorological years and then average the results over the years or calculate the median depositions. The annual depositions calculated in this way can be called as "normalised" in the sense of meteorological variability. Unfortunately, the direct calculations of "normalized" nitrogen depositions are difficult, time consuming and expensive. Therefore, a simplified approach was applied using the source-receptor matrices for oxidized and reduced nitrogen, described in the previous section. The source receptor matrices differ from one year to another depending mainly on meteorological conditions. Therefore, they are often used for prediction of future depositions with a given scenario when meteorological conditions are not known. They have been also used in our approach for calculating normalised depositions to the Baltic Sea basin. In this approach, we have used the source-receptor matrices and depositions as defined in Eq. (D5-D6) and calculated for each of 17-year period 1995-2011 with available EMEP model runs. The "normalised" depositions to the Baltic Sea were calculated for oxidized, reduced and total nitrogen and for each year of the period 1995-2011. In the first step of this process, the annual depositions were calculated for each combination of meteorological and emission year:

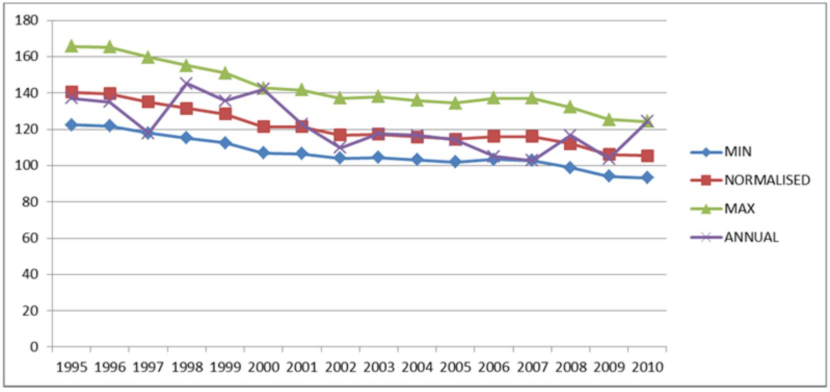
$$\begin{aligned} D^{ox}(ie,im) &= \sum_{i=1}^{ns1} A^{ox}(im) \times E^{ox}(ie) + R^{ox}(ie,im) \\ D^{rd}(ie,im) &= \sum_{i=1}^{ns2} A^{rd}(im) \times E^{rd}(ie) + R^{rd}(ie,im) \end{aligned} \qquad \text{(D7)}$$

Terms $R^{ox}(ie,im)$ and $R^{rd}(ie,im)$ are introduce mainly because of the contribution of BIC (Initial and Boundary Conditions) in the model calculations, additional source for which emissions cannot be specified. For the Baltic Sea basin this additional source is only contributing to oxidized nitrogen deposition, so $R^{rd}(ie,im) = 0$. The normalised deposition of total nitrogen for the emission year ***ie - DN(ie)*** is defined as:

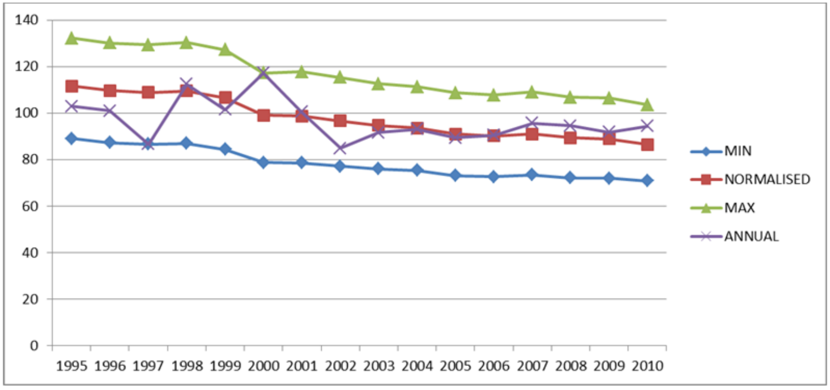
$$\begin{aligned} DN(ie) = MED\{&D^{ox}(ie,1) + D^{rd}(ie,1),\ldots,D^{ox}(ie,im) + D^{rd}(ie,im),\ldots \\ &\ldots,D^{ox}(ie,im) + D^{rd}(ie,im) \end{aligned} \qquad \text{(D8)}$$

In Eq. (D8), MED is the median taken over 16 values which correspond to 17 meteorological years. In addition, the maximum and minimum values are also calculated for each emission year. The results of these calculations for the years 1995-2011 are shown in Figs. 3.12-3.14, for oxidised, reduced and total nitrogen deposition. The normalised depositions for nitrogen are also included in in the Indicator Fact Sheet for nitrogen deposition available on the HELCOM web site. The normalised depositions for HMs and PCDD/Fs are calculated in a very similar way to this described for nitrogen. They are included in the corresponding Indicator Fact sheets for HMs and PCDD/Fs, with the links given in Appendix C.

Normalised deposition of oxidized nitrogen



Normalised deposition of reduced nitrogen



Normalised deposition of total nitrogen

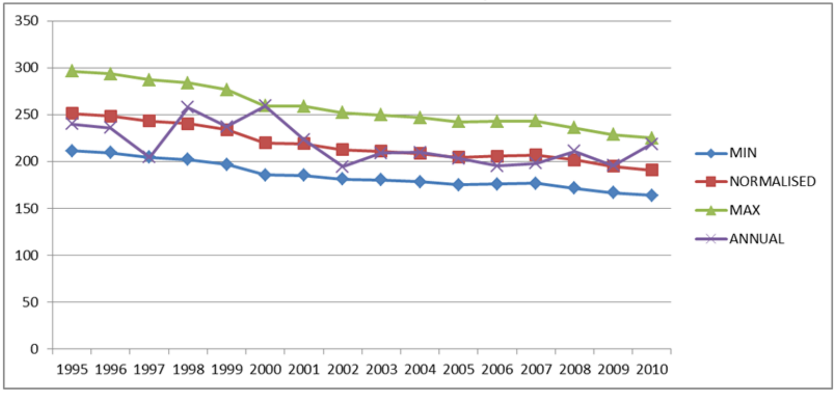


Fig. D2. Normalised depositions of oxidized, reduced and total nitrogen for the period 1995-2010. Minimium, maximum and actual annual values of the deposition are also shown.

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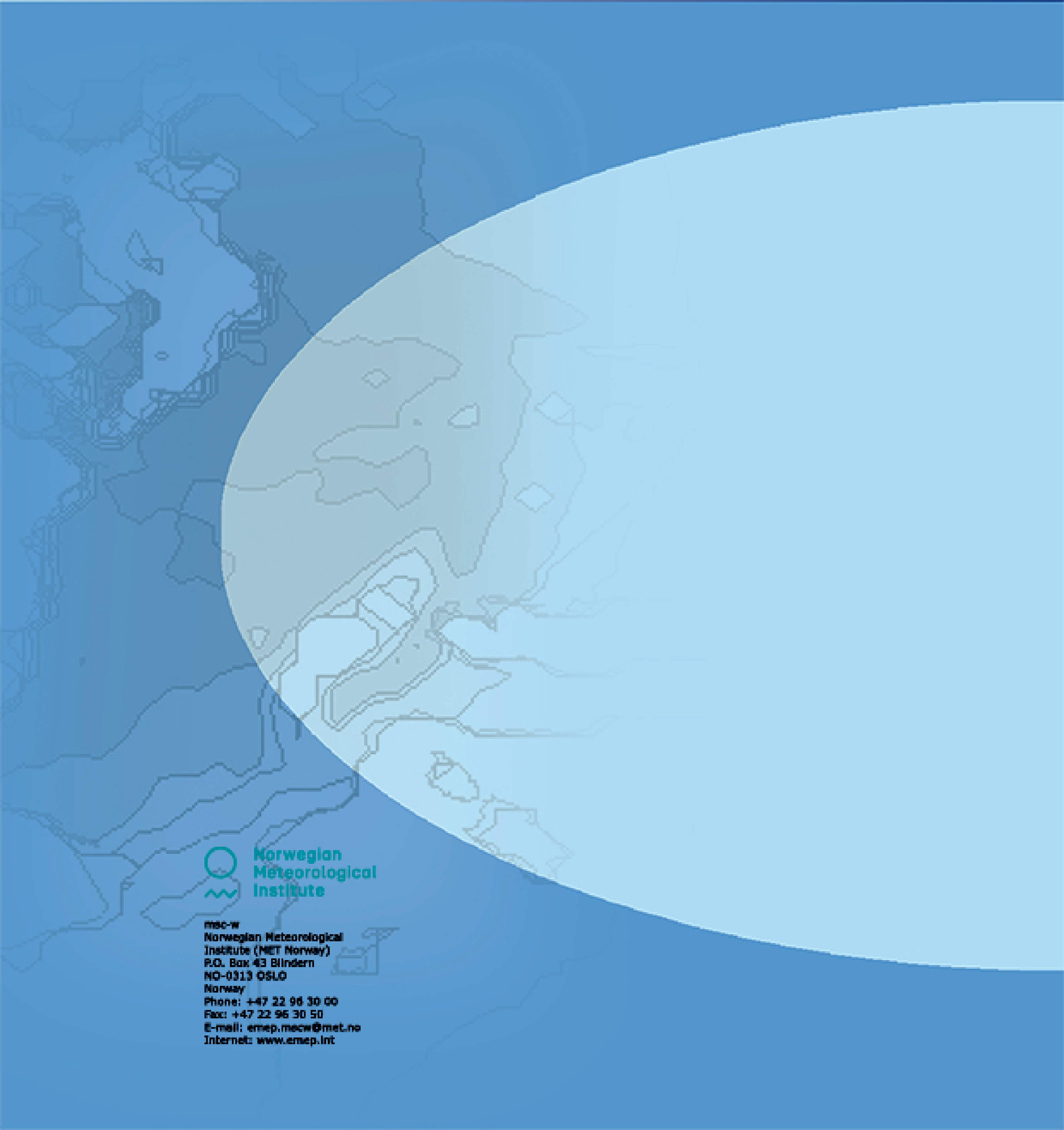
Meteorological Synthesizing Centre – West
Norwegian Meteorological Institute
P.O.Box 43 – Blindern, NO-0313 Oslo, Norway



Norwegian
Meteorological
Institute

msc-w
Norwegian Meteorological
Institute (MET Norway)
P.O. Box 43 Blindern
NO-0313 OSLO
Norway
Phone: +47 22 96 30 00
Fax: +47 22 96 30 50
E-mail: emep.mscw@met.no
Internet: www.emep.int