

1. Executive Summary

Fourth Joint Report. This is the fourth joint report of the EMEP Centres for HELCOM, which concentrates on the results for one particular year - 1998. In contrast to the previous reports (Tarrason *et al.* 1997, Bartnicki *et al.* 1998 and Bartnicki *et al.* 2000) it will be only available on the internet, at the beginning on the EMEP WEB pages, and finally on the HELCOM WEB pages. In this way we could follow the suggestions of the First MONAS Meeting, which was held 8-12 May 2000 in Tallin Estonia. As required by HELCOM, the present joint annual report includes descriptions of the following tasks performed by the centres:

- Updated emission estimates of selected pollutants (two years in arrears). The selected pollutants are: oxidized and reduced nitrogen, cadmium, lead, mercury and lindane.
- Estimates of the annual deposition for the above mentioned pollutants, including separation to wet, dry and total deposition values over sub-basins and catchments of the Baltic sea.
- Deposition allocation budgets for sub-basins and catchments on the country basis.
- Analysis of measurements of observed pollution levels and comparison of the modelling results with measured concentrations at HELCOM stations.

The most important and most interesting, from the policy point of view, conclusions are presented below.

Warning about quality of 1997 nitrogen results. Computed depositions of nitrogen in 1997 are presented in several places of this report, in comparison with the 1998 results. It should be mentioned here that both, the computed 1997 depositions and the 1997 source allocation budgets are not entirely correct. The reason for that is the lack of convective precipitation in the 1997 meteorological data used for nitrogen computations. The underestimation of annual 1997 precipitation lead to an underestimation of computed 1997 deposition of nitrogen compounds. This problem was in more detail discussed on EMEP WEB pages and in the EMEP report (Tarrason and Schaug, 2000).

Emissions. The relative changes of annual total emissions of all compounds between 1997 and 1998, from all HELCOM countries and from the entire EMEP area are shown in Figure 1.1.

Annual total emissions from all HELCOM countries are, for all compounds, lower in 1998 than in 1997. Annual emissions of oxidized and reduced nitrogen from the EMEP area are higher in 1998 than in 1997, whereas, emissions of heavy metals and lindane are lower.

For all compounds there are only small differences between spatial distributions of emissions in 1997 and in 1998. As an example, we present in Figure 1.2 maps of 1997 emissions and 1998 emissions of oxidized nitrogen.

Emissions of oxidized and reduced nitrogen used in the model computations for 1998 were officially reported by EMEP countries to the Steering Body of EMEP. For oxidized nitrogen the so called "Baltic Sea" emissions resulting from international ship traffic are relatively high, e.g. higher than annual total emissions from Denmark. The Baltic Sea emissions remain on the same level in 1997 and in 1998.

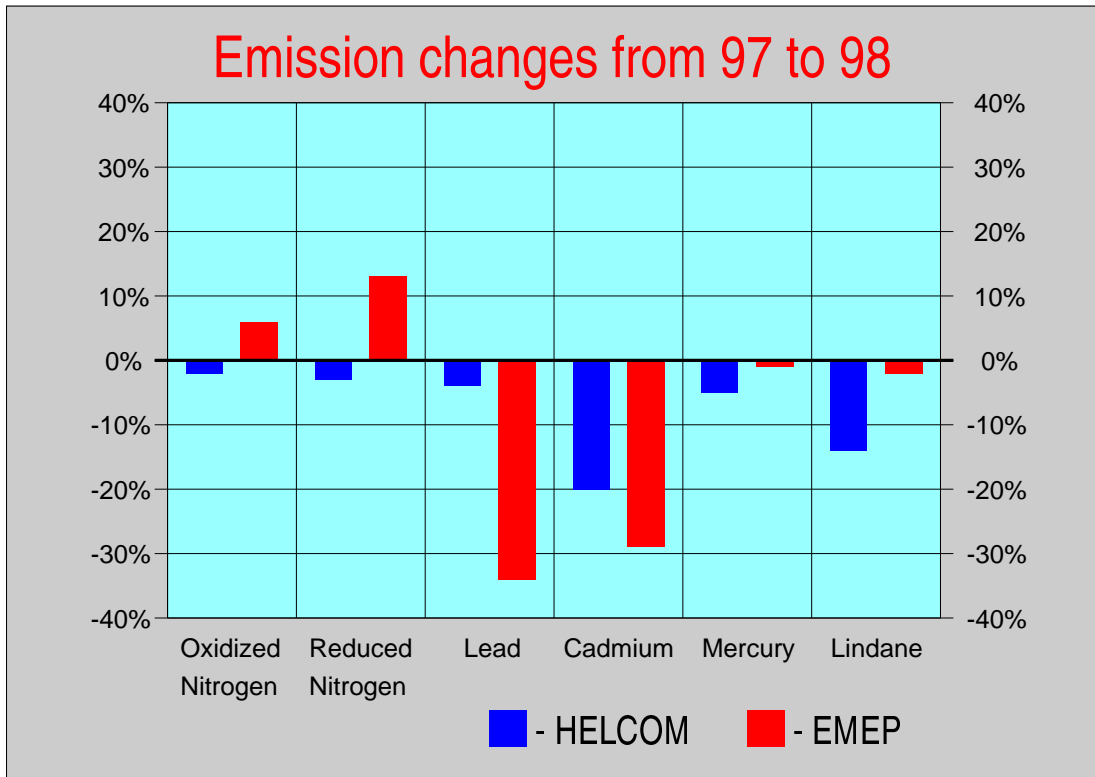


Figure 1.1. Relative changes of annual total emissions of all compounds between 1997 and 1998, from all HELCOM countries and from the entire EMEP area, in % of the 1997 value.

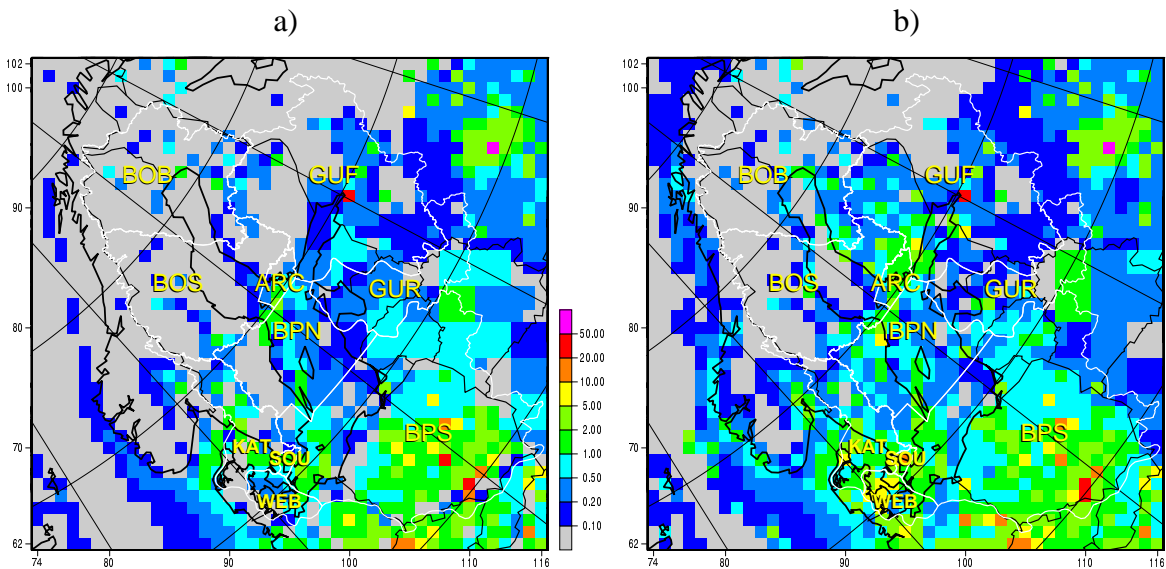


Figure 1.2. Maps of: a) 1997, and b) 1998 annual emissions of oxidized nitrogen in the Baltic Sea region. Units: ktonnes grid⁻¹ yr⁻¹.

In the 1998 computations of lead and cadmium depositions, both anthropogenic and natural sources were included. Natural lead and cadmium emissions in the EMEP area are relatively low, and they account for 5% and 12% of the total emissions, respectively. The official lead and cadmium anthropogenic emission data submitted by countries to EMEP were used in the 1998 computations.

In all of Europe, approximately 50% of mercury emissions are released to the atmosphere in the form of elemental mercury, 30% as inorganic oxidized mercury and 20% as particulate mercury. Natural emissions and re-emissions of mercury account for 50% and 10%, respectively, of the total 1998 annual emissions of mercury in the EMEP area. Both official emissions and expert estimates were used in the mercury computations for 1998.

In the case of lindane, expert estimates of 1996 emissions are used for most of the countries in 1998 model computations. However, where available, the official data from EMEP countries were used for emissions, in total in 11 out of 42 countries. Among HELCOM countries, official 1998 data were only available for Denmark and Sweden (zero 1998 annual emissions in both countries).

Atmospheric load of pollution to the Baltic Sea. Relative changes of annual dry, wet and total (dry+wet) depositions to the Baltic Sea are shown, for all compounds, in Figure 1.3.

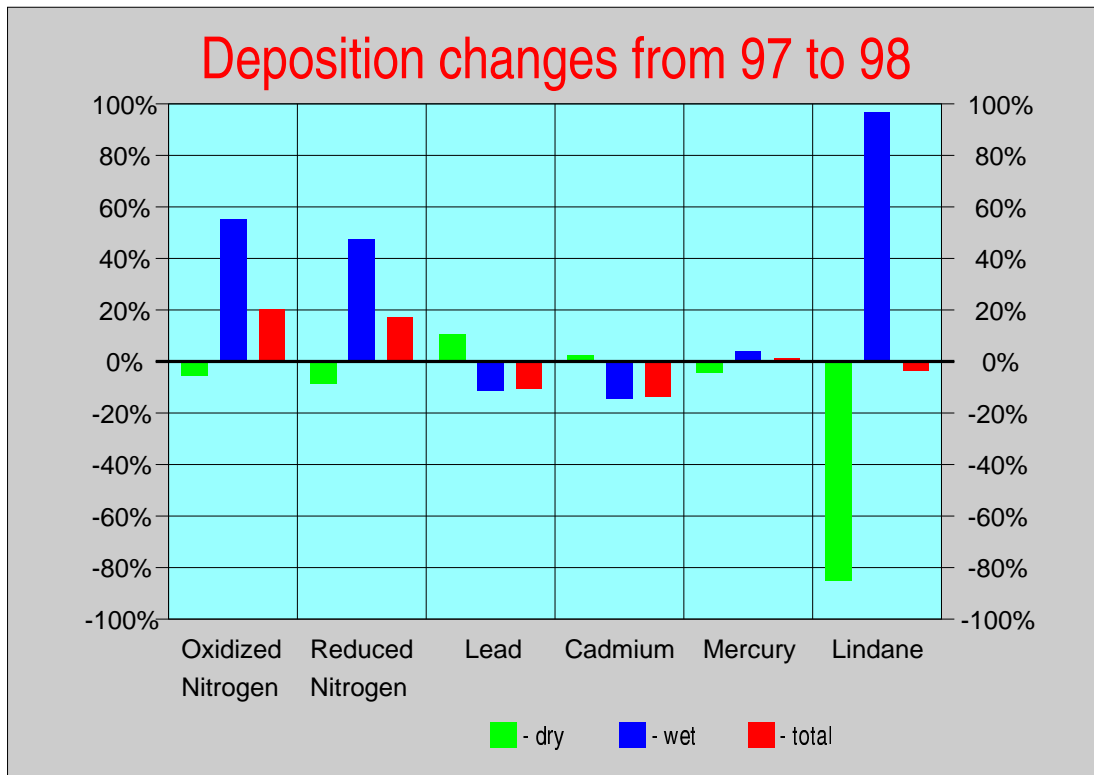


Figure 1.3. Relative changes of annual dry, wet and total (dry+wet) depositions of oxidized nitrogen, reduced nitrogen, lead, cadmium, mercury and lindane to the Baltic Sea basin. Units: % of the 1997 deposition values.

Annual dry depositions of oxidized and reduced nitrogen into the Baltic Sea region are slightly lower in 1998 than in 1997, wet and total depositions of both compounds are significantly higher in 1998 than in 1997, despite lower emissions from the HELCOM countries in 1998 than in 1997. There are three reasons for the increase of total nitrogen deposition in 1998: 1) higher amount of precipitation in 1998 compared to 1997 (higher wet deposition), 2) different other meteorological conditions (e.g. pattern of mixing height, cloudiness and surface roughness) for these two years, and 3) higher 1998 nitrogen emissions outside the HELCOM countries. The largest deposition flux of total (oxidized+reduced) nitrogen, $1584 \text{ mg N m}^{-2} \text{ yr}^{-1}$, was computed for the Western Baltic and Belts (WEB) sub-basin.

Dry depositions of lead and cadmium to the Baltic Sea basin were slightly higher in 1998 than in 1997, whereas, wet and total depositions were lower in 1998 than in 1997. The maxima of lead and cadmium deposition fluxes in 1998, $516 \text{ g km}^{-2} \text{ yr}^{-1}$ and $21.1 \text{ g km}^{-2} \text{ yr}^{-1}$, respectively, were both computed for the Southern Baltic Proper (BPS) sub-basin.

Dry, wet and total depositions of mercury to the Baltic Sea basin remain approximately at the same level in 1997 and 1998. Also total deposition of lindane remains at the same level in 1997 and 1998, but there are large differences in dry and wet depositions of lindane for those years. The maxima of the deposition fluxes for mercury and lindane, $35.4 \text{ g km}^{-2} \text{ yr}^{-1}$ and $37.6 \text{ g km}^{-2} \text{ yr}^{-1}$, respectively, can be noticed in the Sound (SOU) sub-basin of the Baltic Sea.

Concerning spatial distribution of the annual 1998 deposition, there is a clear south-to-north gradient in the deposition for all compounds. As an example the deposition maps for reduced nitrogen and lead is shown in Figure 4. Similar pattern can also be found for oxidized nitrogen, cadmium, mercury and lindane in the maps presented in Chapter 4. The south-to-north gradient in the deposition is mainly forced by a similar pattern in the spatial distribution of emissions in 1998.

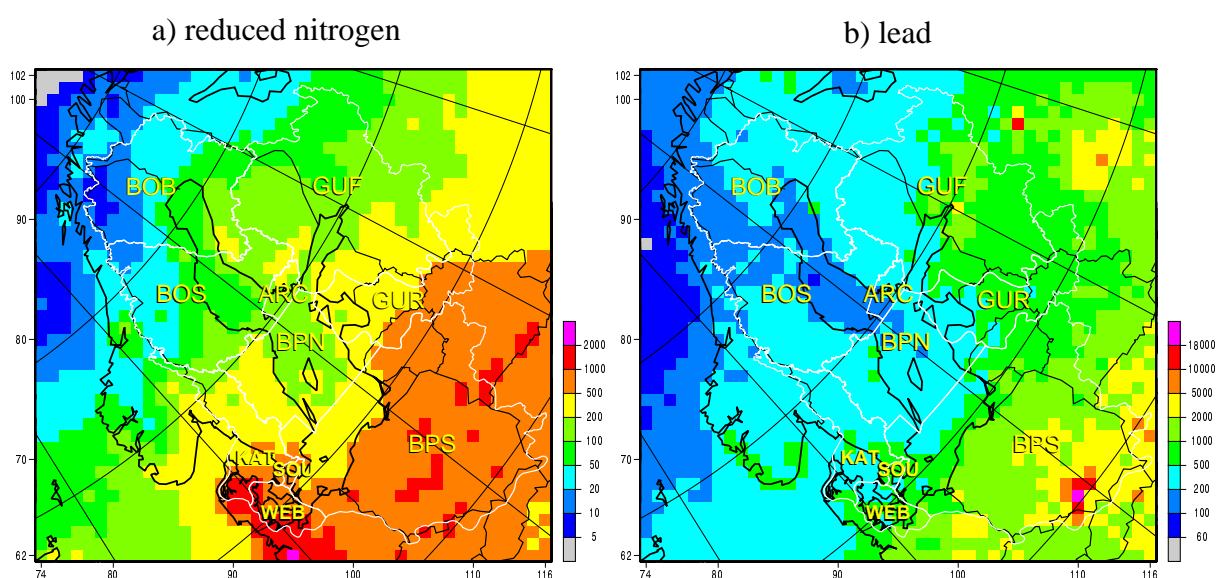


Figure 1.4. Maps of annual 1998 depositions of a) reduced nitrogen and b) lead in the Baltic Sea region. Units for reduced nitrogen - $\text{mg of N m}^{-2} \text{ yr}^{-1}$, and for lead - $\text{kg km}^{-2} \text{ yr}^{-1}$. A clear south-to-north gradient is visible in both deposition maps.

Source allocation budgets for the Baltic Sea. Comparisons of two major contributors to the 1997 and 1998 depositions to the Baltic Sea basin and to the Baltic Sea catchment area are shown in Figures 1.5 and 1.6, respectively.

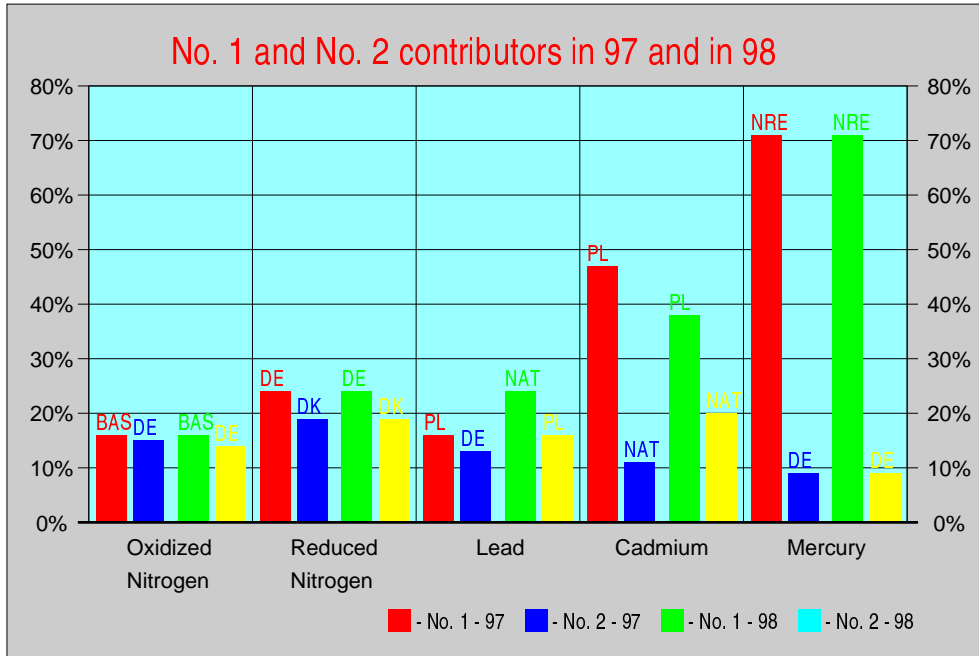


Figure 1.5. Comparison of the two major contributors to the depositions to the Baltic Sea basin in 1997 and in 1998. Units: % of the total depositions to the Baltic Sea Basin. The NRE means depositions caused by the natural emissions and re-emissions of mercury.

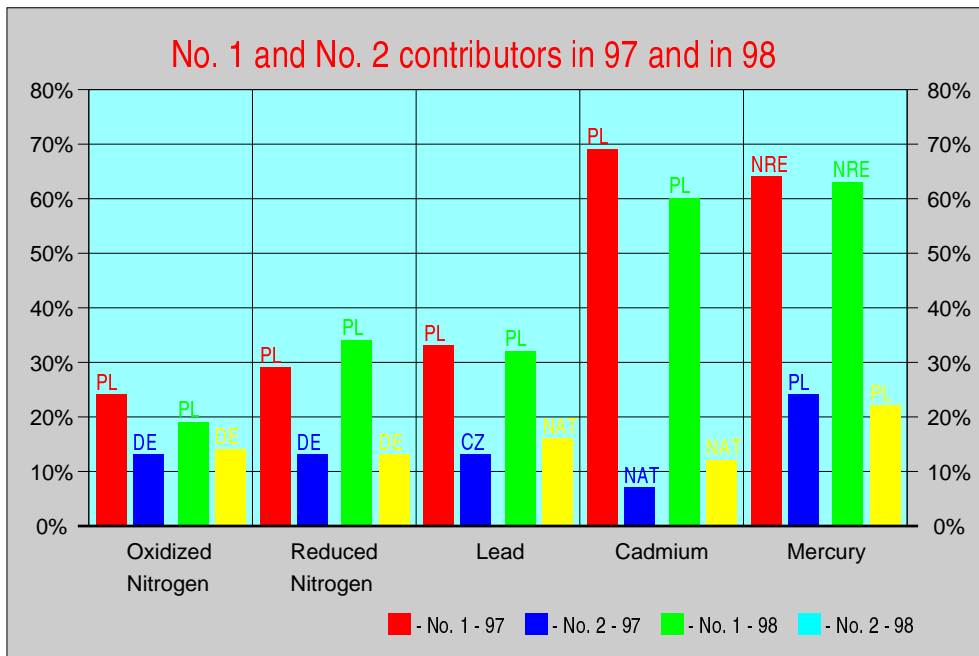


Figure 1.6. Comparison of the two major contributors to the depositions to the Baltic Sea catchment in 1997 and in 1998. Units: % of the total depositions to the Baltic Sea Basin. The NRE means deposition caused by the natural emissions and re-emissions of mercury.

The two major contributors to the depositions of nitrogen compounds in the Baltic Sea in 1997 and 1998 are the same: Poland and Germany, both for its basin and catchment. Also the values of their contributions are similar for those two years.

For lead and cadmium contributions to the depositions in the Baltic Sea in 1997 and 1998, natural emissions became more important in 1998 as compared to 1997, both for its basin and catchment area.

In the case of mercury natural emissions and re-emissions are the major contributor to the depositions to the Baltic Sea basin and catchment area, both in 1997 and in 1998.

Observed pollutant concentrations at HELCOM stations. At the HELCOM stations no significant increase or decrease in nitrogen oxide (NO_2) concentrations in air was observed during the period 1994 - 1998. The concentrations tend to be higher during winter and autumn months. The annual cycles of total oxidized nitrogen ($\text{HNO}_3 + \text{NO}_3\text{-N}$) cycles are generally not strong, but tend towards a winter maxima. The same can be stated for the total reduced nitrogen ($\text{NH}_3 + \text{NH}_4^+\text{-N}$), although concentrations may also be high during spring and autumn months.

At several HELCOM sites a decrease in weighted annual nitrate (NO_3) and ammonium (NH_4) concentrations in precipitation can be observed for the period 1994 to 1998. This is especially true in the southern part of the Baltic Sea showing relatively high concentration levels. Weighted annual pH values increased at most stations, likely related to the decreasing sulphate (SO_4) concentrations in precipitation.

Monthly NO_3 and NH_4 concentrations in precipitation are lower in the northern part of the Baltic Sea. The highest peak values in 1998 were observed at the German station Zingst and the Finnish station Utö during the winter months. Highest peak NH_4 concentrations in 1998 were measured at Zingst (Germany), Keldsnor (Denmark) and Vavihil (Sweden).

Annual mean concentrations of heavy metals in the air did not show clear changes between 1994 and 1998. In 1998 the highest Pb values were found at Zingst (Germany), Keldsnor (Denmark) and Preila (Lithuania).

Weighted annual average heavy metal concentrations in precipitation decreased over the period: 1994 - 1997. However, at several stations the values increased again in 1998. Differences in annual heavy metal concentrations at the single HELCOM stations are mainly due to differences in monthly peak values observed or calculated over the period: 1994 - 1998.

In 1998, mean monthly lead concentrations in precipitation showed highest values at the stations Rucava and Preila. Monthly cadmium concentrations in 1998 were usually below $0.4 \mu\text{g l}^{-1}$ at all stations. The lowest values were measured in August and September 1998. A high peak concentration was observed at the Latvian site Rucava in April 1998.

The monthly volume weighted mercury (Hg) concentration at the German station Zingst was in 1996 much higher compared to the concentration given for 1998 (1994: 10.79 ng l^{-1} , 1995: 16.49 ng l^{-1} , 1996: 18.47 ng l^{-1} and 1997: 13.13 ng l^{-1} , 1998: 13.14 ng l^{-1}). Annual Hg values (1998) at the Swedish stations Rörvik and Vavihil were lower, 7.0 and 8.3 ng l^{-1} , respectively.

Concentrations of lindane in precipitation have only been reported for the German station Zingst, and there are no major changes in 1998 as compared to 1997.

Comparison of measurements and model results. Comparison of measured and modelled values of NO₂, precipitation, wet deposition of oxidized nitrogen, and wet deposition of reduced nitrogen, for all available HELCOM stations in 1998 is shown on the scatter diagrams in Figure 1.7.

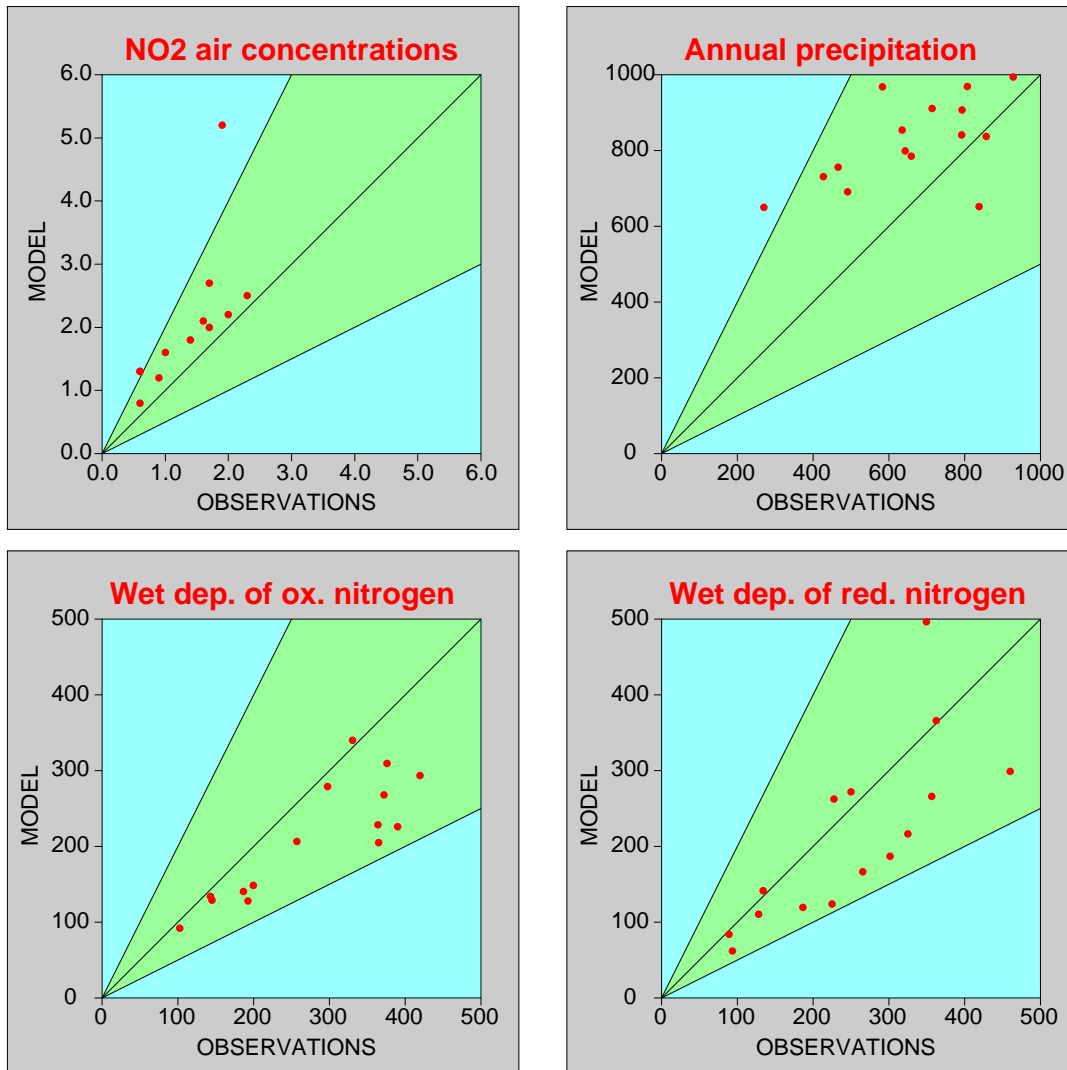


Figure 1.7. Comparison of measured and modelled values of: 1998 annual average NO₂ air concentrations (in $\mu\text{g m}^{-3}$), 1998 annual precipitation (in mm yr^{-1}), 1998 wet deposition of oxidized nitrogen (in mg N m^{-2}), 1998 wet deposition of reduced nitrogen (in mg N m^{-2}).

In case of nitrogen, there is a better agreement between measured and modelled values of annual average NO₂ concentrations, annual depositions of oxidized nitrogen and annual depositions of reduced nitrogen in 1998 than in 1997. Although annual average NO₂ concentration measured

at Preila (LT51R) station seems to be extremely high ($12.48 \mu\text{g m}^{-3}$), measured average with and without Preila station are closer to computed value in 1998 than in 1997. Concerning precipitation amount, in 1998 model results overestimated the annual mean averaged over all HELCOM stations by 23%, whereas, in 1997 model underestimated the measured mean by 14%. Annual average measured and computed wet depositions of oxidized and reduced nitrogen are also in better agreement in 1998 (model underestimation of 25% and 18%, respectively) than in 1997 (model underestimation of 42% and 41%, respectively). Both modelled and measured annual wet depositions of oxidized and reduced nitrogen are higher in 1998 than in 1997.

An application of the official EMEP data in the emission inventories of lead and cadmium leads to an underestimation of the measured concentrations by the model results, both in 1997 and in 1998 computations. Scatter diagrams with comparison of measured and modelled 1998 air concentrations of lead and cadmium at available HELCOM stations are shown in Figure 1.8.

Due to lack of measurements for lindane at HELCOM stations in 1998, model results for this compound were compared with available measurements at EMEP stations. Scatter diagrams with comparison of measured and modelled 1998 concentrations in precipitation of mercury (HELCOM stations) and lindane (EMEP stations) are shown in Figure 1.9. In both cases the model overestimates measured values.

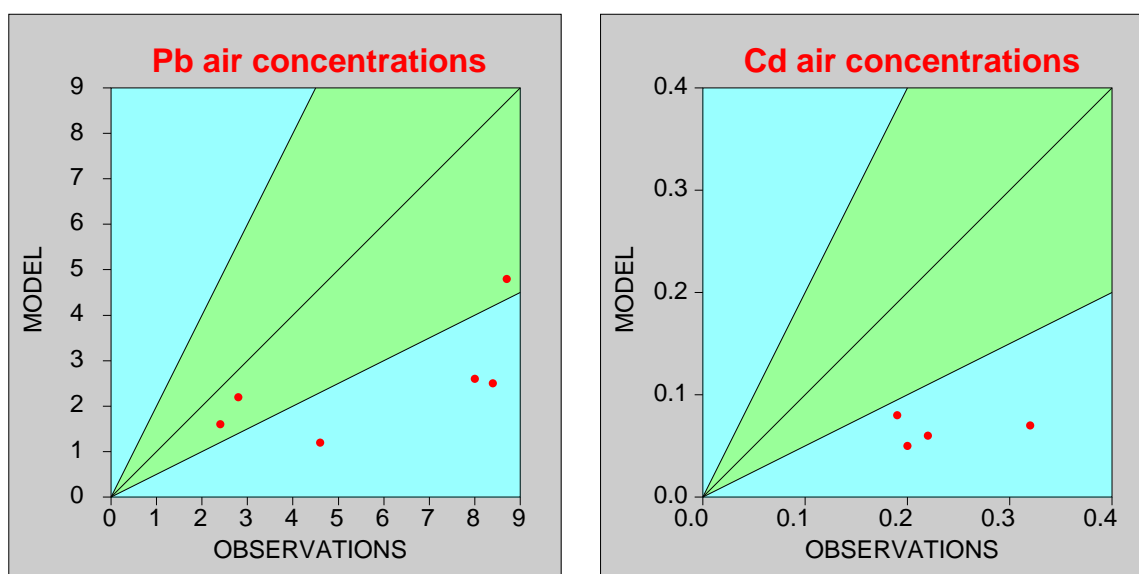


Figure 1.8. Comparison of measured and modelled values of 1998 annual average Pb and Cd air concentrations. Units: ng m^{-3} .

Results of the comparison for heavy metals and lindane showed that computed concentrations of lead and cadmium are lower than measured ones by a factor of 2-4. This can be possibly caused by either an underestimation of emission data, or inevitable model simplification of atmospheric processes, or by the problems connected with the complexity of heavy metal measurements. Additional analysis of the results indicated that the underestimation of computed concentrations for lead and cadmium is most likely connected with the uncertainties of emission data used for modelling. Both official anthropogenic emission data and estimations of natural emissions of these compounds have sufficient uncertainties. For instance, comparing official emission data for 1990 and for 1998 it should be noted that the latter emissions are significantly lower.

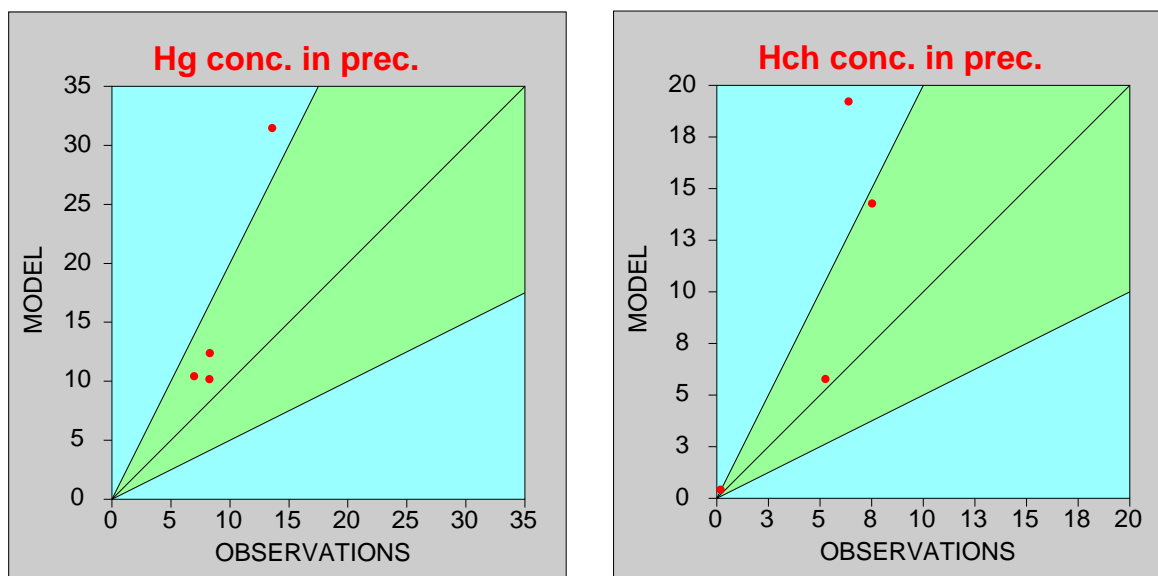


Figure 1.9. Comparison of measured and modelled values of 1998 annual average Hg and lindane concentrations in precipitation. Units: ng l^{-1} .

While emission data in the entire HELCOM region has fallen 8 times for lead and 2 times for cadmium, emission decrease on the level of individual countries is even greater. For some countries the ratio of the 1990 to 1998 lead emissions exceed one order of magnitude (Denmark, Finland, Sweden). It is natural that such a decrease of emission estimates should ultimately result in a serious reduction of the computed concentrations of lead and cadmium. In this context, it is important to mention that measured concentrations of these pollutants carried out at the monitoring stations around the Baltic Sea (Berg et al., 1996; Berg et al., 1997; Berg and Hjellbrekke, 1998) do not quite indicate this sharp decrease of concentrations from 1990 to 1998. Recently within the EMEP the process of the refinement and submission of emission data for heavy metals was noticeably intensified. Therefore there is a hope that this progress will increase the accuracy of modelling results and improve the agreement between the computed and observed concentrations of lead and cadmium.

The other reason of disagreement between measured and computed concentrations of lead and cadmium can be connected with inevitable model simplification of atmospheric processes and with a possible underestimation of input of natural sources of lead and cadmium. The uncertainties connected with modelling approaches used at MSC-E were extensively analysed. MSC-E carried out two model intercomparison studies for lead and cadmium in co-operation with experts from several scientific organisations of Germany, Sweden, the Netherlands, and Norway. Several models along with MSCE-HM model took part in these studies carried out on the basis of the data for 1990 (Sofiev et al., 1996; Gusev et al., 1999). The results showed that participated models were able to simulate concentrations and deposition fluxes on an annual basis with an uncertainty of a factor 2. Furthermore MSC-E performed an assessment of model uncertainty and sensitivity study (Ilyin et al., 2000). The analysis has shown that the uncertainties of modelling results due to the modelling approaches are within the range of 30-50%.

Therefore, taking into account the results of this analysis, the most likely reason of the underestimation of computed concentrations of lead and cadmium can be connected with the under-

estimated emission data. It means that actual atmospheric load of these compounds to the Baltic Sea is most likely higher than estimated values of depositions.

Results of the comparison for mercury showed that the computed concentrations are in reasonable agreement with the measurements. The differences are within a range of a factor 1-2. At present MSC-E is carrying out a similar model intercomparison study for mercury models in cooperation with experts from Germany, Sweden, Denmark, Norway, USA, and Canada. It is believed that the results of this study will contribute to a further development of the EMEP/MSCE model for mercury.

The comparison of modelling results for lindane with available measurements from EMEP monitoring sites showed that computed concentrations exceed measured ones by a factor of 1-3. This can be caused by several reasons, in particular, the overestimation of lindane emissions, uncertainties in modelling approaches, and uncertainties of the physical-chemical properties of lindane used in the MSCE-POP model.

More details concerning all major points discussed in this summary can be found in different Chapters of this report.