

4. Atmospheric Supply of Nitrogen to the Baltic Sea in 2001

Nitrogen deposition to the Baltic Sea is caused mainly by the emission sources in the HELCOM countries and ship emissions. However, in some cases, emissions from other sources in the EMEP domain have also important contribution to the deposition. Therefore, in this chapter we present the recent emission data officially submitted to EMEP for individual HELCOM countries, all HELCOM sources, ship emissions and all EMEP sources. All emission data presented here were published in the EMEP report (Vestreng, 2003).

2.1 Nitrogen emissions

Comparisons of annual emissions of nitrogen oxides, ammonia and total nitrogen (nitrogen oxides + ammonia) in the years 2000 and 2001, from individual HELCOM countries, are shown in Figures 4.1, 4.2 and 4.3, respectively.

In case of nitrogen oxides emission, out of nine HELCOM countries, reduction between 1.6% and 7.3% can be noticed in five countries from the year 2000 to 2001 (Figure 4.1). Largest emission reduction, 7.3%, can be seen in the country with relatively small nitrogen oxides emission – Estonia. In three countries, Germany, Lithuania and Latvia, emissions increased by 0.5%, 14.6% and 20%. In the HELCOM Party with the largest emissions, Russian Federation, emissions remain on the same level in 2000 and 2001, but this information is based on interpolation and not on the data which should be officially submitted to EMEP.

In case of ammonia annual emissions in the year 2001 slightly increased in one HELCOM Party, Germany, compared to the year 2000. In Denmark Poland and Sweden, annual emissions were, 1.9%, 4% and 5.3% lower in 2001 than in 2000. Ammonia emissions in Russian Federation remained on the same level in 2000 and in 2001.

Concerning total nitrogen emissions (sum of nitrogen oxides and ammonia) reduction between 2000 and 2001 can be noticed for five HELCOM countries: Finland, Denmark, Sweden, Poland and Estonia, by 1.2%, 2.1%, 3%, 4%, 4.6%, respectively. Increase of annual emissions can be seen in three countries Germany, Lithuania and Latvia by 12.2%, 3.4% and 10.4%, respectively.

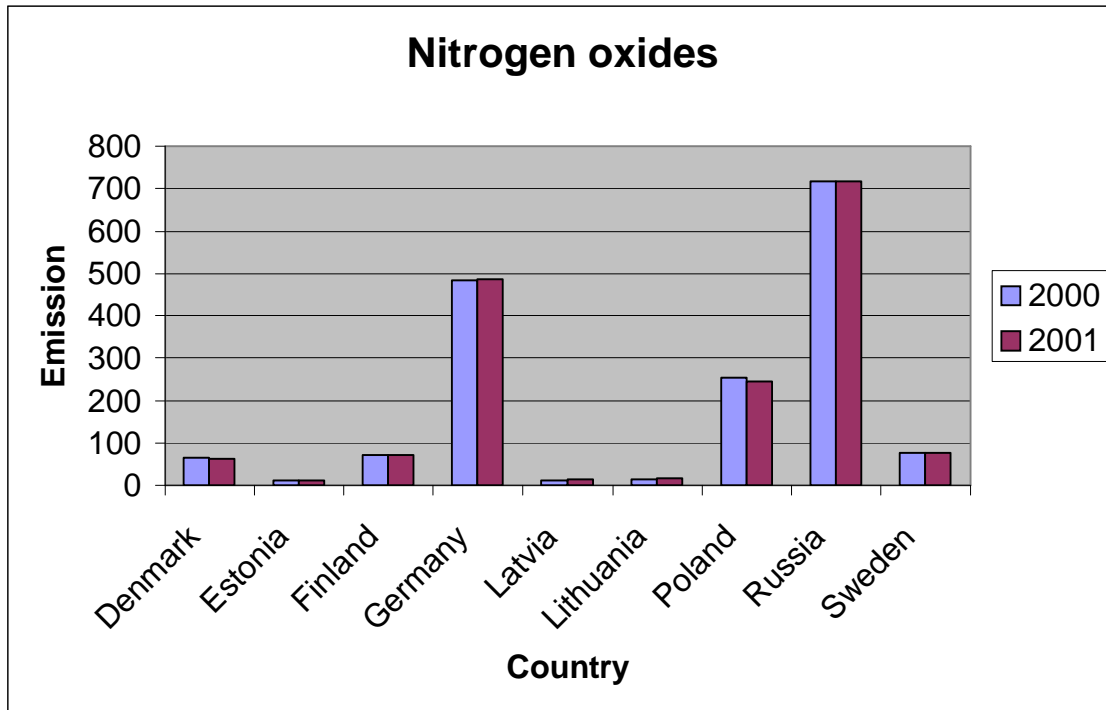


Figure 4.1. Annual emissions of nitrogen oxides from individual HELCOM countries in the years 2000 and 2001. Units: Gg/yr.

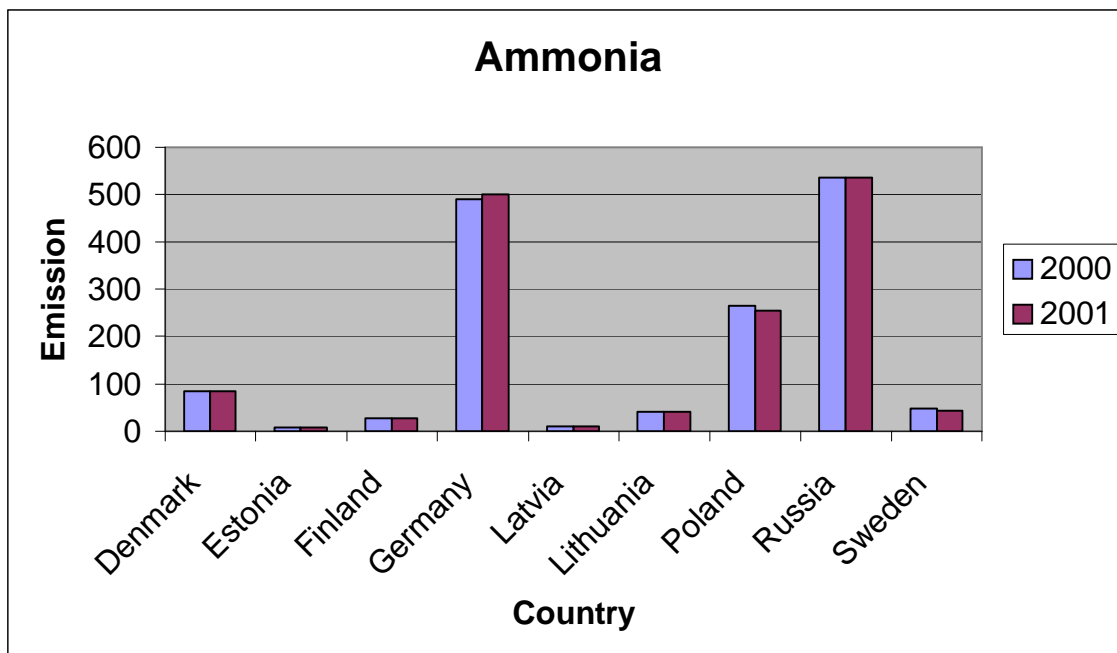


Figure 4.2. Annual emissions of ammonia from individual HELCOM countries in the years 2000 and 2001. Units: Gg N/yr.

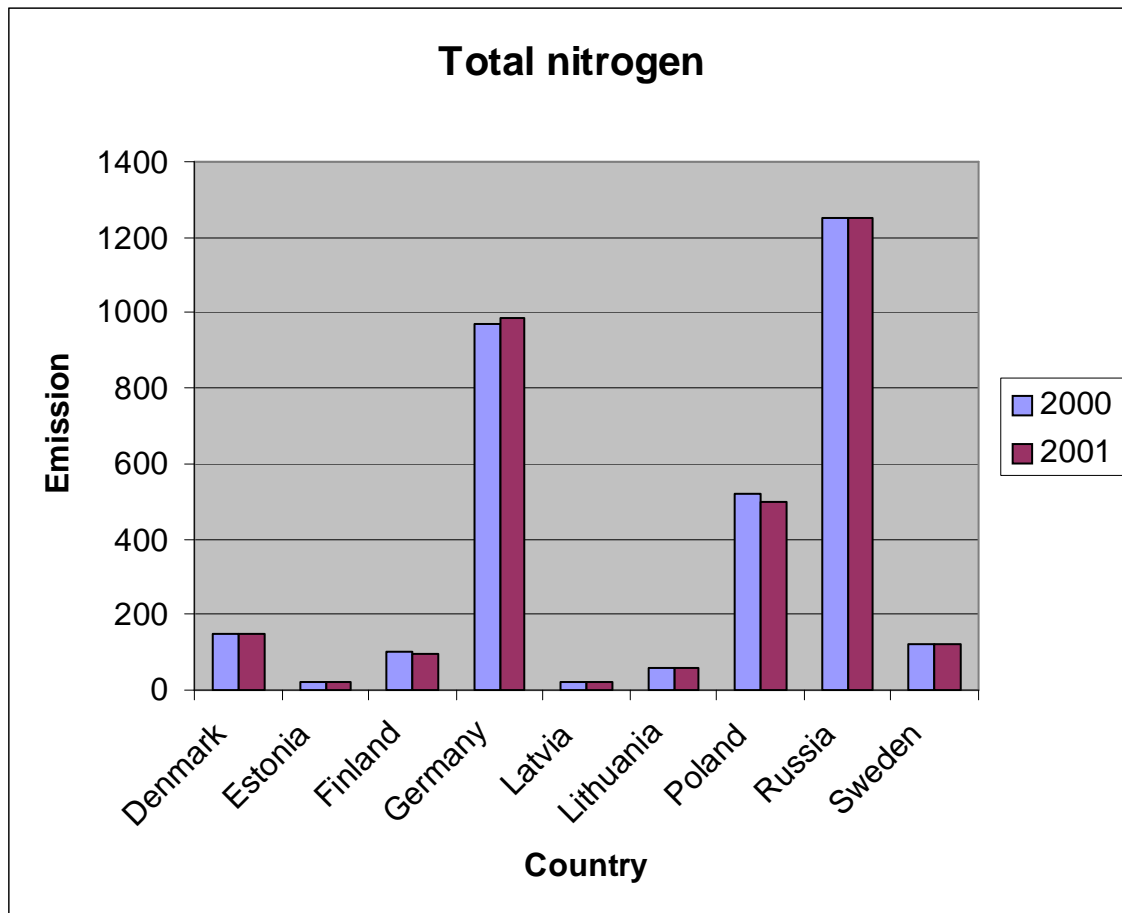


Figure 4.3. Annual emissions of total nitrogen (nitrogen oxides + ammonia) from individual HELCOM countries in the years 2000 and 2001. Units: Gg N/yr.

Annual emissions of oxidized nitrogen, ammonia and total nitrogen from all HELCOM sources (sum of emissions from nine HELCOM countries) in the years 2000 and 2001 are shown in Figure 4.4. Small reduction of these emissions can be noticed between 2000 and 2001, 0.5%, 0.4% and 0.4% for oxidized nitrogen, ammonia and total nitrogen, respectively.

Annual emissions of oxidized nitrogen, ammonia and total nitrogen from the entire model domain (sum of emissions from all EMEP countries and seas) in the years 2000 and 2001 are shown in Figure 4.5. Also in this case a small reduction of the emissions can be noticed between 2000 and 2001, 0.6%, 0.3% and 0.7% for oxidized nitrogen, ammonia and total nitrogen, respectively.

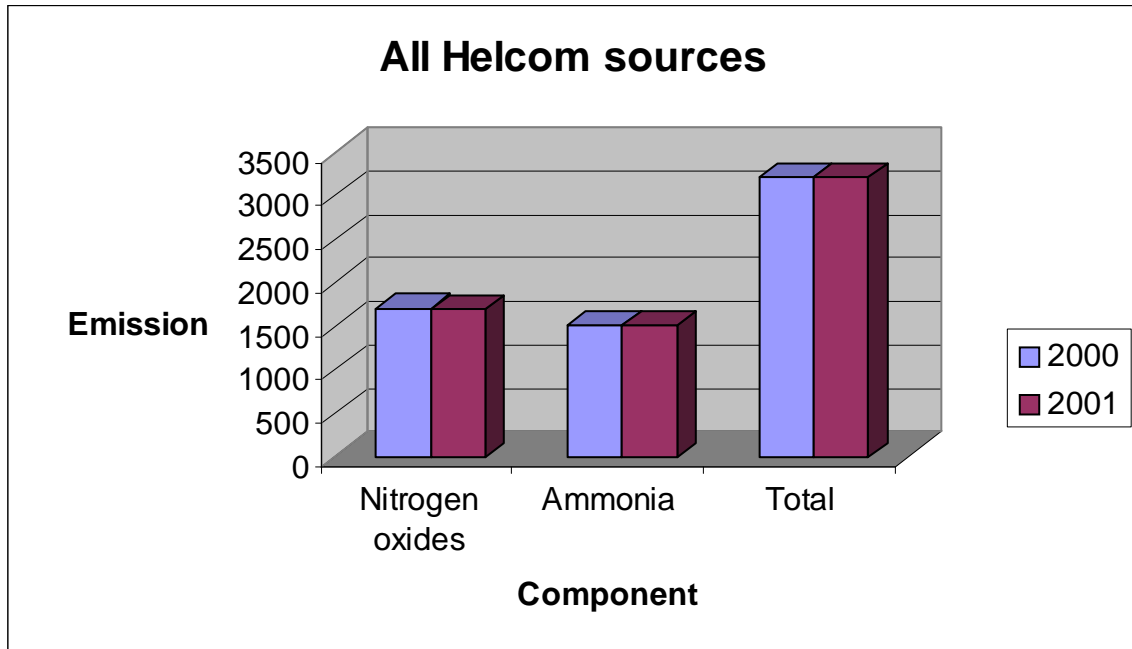


Figure 4.4. Annual emissions of oxidized nitrogen, ammonia and total nitrogen (nitrogen oxides + ammonia) from all HELCOM countries (sum of individual emissions) in the years 2000 and 2001. Units: Gg N/yr.

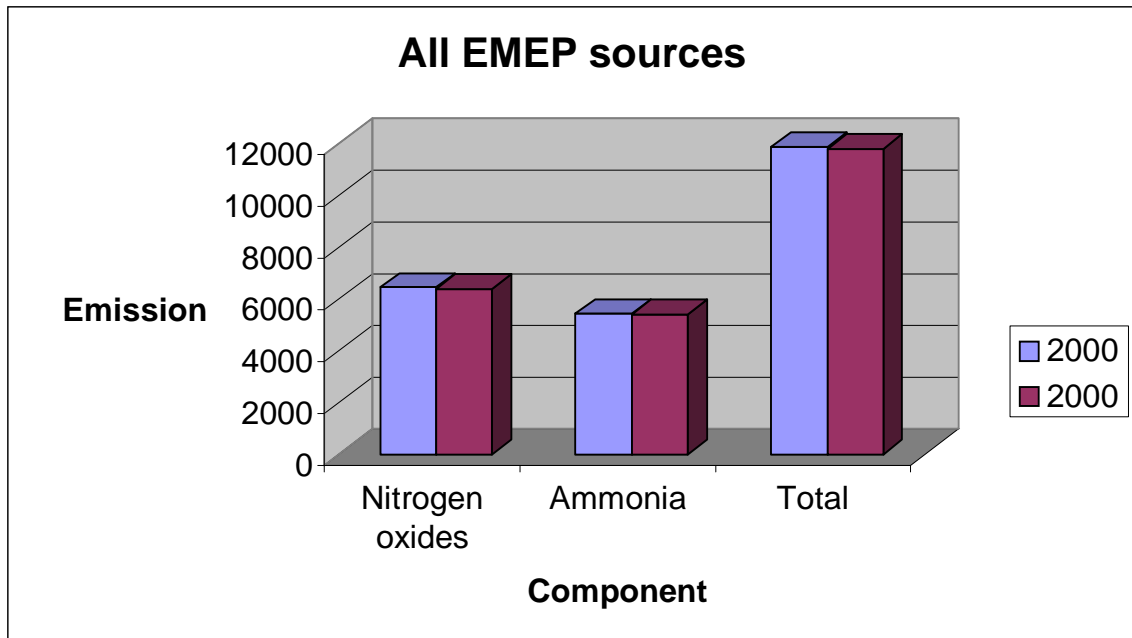


Figure 4.5. Annual emissions of oxidized nitrogen, ammonia and total nitrogen (nitrogen oxides + ammonia) from all HELCOM countries (sum of individual emissions) in the years 2000 and 2001. Units: Gg N/yr.

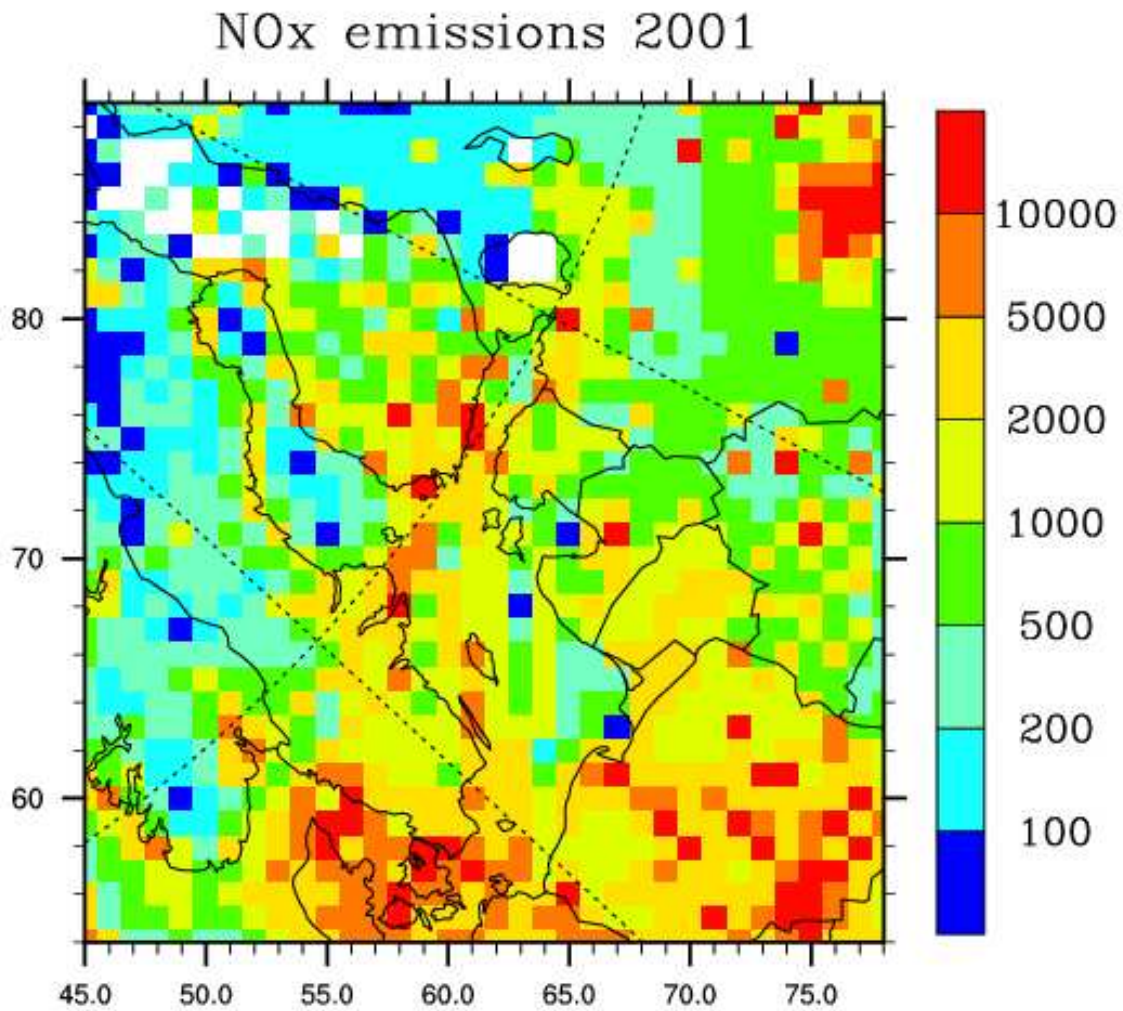


Figure 4.6. Map of annual emission of oxidized nitrogen in the Baltic Sea region in 2001. Units: Mg of N per year and per 50×50 km grid cell.

Spatial distribution of annual 2001 emissions of nitrogen oxides on and around the Baltic Sea is shown in Figure 4.6. Emissions from the international ship traffic on the Baltic Sea, which will be described in more detail later in this Chapter, are included in Figure 4.6. Major local sources of nitrogen oxides are related to large cities and around the Baltic Sea and intensive ship traffic on the sea.

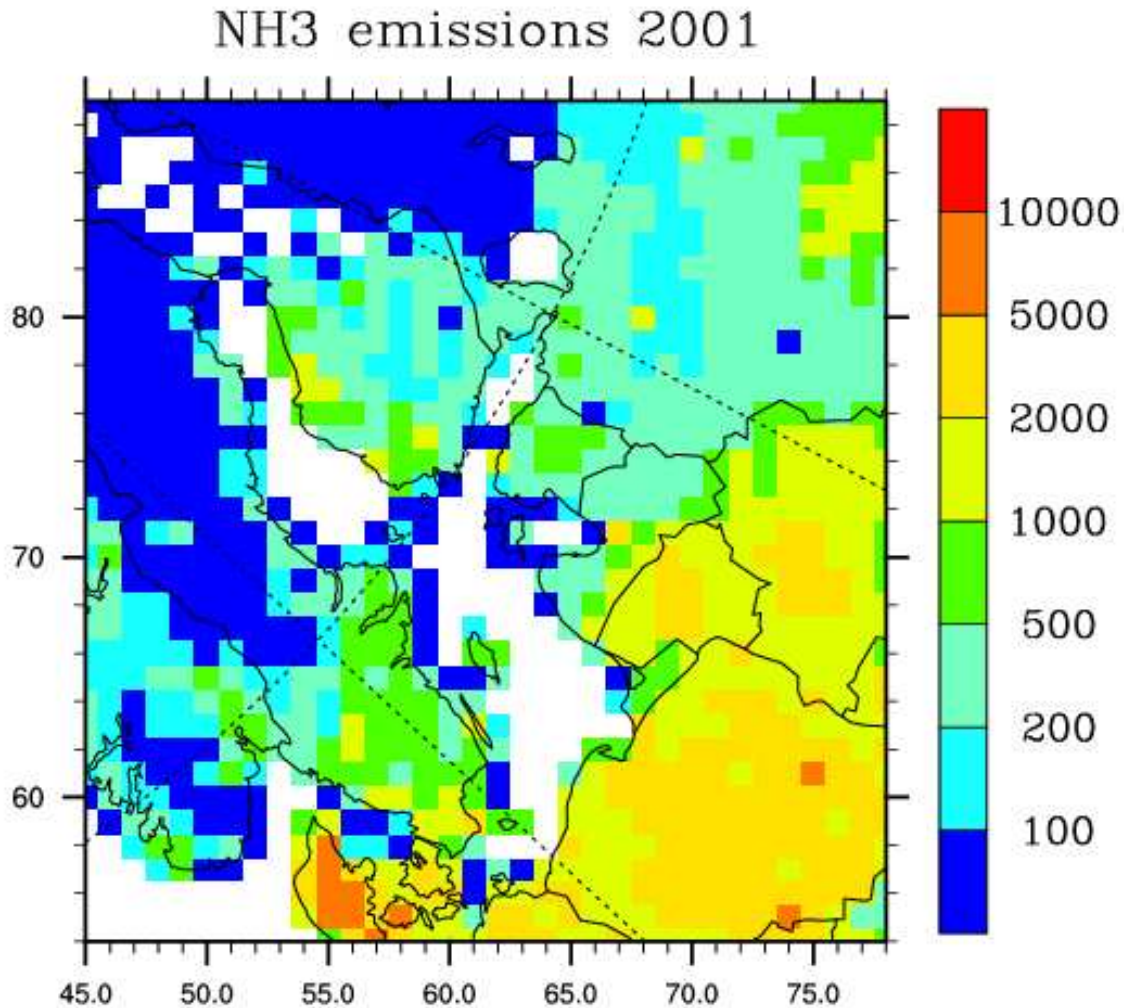


Figure 4.7. Map of annual emission of ammonia in the Baltic Sea region in 2001. Units: Mg of N per year and per 50×50 km grid cell.

Spatial distribution of annual 2001 emissions of ammonia around the Baltic Sea is shown in Figure 4.7. Ammonia emission sources are mostly located in the south and south-west coast, mainly affecting Kattegat and the Belt Sea sub-basin of the Baltic Sea. In this case, there are no emission sources on the sea. A clear south to north gradient of emissions can be noticed in Figure 4.7.

Nitrogen emissions from the international ship traffic on the Baltic Sea, are only available for one year – 1990. Total annual emissions of nitrogen oxides from the international shipping operation on the Baltic Sea are relatively high, 353 ktonnes (NO_x), compared to

annual emissions from the individual HELCOM countries, for the same year. Map of annual nitrogen oxides emissions from the ship traffic on the Baltic Sea in 1990 is shown in Figure 4.8.

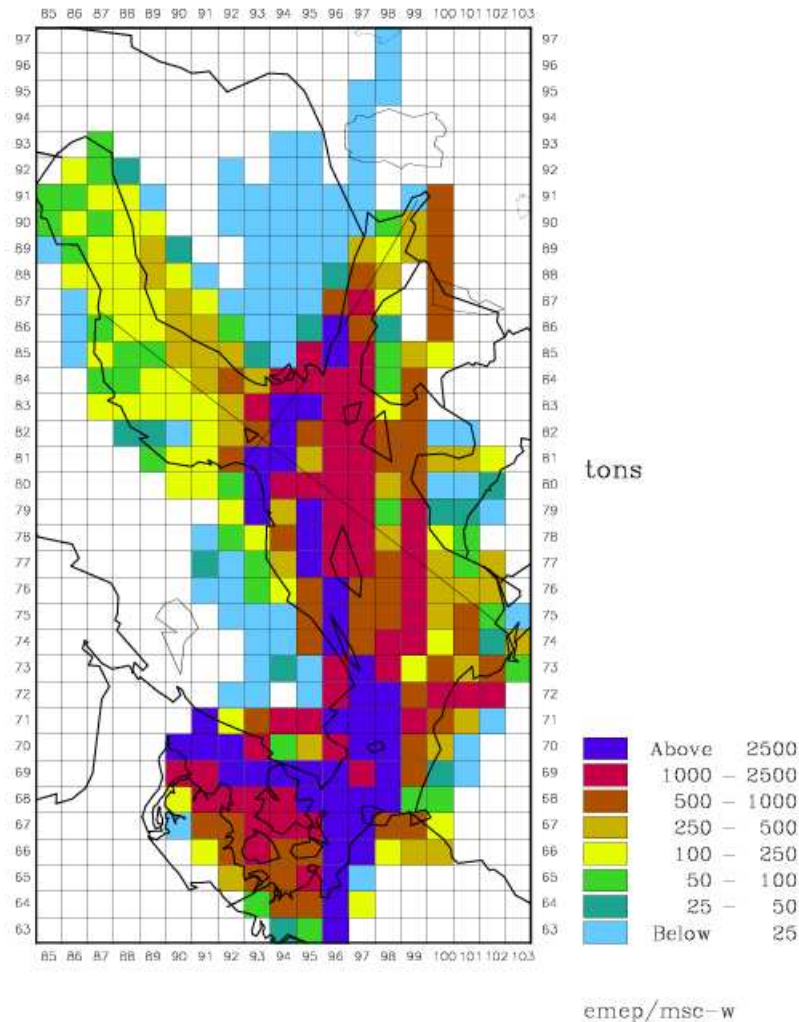


Figure 4.8 Map of annual emissions of nitrogen oxides from the international ship traffic on the Baltic Sea in 1990. Units: Gg of N per year and per 50x50 km grid cell.

Maps with time series of annual emissions from the individual HELCOM countries, for the period 1991 – 2001 are shown in Figures 4.9 and 4.10, for nitrogen oxides and ammonia, respectively. Both, emissions of oxidized nitrogen and reduced nitrogen are significantly lower in 2001 than in 1991.

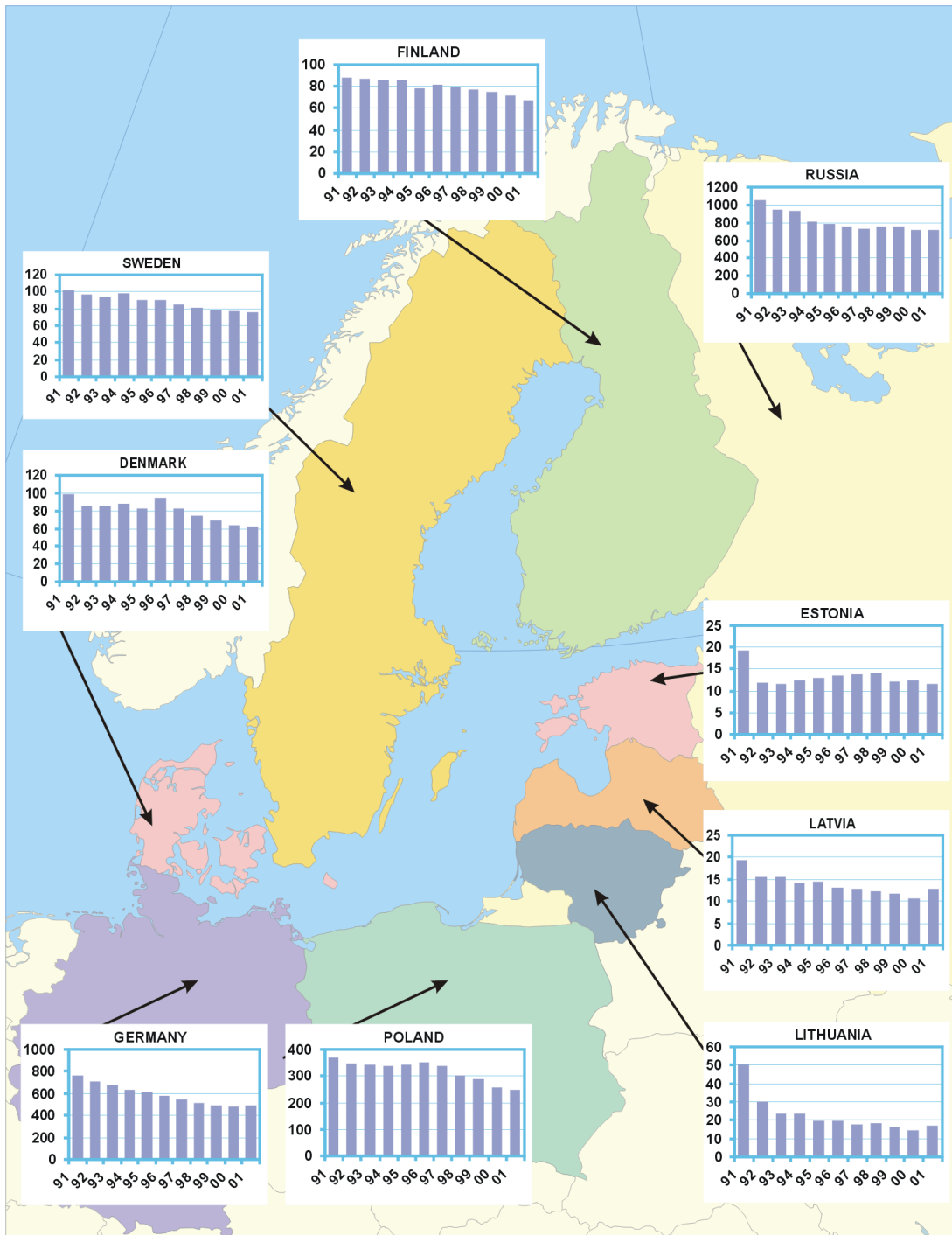


Figure 4.9 Time series of annual ammonia emissions from individual HELCOM countries, for the period 1991 – 2001. Units: ktonnes of N per year.

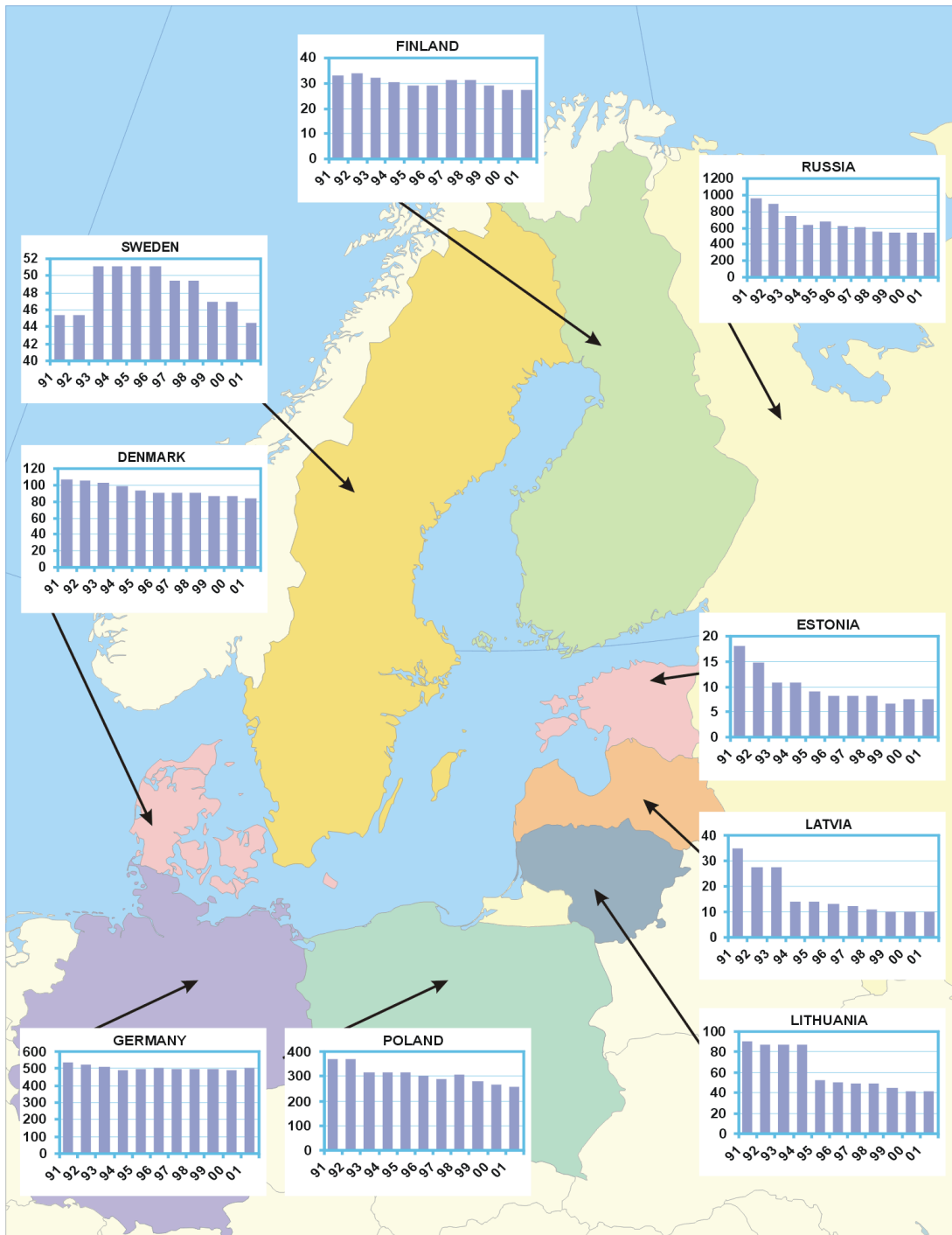


Figure 4.10 Time series of annual emissions of nitrogen oxides from individual HELCOM countries, for the period 1991 – 2001. Units: ktonnes of N per year.

4.2 Annual deposition of nitrogen

Spatial distributions of annual 2001 deposition fluxes of oxidized, reduced and total (oxidized + reduced) nitrogen, on and around the Baltic Sea, are shown in Figures 4.11, 4.12 and 4.13, respectively. There is a clear south-East to North-West gradient in the deposition fluxes. For all three: oxidized, reduced and total nitrogen, the highest deposition fluxes can be noted in the Belt Sea (BES) sub-basin/catchment and the lowest in the Bothnian Bay (BOB) sub-basin/catchment.

Dry and wet annual 2001 deposition fluxes of total (oxidized and reduced nitrogen), on and around the Baltic Sea, are shown in Figures 4.14 and 4.15, respectively. Wet deposition is significantly larger than dry deposition over the entire Baltic Sea region.

Annual 2001 dry, wet and total depositions, as well as total deposition fluxes of oxidized, reduced and total nitrogen are given in Tables 4.1, 4.2 and 4.3 respectively. Table 4.4 presents annual 2001 nitrogen depositions and deposition fluxes of total nitrogen to all catchments of the Baltic Sea. These tables confirm the domination of wet deposition of nitrogen in all sub-basins and catchments of the Baltic Sea. Contribution of wet deposition of nitrogen to total (wet + dry) deposition to the entire Baltic Sea basin in 2001 is 76%.

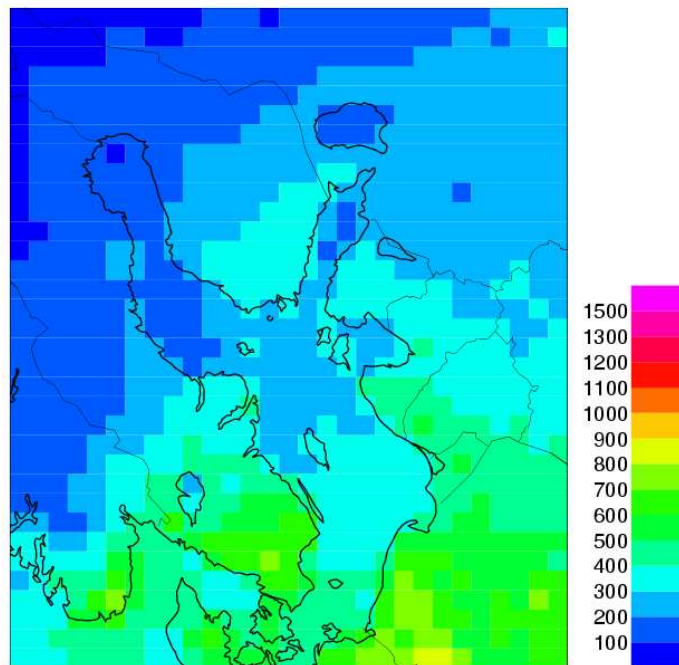


Figure 4.11. Map of annual deposition flux of oxidized nitrogen (dry + wet) in 2001.
Units: mg N m⁻² yr⁻¹.

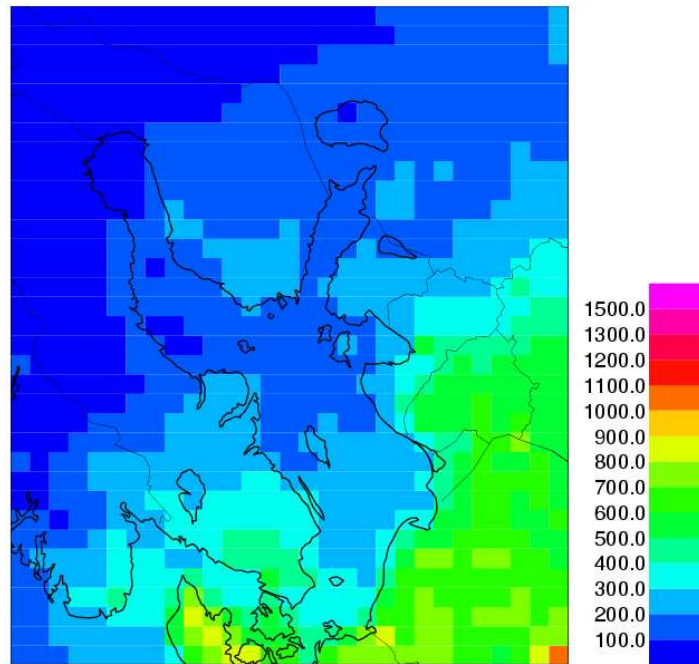


Figure 4.12. Map of annual deposition flux of reduced nitrogen (dry + wet) in 2001.
Units: mg N m⁻² yr⁻¹.

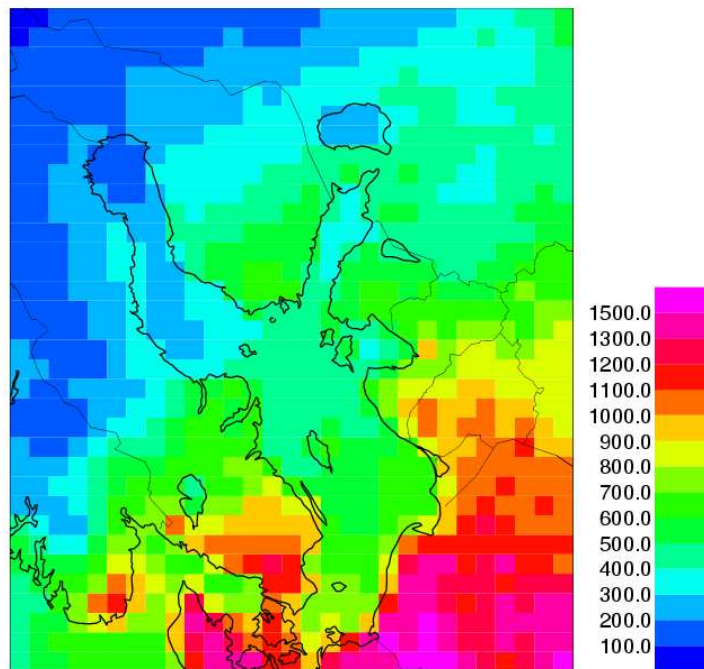


Figure 4.13. Map of annual deposition of total (oxidized + reduced) nitrogen in 2001.
Units: mg N m⁻² yr⁻¹.

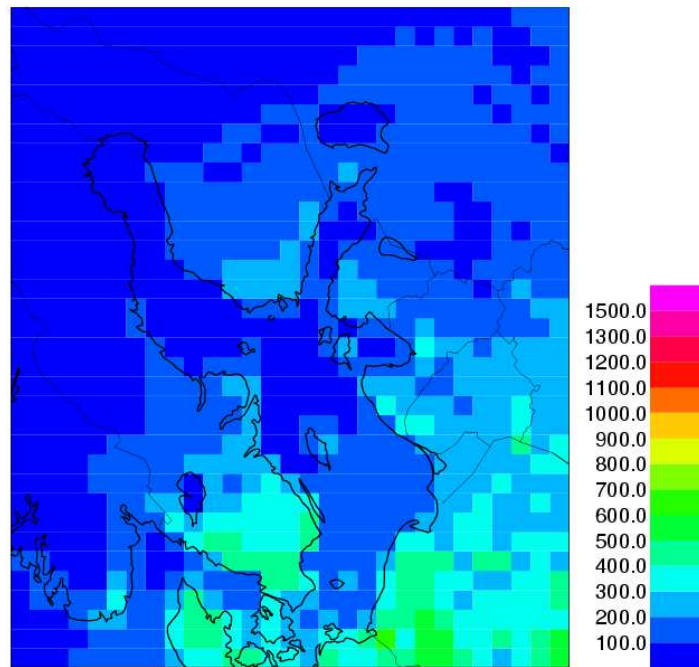


Figure 4.14. Map of annual dry deposition of total nitrogen (oxidized + reduced) in 2001.
Units: mg N m⁻² yr⁻¹.

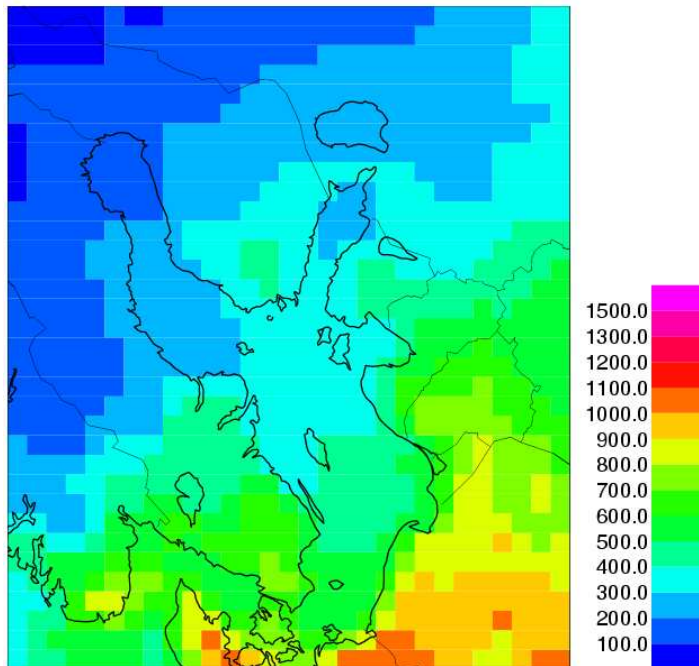


Figure 4.15. Map of annual wet deposition of total nitrogen (oxidized + reduced) in 2001.
Units: mg N m⁻² yr⁻¹.

Table 4.1. Annual dry, wet, and total depositions (ktonnes year⁻¹) and total deposition fluxes (mg m⁻² year⁻¹) of oxidized nitrogen to the Baltic Sea sub-basins in 2001.

Deposition	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
<i>Dry</i>	5.1	2.1	1.6	19.4	2.1	2.2	32.6
<i>Wet</i>	16.0	5.1	3.8	54.1	8.5	7.4	94.9
<i>Total</i>	21.2	7.2	5.4	73.4	10.6	9.7	127.4
<i>Flux</i>	181	240	290	349	518	414	306

Table 4.2. Annual dry, wet, and total depositions (ktonnes year⁻¹) and total deposition fluxes (mg m⁻² year⁻¹) of reduced nitrogen to the Baltic Sea sub-basins in 2001.

Deposition	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
<i>Dry</i>	1.6	0.7	0.6	10.7	4.5	2.7	20.9
<i>Wet</i>	11.7	4.0	3.2	41.5	8.6	6.5	75.5
<i>Total</i>	13.3	4.7	3.8	52.2	13.1	9.2	96.4
<i>Flux</i>	114	156	207	248	641	394	232

Table 4.3. Annual dry, wet, and total depositions (ktonnes year⁻¹) and total deposition fluxes (mg m⁻² year⁻¹) of total nitrogen to the Baltic Sea sub-basins in 2001.

Deposition	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
<i>Dry</i>	6.7	2.8	2.2	30.1	6.6	5.0	53.5
<i>Wet</i>	27.7	9.1	7.0	95.6	17.1	13.9	170.4
<i>Total</i>	34.5	11.9	9.2	125.7	23.8	18.9	223.8
<i>Flux</i>	296	396	497	597	1158	810	538

Table 4.4. Annual dry, wet, and total depositions (ktonnes year⁻¹) and total deposition fluxes (mg m⁻² year⁻¹) of total nitrogen to the Baltic catchments in 2001.

Deposition	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea catchment
<i>Dry</i>	43.3	51.5	26.8	209.8	17.1	28.4	376.8
<i>Wet</i>	118.7	127.2	73.8	528.4	41.1	61.8	951.1
<i>Total</i>	162.0	178.7	100.6	738.1	58.3	90.2	1327.9
<i>Flux</i>	268	398	641	967	1250	1250	636

A map with time series of oxidized, reduced and total (oxidized + reduced) nitrogen annual nitrogen depositions to six sub-basins of the Baltic Sea, in the period 1996 - 2001 is shown in Figure 4.16.

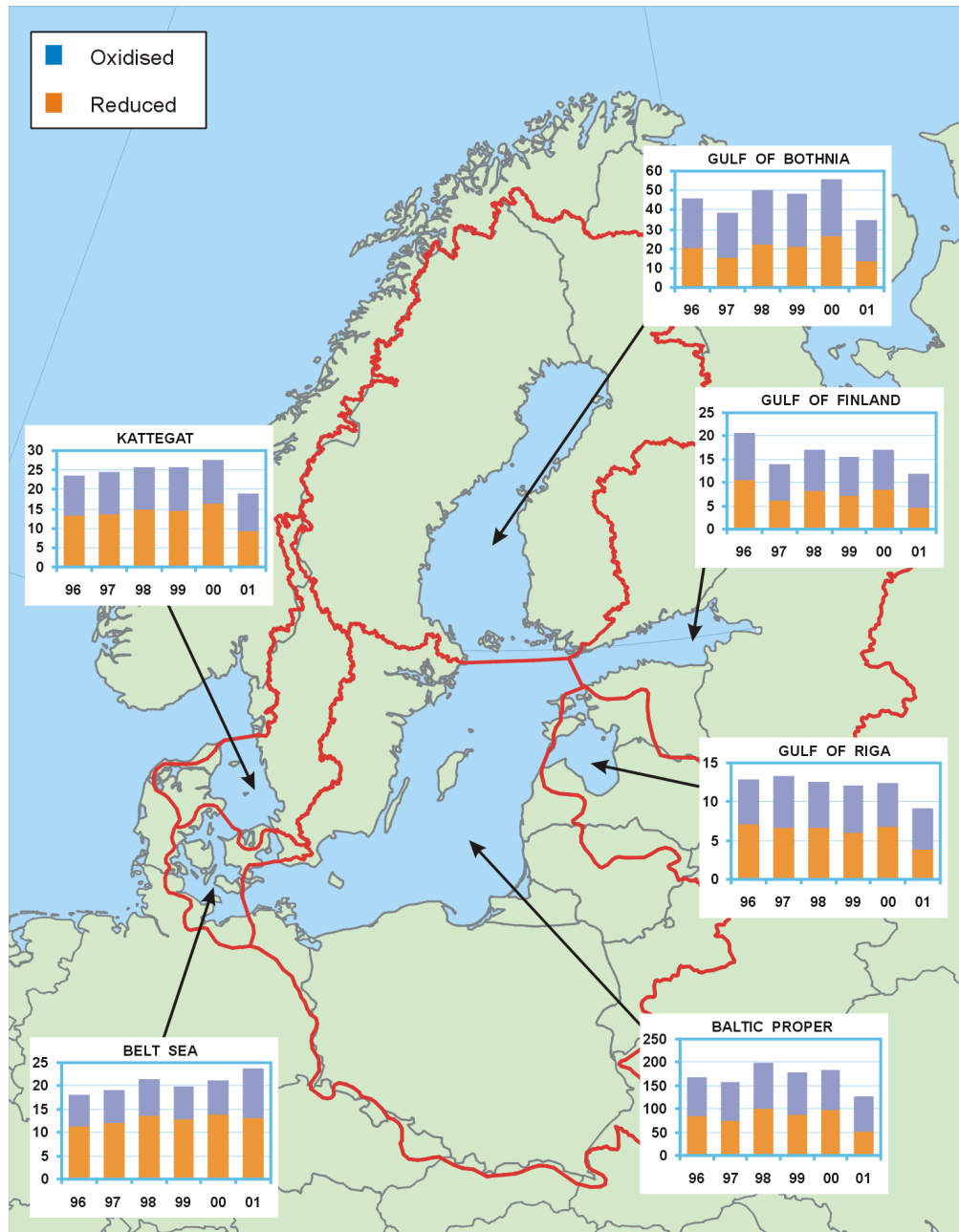


Figure 4.16. Map of annual wet deposition of total nitrogen (oxidized + reduced) in 2001. Units: mg N m⁻² yr⁻¹.

All kinds of nitrogen deposition to five basins (all except Belt Sea) are reaching minimum in 2001. Contrary, deposition of total nitrogen to Belt Sea sub-basin in 2001 is the largest for the entire period. There are several possible reasons for generally lower depositions in 2001: new model version applied to 2001 calculations, different meteorological conditions in 2003 and especially lower precipitation amount in the Central and Northern Baltic in 2001. On the other hand, relatively large annual precipitation amount in the Belt Sea region in 2001 may be the main reason for larger 2001 deposition in this region. Distribution of annual precipitation over the Baltic Sea region in 2001 is shown in Figure 4.17.

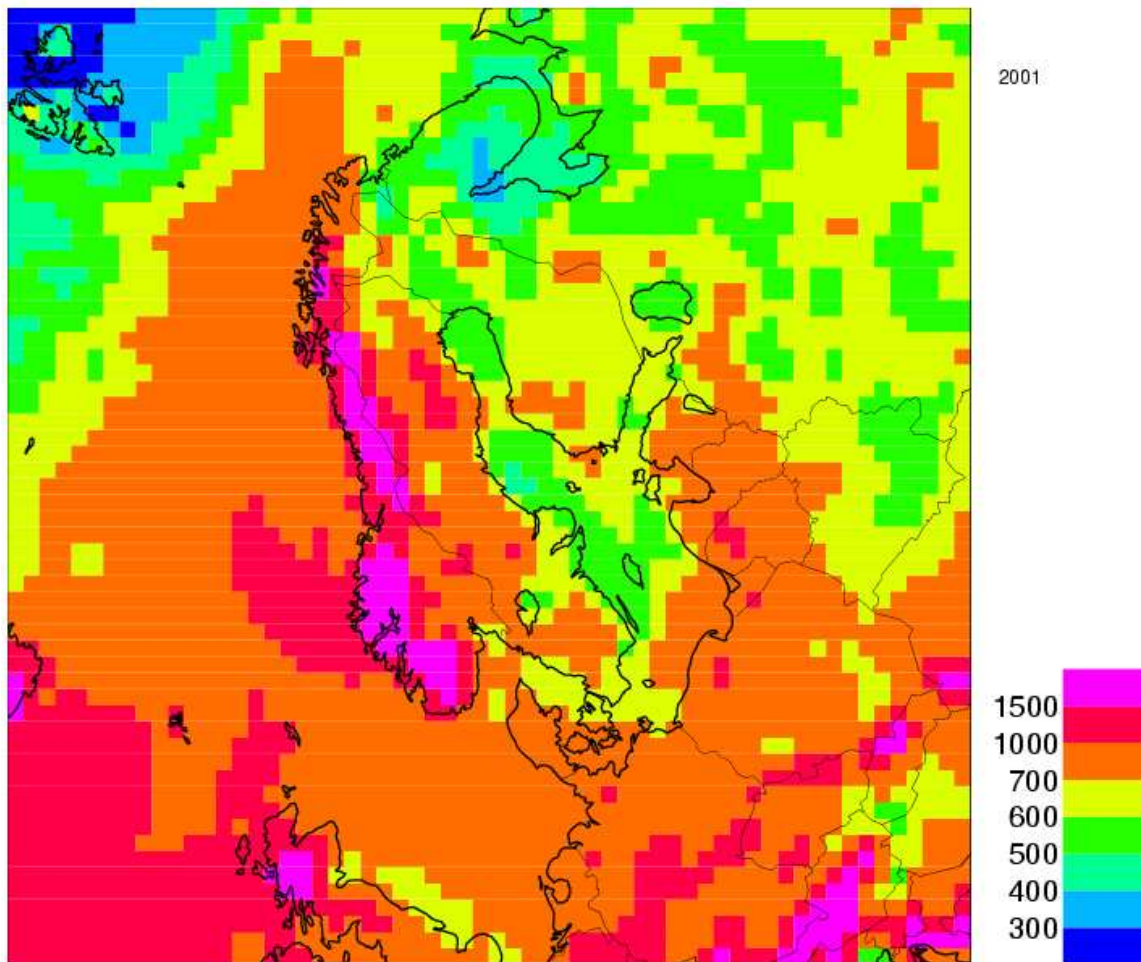


Figure 4.17. Map of annual precipitation amount in 2001. Units: mm.

4.3 Monthly depositions of nitrogen

Monthly 2001 depositions of oxidized, reduced and total nitrogen to the entire Baltic Sea basin and the Baltic Sea catchment are shown in Figures 4.18 and 4.19, respectively. There is no regular pattern in the deposition, except the period between May and September. In this period, all deposition types to the Baltic Sea basin have minimum in May and then they systematically increase until September, when they reach the maximum. Such a pattern is mostly caused by weather conditions and monthly variations of nitrogen emissions.

Maxima of all depositions to the Baltic Sea catchment, but especially maximum of reduced nitrogen deposition, occur in April when intensive agricultural activities take place over the land.

4.4 Source allocation of nitrogen deposition

Not only local emission source of nitrogen (HELCOM Parties, Ship traffic) but also distant emission sources (e.g. UK and Italy) contribute to nitrogen deposition into the Baltic Sea.

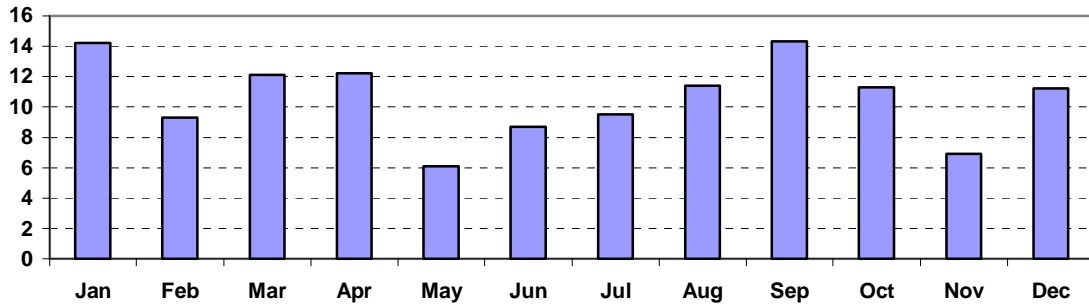
Comparison of contributions of nitrogen oxides emissions from the HELCOM Parties to oxidized nitrogen deposition into the Baltic Sea Basin in the years 1997 and 2000 is given in Figure 4.20. Similar comparison for ammonia emissions and reduced nitrogen depositions is shown in Figure 4.21.

In case of nitrogen oxides emissions, ship traffic and emissions from Germany and Poland are the main contributors to the deposition, both in 1997 and in 2000. Major difference between 1997 and 2000 is significant rise of the German and Polish contributions with small decrease of emissions in the Scandinavian countries. Contributions of emissions from Estonia, Lithuania and Latvia are relatively small among HELCOM Parties.

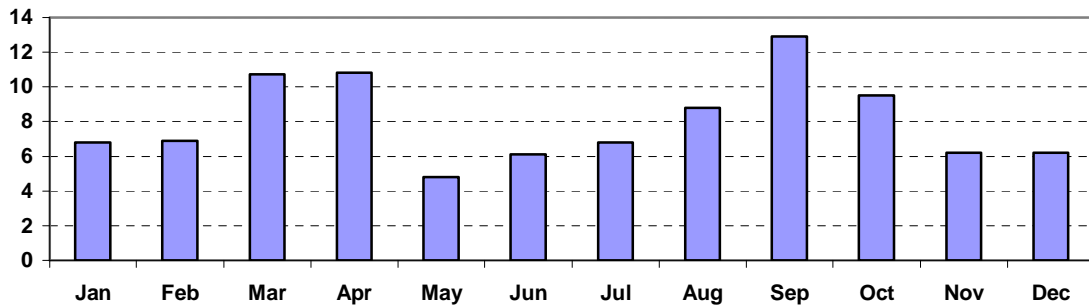
Annual emissions of ammonia from Germany and Poland are the main contributors to reduced nitrogen deposition in the Baltic Sea, especially in 2000. However, emissions from Denmark and Sweden also contribute substantially to the deposition, especially in 1997. A sharp reduction of the Estonian and Swedish contributions can be noticed between 1997 and 2000, whereas contribution of Polish emissions is definitely higher in 2000 than in 1997.

Ten countries (sources) with highest contributions of nitrogen oxides emissions to annual deposition of oxidized nitrogen into the Baltic Sea basin in the year 2000 are shown in Figure 4.22. Ten countries with highest contributions of ammonia emissions to annual deposition of reduced nitrogen into the Baltic Sea basin in the year 2000 are shown in Figure 4.23.

(a) oxidized nitrogen



(b) reduced nitrogen



(c) total (oxidized + reduced) nitrogen

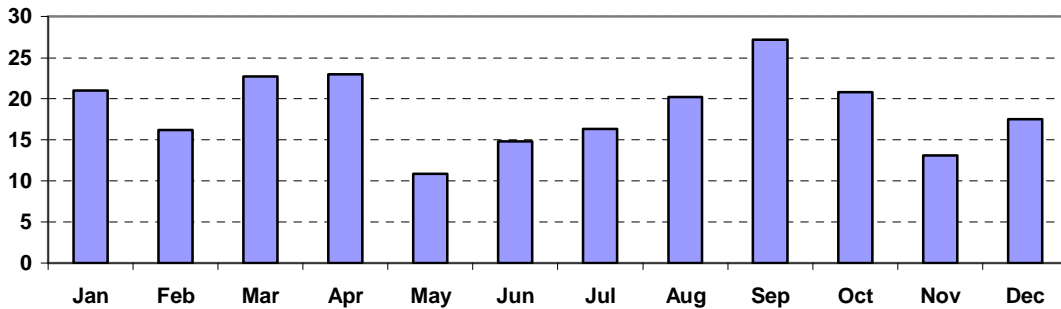
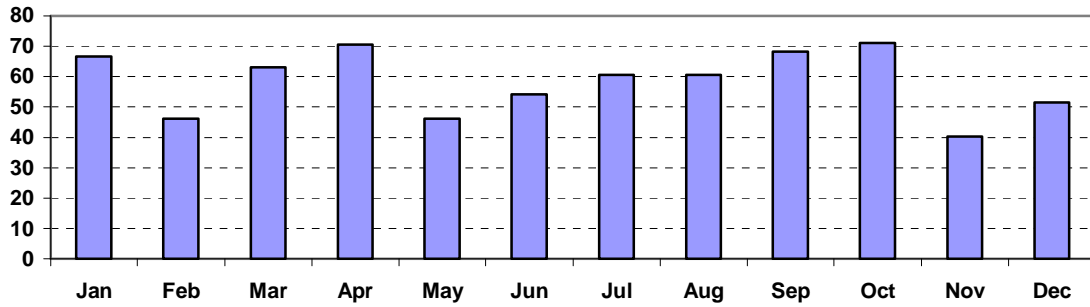
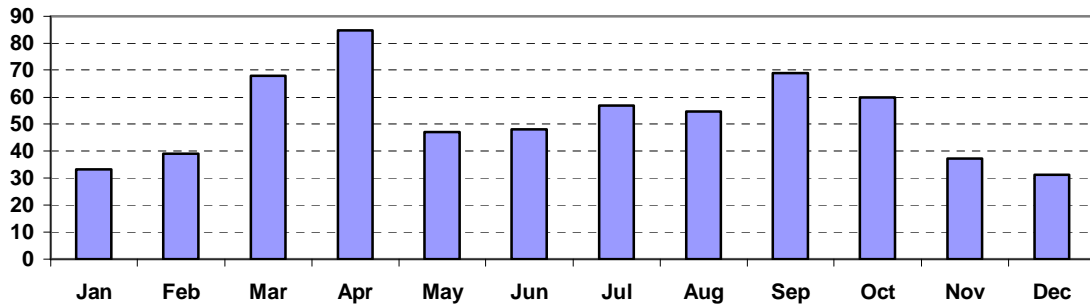


Figure 4.18. Monthly depositions of: a) oxidized, b) reduced and c) total (oxidized +reduced) nitrogen to the entire Baltic Sea basin in 2001. Units: ktonnes N month⁻¹.

(a) oxidized nitrogen



(b) reduced nitrogen



(c) total (oxidized + reduced) nitrogen

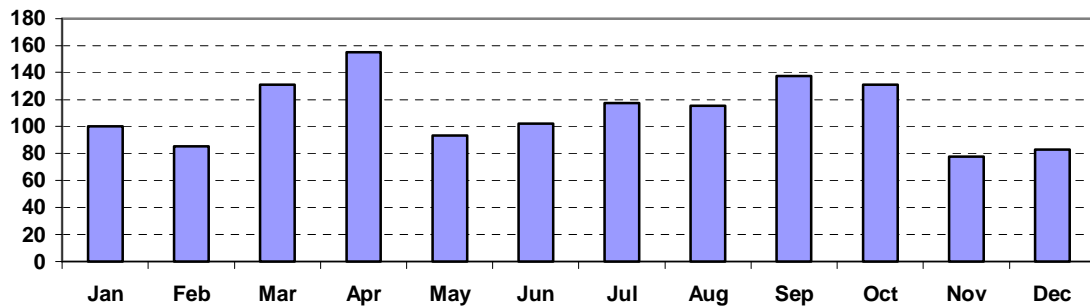


Figure 4.19. Monthly depositions of: a) oxidized, b) reduced and c) total (oxidized +reduced) nitrogen to the entire Baltic Sea catchment in 2001. Units: ktonnes N month⁻¹.

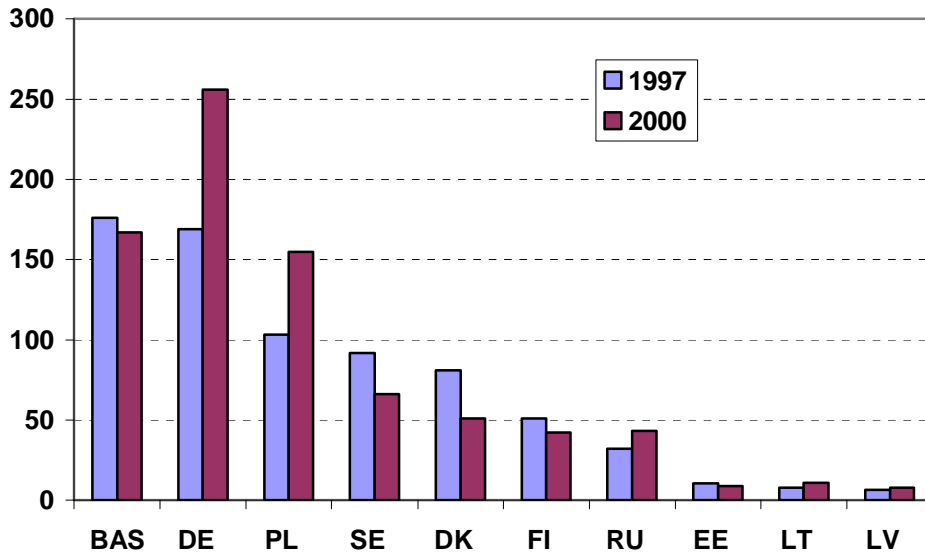


Figure 4.20. Contributions of the HELCOM Parties annual nitrogen oxides emissions to annual deposition of oxidized nitrogen into the Baltic Sea basin. Units: ktonnes N year⁻¹.

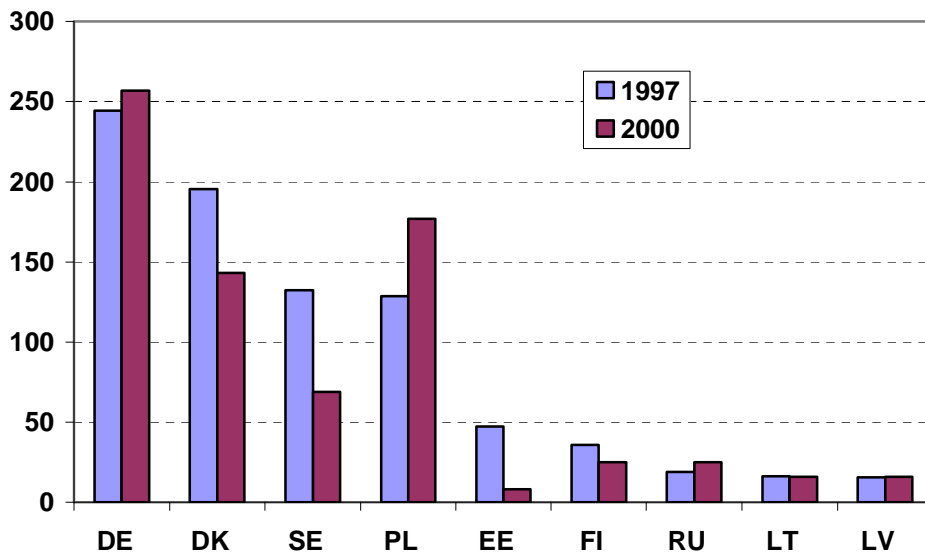


Figure 4.21. Contributions of the HELCOM Parties annual ammonia emissions to annual deposition of reduced nitrogen into the Baltic Sea basin. Units: ktonnes N year⁻¹.

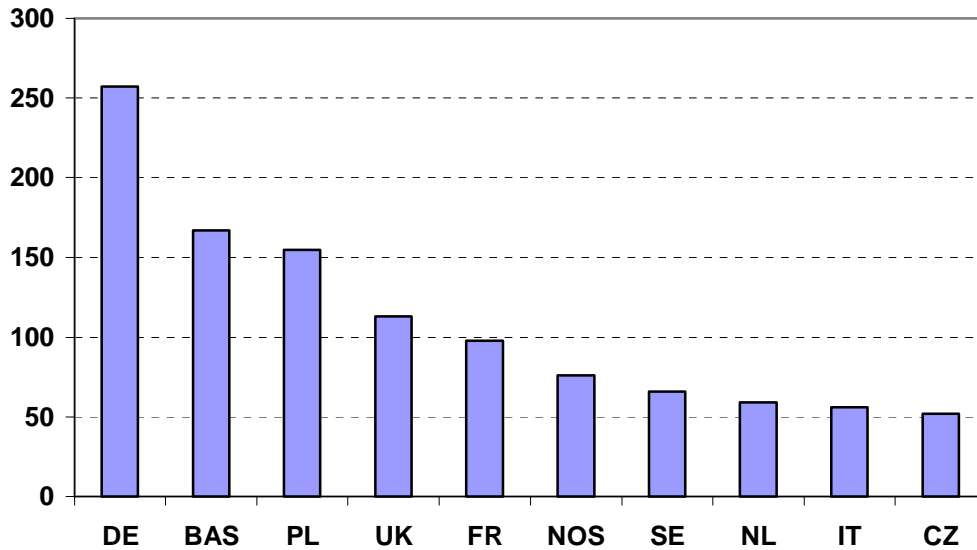


Figure 4.22. Top ten countries (sources) with highest contributions of nitrogen oxides emissions to annual deposition of oxidized nitrogen into the Baltic Sea basin in the year 2000. Units: ktonnes N year⁻¹.

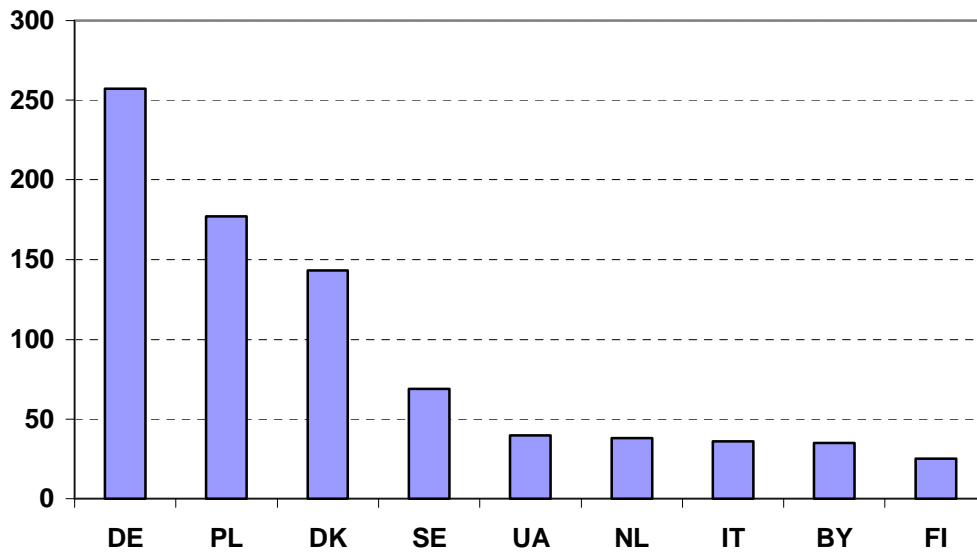


Figure 4.23. Top ten countries with highest contributions of ammonia emissions to annual deposition of reduced nitrogen into the Baltic Sea catchment in the year 2000. Units: ktonnes N year⁻¹.

Both, for oxidized and reduced nitrogen deposition, Germany is definitely the largest contributor. In case of oxidized nitrogen deposition, contribution of emissions from the international ship traffic on the Baltic Sea is the second largest and it is approximately the same as contribution of Polish sources (third largest). In this case, contribution of distant sources, like UK, French and Italian emissions, is also significant. It is interesting to observe that the contribution of emissions from the international ship traffic over North Sea is also large – number six in the list.

Information about nitrogen oxide emissions from the international ship traffic on the Baltic Sea, used in the model calculations is relatively old – from 1990. Most likely, these emissions remain on the same level or are even higher at present than in 1990. Therefore, it is important to use the updated inventory as soon as they are available.

In case of reduced nitrogen deposition, contribution of emissions from the HELCOM Parties, and especially from Germany, Poland, Denmark and Sweden, dominate in the deposition. However distant countries, such as Netherlands and Italy are also among ten major contributors to the deposition.

4.5 Comparison of model results with measurements

The EMEP Unified Eulerian model system has undergone a major overhaul the last two years, where the previous EMEP models (Lagrangian as well as Eulerian) have been merged and re-written in order to produce the Unified EMEP Eulerian model. The model has been carefully documented in EMEP Status Report 1/2003, Part I and verified against measurement data at EMEP stations for nine different years (1980, 1985, 1990, 1995-2000) in EMEP Status Report 1/2003, Part II. Model verification for 2001 has been shown in Part III.

The agreement between model predicted and observed air and precipitation data heavily depends upon an adequate description of emissions. This includes both a reasonable estimates of national totals, gridded (source sector) data and temporal distribution of emissions. As described in detail in EMEP/MS-C-W Note 1/2003 (Vestreng, 2003), the method for creating gridded sector emission data (GS) used as input to the model has been substantially revised this year. When available, reported GS data are used directly in the model calculations, but at present this applies only for 11 Parties. For all other countries, expert estimates on the gridded and sectoral distribution of emissions need to be developed. This year information on large point sources and population statistics have been extensively used in the new scaling procedure to derive gridded sector data when information from the Parties are missing.

Thus, the results presented this year differ from previous years calculations due to 1) revision of the Unified EMEP model and 2) review and revision of the emissions.

In this section we will concentrate on the model performance for nitrogen compounds at the HELCOM sites. Note that the agreement between model results and observations depends not only on the model performance and the adequacy of emissions, but also on the quality and representativeness of the measurement sites. Thus, the following discussion on model *underestimation* and *overestimation* simply imply that the calculated values are lower or higher than the observations, and does not refer to model deficiency only.

4.5.1 Air concentrations

Measurements of particulate ammonium and nitrate are available only from three sites, RU16, LV10 and PL4. Therefore, it is difficult to conclude anything on the model performance for these compounds based on the HELCOM measurements. However, observations for the sum of ammonia plus ammonium and total nitrate (nitric acid plus particulate nitrate) have been reported by 8 HELCOM sites.

In figure 4.23 we present monthly time-series for $\text{NH}_3+\text{NH}_4^+$ for model versus observations. The overall agreement between measurement and model is good. It is especially encouraging that the model manages to reproduce the seasonal cycle of ammonia plus ammonium. The model shows, however, a tendency to overestimate winter concentrations and underestimate summer concentrations. In Fagerli *et al.* (2003) it was suggested that the overestimation in winter is caused by an overestimation of ammonium aerosol. The underestimation of summer concentrations compared to measurements is probably due to problems in the modeling of the spatially variable ammonia gas in combination with influence of local sources on measurements.

In figure 4.24 we present monthly time-series for the sum of nitric acid and nitrate in air for HELCOM stations that delivered data for 2001. The observed seasonal variation of total nitrate in air is well reproduced by the model. However, the model predicts somewhat higher nitrate concentrations in air than measurements, especially in winter. This may be caused by too high conversion of nitric acid to nitrate in the cold periods. Preliminary results (Fagerli *et al.* 2003) suggest that the implementation of a more comprehensive equilibrium chemistry module (EQSAM, Metzger *et al.* (2002a), Metzger *et al.* (2002b)) gives total nitrate concentrations in better agreement with observations due to a more correct gas-particulate partitioning.

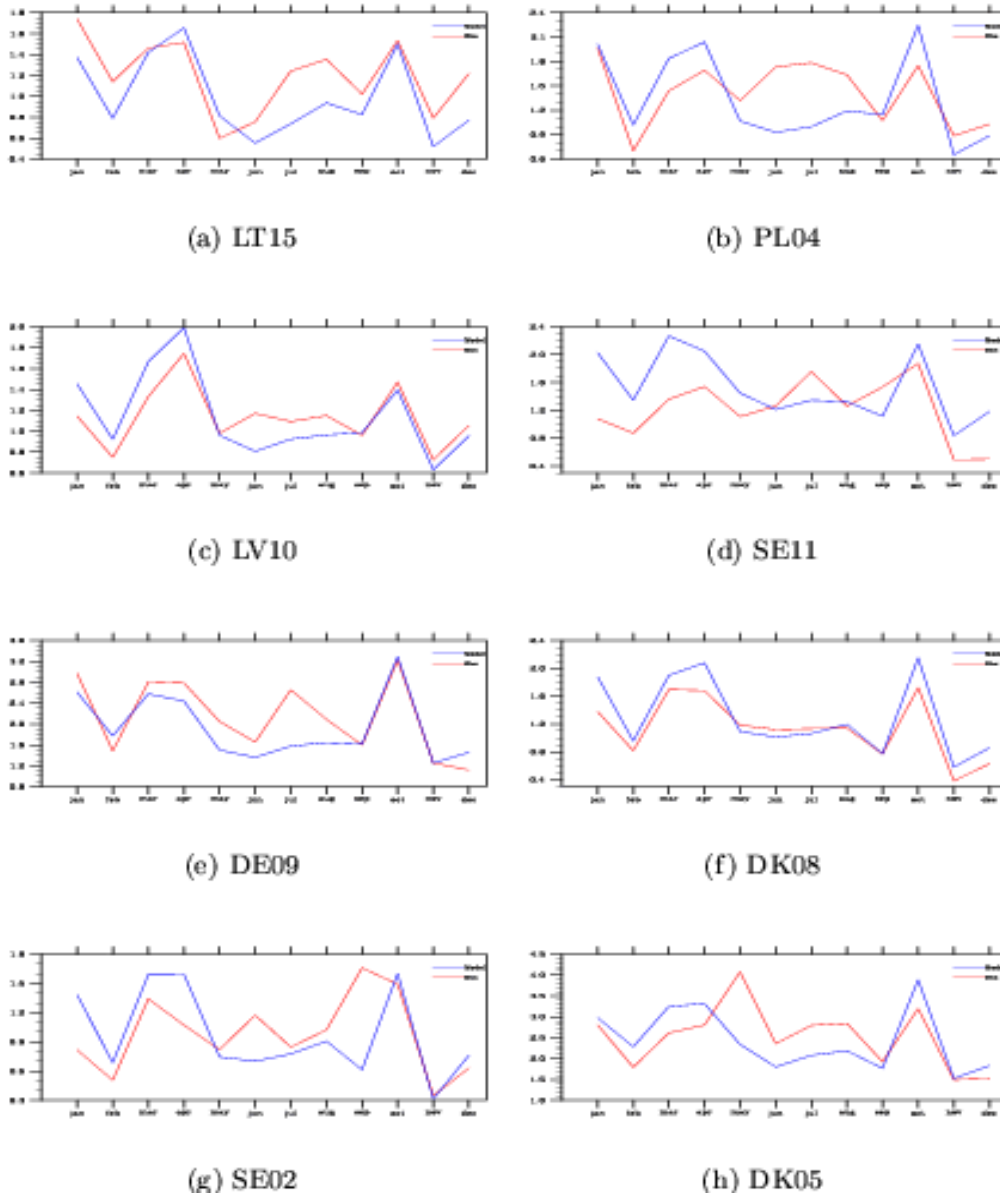


Figure 4.23. Monthly time-series for modelled versus measured concentrations of ammonia plus ammonium in air in 2001. Units: mg N m^{-3} .

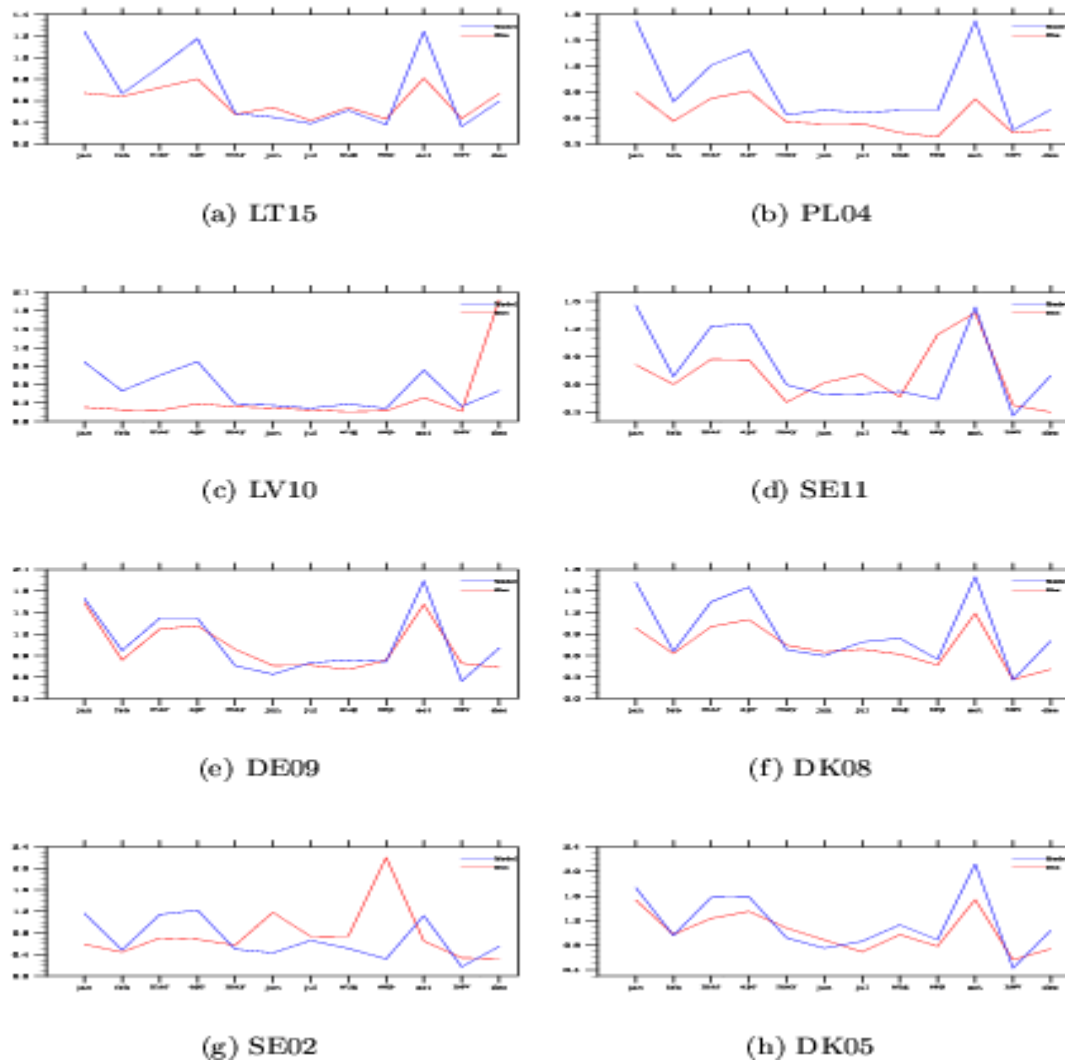


Figure 4.24. Monthly time-series for modelled versus measured concentrations of nitric acid plus ammonium nitrate in air in 2001. Units: $\mu\text{g N m}^{-3}$.

4.5.2 Concentrations in precipitation

The correlation between model and measurements for concentrations in precipitation and wet depositions will to a large extent depend on the modeled precipitation field. However, the precipitation field pattern is very patchy (e.g. influenced by local topographic effects), and the regional scale EMEP model is unable to resolve this sub grid scale distribution. A typical problem arises with small scale showers. In reality

precipitation is high in a small area of a given grid, but a large fraction of the grid should remain dry. Within the model, however, this precipitation is averaged out to cover the whole grid at a lower intensity. Thus, even though average precipitation amounts may be simulated well, the model experiences precipitation more often, but in lower amounts, than occur in reality.

For the reasons given above, it is clear that the comparison results for components in precipitation are expected to be worse than those for air concentrations.

In Figures 4.25 and 4.26 we compare modelled and measured monthly concentrations of oxidized and reduced nitrogen in precipitation. For the majority of the stations, the modelled and measured concentrations agree well. However, concentrations of ammonium and nitrate in precipitation are somewhat underestimated. It is well known (e.g. Cape and Leith, 2002) that dry deposition of NH_3 to open bulk collectors can account for a substantial part of the measured wet deposition. Thus, the apparent under prediction of wet depositions may partly be caused by the bias in measured wet deposition. Further work is needed in order to fully understand the reason for the discrepancy between modelled and measured nitrate and ammonium wet depositions.

4.5.3 Concluding remarks

In general, the comparison between model and measurements for concentrations of nitrogen compounds is good and comparable to the model performance for the EMEP sites. Thus, the model provides a reliable tool in order to assess the nitrogen supply to the Baltic Sea.

It should be noted, however, that in order to make a proper evaluation of model performance, monthly measurements are not satisfactory. For example, it happens that measurements are contaminated or that a measurement site is closed down for a day or two during a month. When comparing the model and the measurement data, the comparison should be done only for days when both modeled and measured data are available. In addition, daily data is needed in order to examine e.g. how the model performs during specific meteorological situations. Thus, to a large extent we rely on the evaluation of the Unified EMEP model against daily measurements from the total EMEP network when drawing conclusions on the model performance for the HELCOM sites.

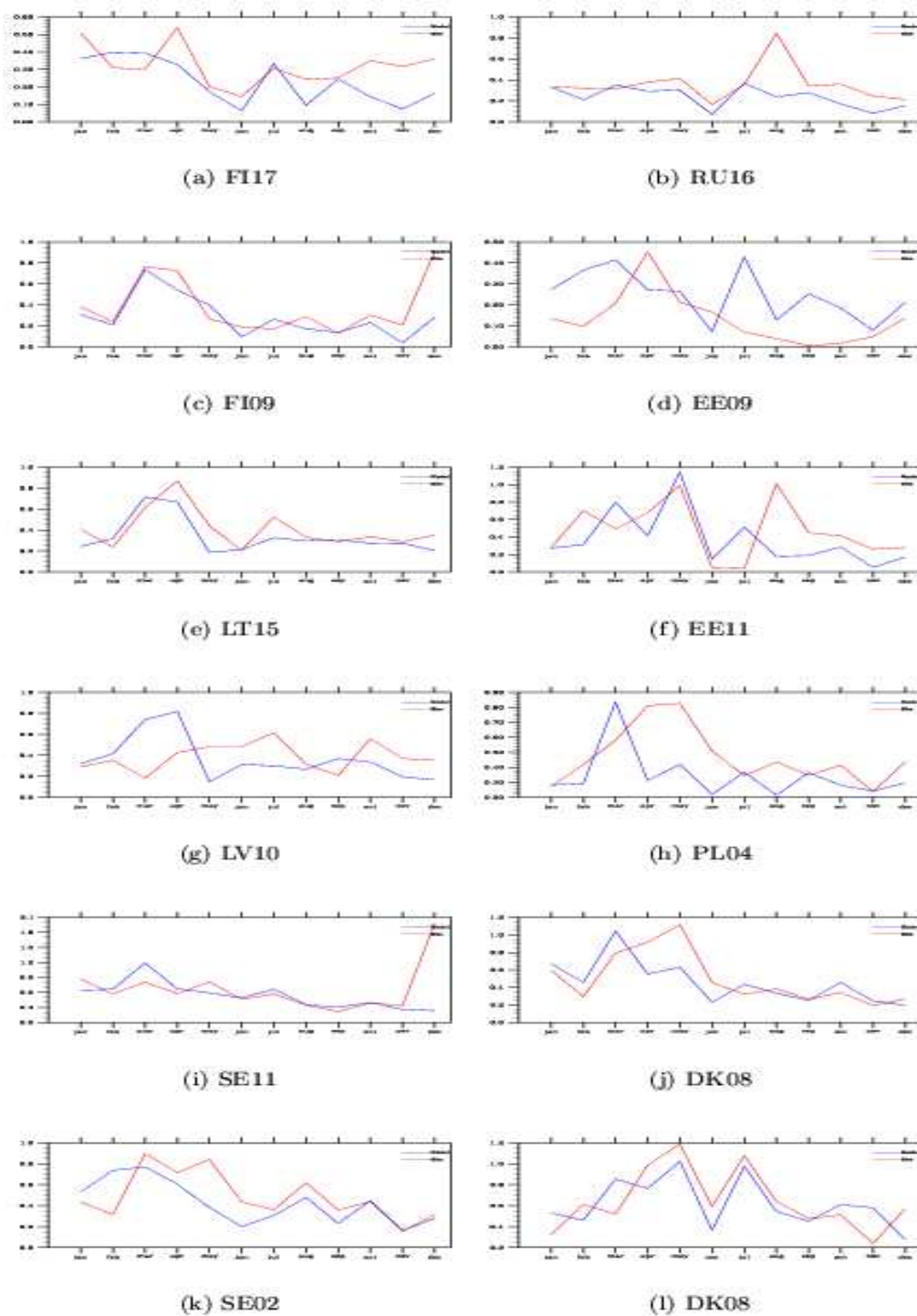


Figure 4.25. Monthly time-series for modelled versus measured concentrations of ammonium nitrate in precipitation in 2001. Units: $\mu\text{g N l}^{-1}$.

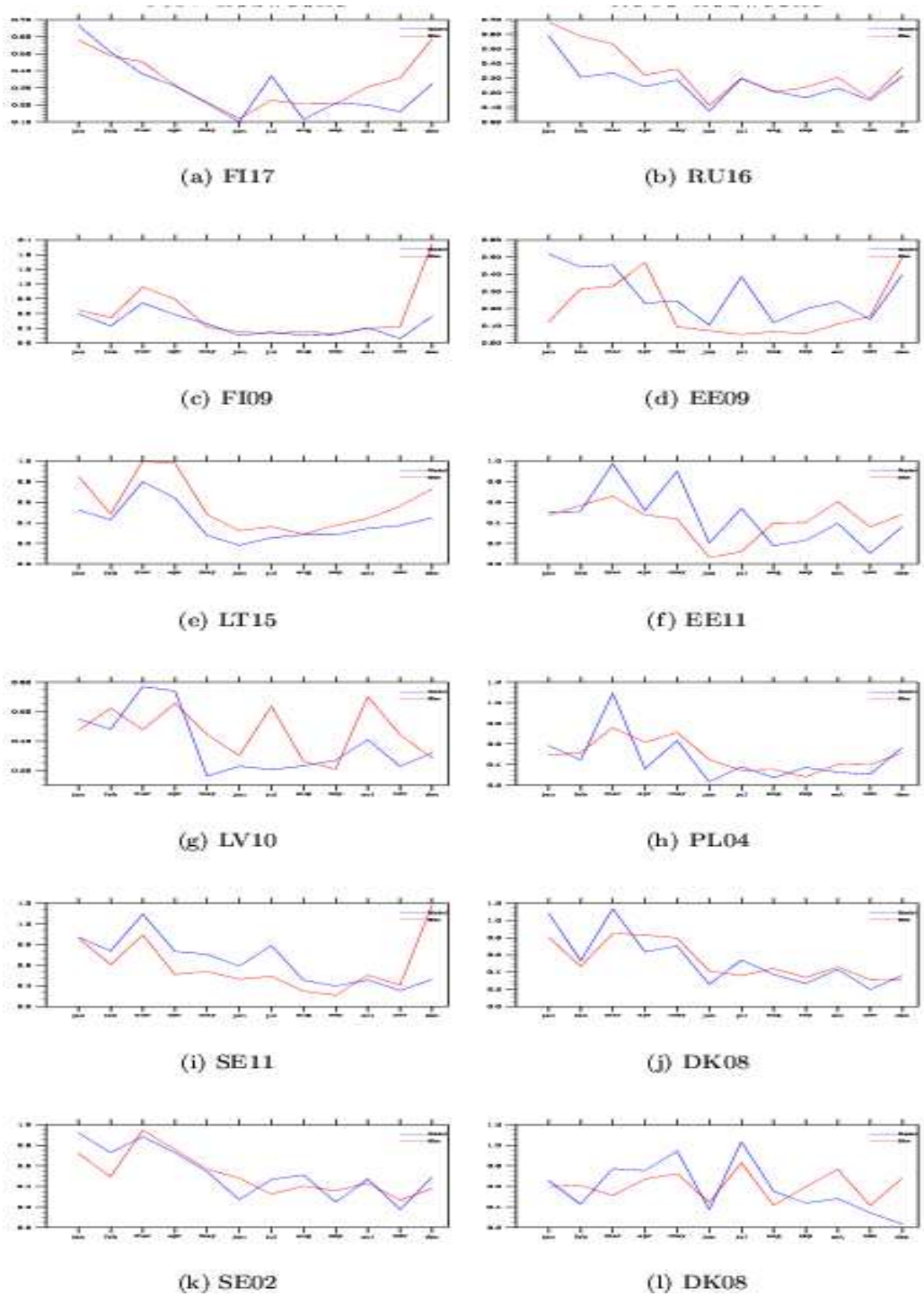


Figure 4.26. Monthly time-series for modelled versus measured concentrations of oxidized nitrogen in precipitation in 2001. Units: $\mu\text{g N m}^{-3}$.