Atmos. Chem. Phys. Discuss., 11, C2490–C2499, 2011
www.atmos-chem-phys-discuss.net/11/C2490/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric deposition of nitrogen to the Baltic Sea in the period 1995–2006" *by* J. Bartnicki et al.

J. Bartnicki et al.

jerzy.bartnicki@met.no

Received and published: 29 April 2011

We would like to thank the Anonymous Referee1 for thorough revision of our manuscript, as well as important and inspiring comments and corrections. We quote each comment first and then reply to it.

General comment:

While there are no major faults with the basic science and the conclusions are straightforward, I am a little concerned that no attention is paid to the uncertainties in the model results, and no validation (e.g. monitoring) data are shown. In the introduction, mention is made of modelled deposition estimates by 2 other models: MATCH with an estimated deposition to the Baltic of 261-300 GgN/yr for the period 1992-2001 and

ACDEP with an estimate of 318 GgN/yr for 1999. Even taking into account the alleged reduction in N emissions and deposition after 2000, as well as the time shift between the periods considered in the different modelling studies, the deposition estimates provided by the EMEP model (200-230GgN/yr) are significantly (25%) lower than those of both MATCH and ACDEP. The discussion ought to address this issue and attempt to explain the differences. The EMEP model results should be put in the context of other studies, whether model or monitoring based, in order to assess the validity of the output. For example, if there was a 20% reduction in wet deposition for the period 2002-2006 compared with 1995-2001, as suggested clearly by Figure 6, then the signal should be visible from monitoring network data for wet deposition around the Baltic Sea. Are there any such data (the introduction suggests there are), and if so, do they confirm the reduction/trend in wet deposition over the years? by the same margin?

Reply:

This important point is mainly related to model verification issue and it was raised by both Referees. The EMEP model is systematically verified every year on the latest available measurement data and this was the reason why we slightly limited the scope of this subject in our manuscript. In addition, there has been a study (Fagerli and Aas, 2008) focused on comparison of modelled and measured nitrogen in air and precipitation in the long term period 1980-2003. Altogether 33 EMEP sites located in different parts of Europe were used in this study which indicated a decline of ammonium and nitrate in precipitation both in measurements and model results. However, out of 33, only 5 sites reported concentration in precipitation in the locations close to the Baltic Sea basin. Therefore, we decided to expand the subject of model verification in the revised manuscript and include the results of additional research. Thanks to Wenche Aas from the Norwegian Institute of Air Research, we have received the data with measured nitrogen concentrations in precipitation at so called HELCOM stations, which are in fact the EMEP stations reporting in slightly different way for the HELCOM purpose. We have examined 21 HELCOM sites, of which 11 reported annual nitrogen concentrations in precipitation and precipitation amount for the entire period 1995-2006. We have used the 11 stations with data available over the entire period for comparing observed annual wet deposition with the modelled wet deposition. In addition, annual precipitation amount to the Baltic Sea basin is also compared as average from measurements over 11 stations and average over all model grids covering Baltic Sea basin. This comparison is shown in Fig. 1. For all stations there is a relatively good agreement between average observed and modelled deposition values over the entire period, but the correlation is not so good, mainly because of differences in modelled and observed precipitation amount for individual stations. This good agreement indicates that the most likely, additional reason for the differences in nitrogen deposition to the Baltic Sea basin calculated by the EMEP model and by the MATCH and ACDP models is related to different emission inventories used for the calculations. Both nitrogen and sulphur emissions were slightly lower in the EMEP calculations after latest revisions in 2010. However, different meteorology still remains an important factor for the differences. The inter-annual variation of measured annual nitrogen deposition is similar, but larger than the inter-annual variation of modelled deposition. However, correlation between annual measured and modelled values is not very good, typically around 0.3 and this is a follow up of a bad correlation between measured and modelled annual precipitation. The modelled precipitation comes from the numerical weather prediction model and represents the entire grid square, whereas the realistic variation of precipitation within one grid square is high. In some cases it can be higher than 100% (e.g. model grid square which includes Bergen in Norway). When the model wet deposition is calculated with precipitation from the station, the correlation between measured and modelled deposition is much better, above 0.8. The examination of data behind Fig. 6 show that the reduction in modelled wet deposition in the period 2001-2006 compared to period 1995-2000 is 13%. Similar reduction is present in the measured deposition for the stations in Fig. 1. Except for two stations DK05 Keldsnor and PL04 Leba, there is a clear reduction for measured wet deposition in the period 2001-2006 - on average 10%. This value is quite similar and only slightly lower than predicted by the model

calculations. The above discussion and Fig. 1 are included in the revised manuscript.

Specific comment:

1- In the section on temporal changes in N emissions, especially p1808, l10-19, emissions are described as having been 'reduced during the considered period by 5% and 18% for NOx and NH3, respectively'. The statement makes it sound as though there had been a continuous and steady decline throughout the period, but in fact, for NOx, emissions dropped from 1995 only until 2000-2001, but then clearly started to increase again until 2005, just as steadily as they had decreased from 1995 to 2000. This should be pointed out in the text; it seems on the basis of these data that total N emissions in the region stabilised at the turn of the century. What do current data (since 2005) indicate for NOx? Are the reductions in NH3 emissions currently being outweighed by increases in NOx emissions on land and by shipping?

Reply:

We agree that the statement about emission reduction can be misleading. We propose a new formulation in the revised manuscript: Total HELCOM emissions (as a sum from all Contracting Parties) have been reduced during the considered period by 5% and 18% for nitrogen oxides and ammonia, respectively (Fig. 3). However, the reduction pattern was quite different. Nitrogen oxides emissions decreased almost linearly from 1995 to 2000 and then increased with some oscillations from 2002 to 2006. On the other hand, a steady decrease in ammonia emissions can be observed for the entire period. The ship emissions have increased by 30% in the same period in the pattern very close to linear. After 2006, in the years 2007 and 2008, both nitrogen oxides and ammonia emissions, as a sum from all HELCOM Contracting Parties and ship traffic, remain on the same level as in 2006, within 1% accuracy (CEIP, 2010).

Specific comment:

2- Related to the previous comment, the reduction in modelled total deposition is some-

what presented (p1809, l19-25) as a more or less continuous trend with strong interannual variability ("The level of annual total nitrogen deposition into the Baltic Sea basin has changed from 230Gg in 1995 to 199Gg in 2006"). But what is actually striking in Figure 5 is the sudden (and permanent) drop in 2001 in wet deposition (both oxidised and reduced), with annual wet deposition levels in the years before 2001 almost all (6/7) in the range 70-90 GgN, and all post-2001 numbers in the range 60-70 GgN. Total nitrogen deposition does not decrease by 13% during the entire period (cf p1809, l23-24), but only during the first half of that interval, and then stabilises, with the strong reduction in precipitation around the year 2001 playing a dominant role. Also, it would be useful to know if the reported variations in precipitation of Fig.6 (presumably modelled data from NWP model output? please say) were confirmed by ground observations around the Baltic Sea. The text makes the policy-relevant comment that the possibility of increased deposition of nitrogen after an emission reduction has taken place (p1810, l23) should be borne in mind, owing to interannual meteorological variability, but presumably the need to consider longer-term datasets is already a well established wisdom among policy makers.

Reply:

Total deposition pattern shows (Fig. 7) large inter-annual variability in the period 1996-2002 and sudden drop (28%) between the years 2000 and 2002. This sudden drop is mostly caused by the drop in modelled precipitation from year 2001 to 2002. Precipitation data for the model calculations was provided by the numerical weather prediction model PARLAM. Similar drop (18%), but from year 2001 to 2003 is also visible in precipitation in Fig. 1 where the observed annual precipitation averaged over 11 HELCOM stations is shown. The inter-annual variation in modelled and measured precipitation data shown in Fig 1 is similar.

Specific comment:

3- Monthly variability in wet deposition: it is argued (p1811, l14-16) than the month

to month variability is much higher for wet than for dry deposition because of large differences in monthly precipitation, but presumably this is also the result of variations in concentrations in air and in rain water. By contrast, for dry deposition, deposition rates are not necessarily correlated with high air concentrations, since conditions which favour high atmospheric concentrations (low wind speeds, suppressed turbulence) do not favour large deposition velocities.

Reply:

We agree with these comments and they are included in the revised version of the manuscript.

Specific comment:

4- p1812, l3-4: the modeling experiment gives more insight into the importance of meteorology in transporting pollutants, and also into their atmospheric chemistry and removal rates from the atmosphere

Reply: This sentence is included in the revised version of the manuscript.

Specific comment:

5- p1812, l29: the period of high variability is 1997-2002, not 1996-2003. In Fig.11, it would be useful to show both dry and wet deposition, rather than just total, to show whether varying meteorology affects dry and wet deposition in a similar fashion. It would also be useful, in Figure 9, to show the min, max and mean monthly deposition for both dry and wet deposition, as well as total, to show whether dry and wet deposition both peak at the same time of year. Rainfall and windspeed tend to be correlated on a seasonal basis, but, while rainfall scavenges N compounds from the gas phase and thus reduces the fraction that is available for dry deposition to the sea, high wind speeds increase the roughness of the sea surface and enhances the efficiency of dry deposition (higher Vd). Please comment.

Reply:

Following Referees suggestion, Fig. 11 has been changed so that both dry and wet deposition is shown in addition to total deposition (Fig. 2). There is more variability in wet deposition, with the range 93-121% of average than in dry deposition with the range 92-110% of average. However minimum and maximum of both wet and dry deposition occurs in the same years, 1997 and 2000, respectively, resulting in clear minimum and maximum of the total deposition in the same years. Also Fig. 9 has been changed, so that monthly minimum, maximum and average are shown not only for total, but for dry and wet deposition as well (Fig. 3). Unfortunately, we have not been able to examine the surface wind speed over the Baltic Sea, but we agree with the Referee that it is correlated with precipitation on seasonal basis. It is visible in new Fig. 9 (Fig. 3) for the autumn months October and November when the strong storms on the Baltic Sea are most frequent and surface wind speed reaches the seasonal maximum. For these two months, the highest values are visible in monthly dry, wet and total deposition. The modified Figures 9 and 11, and above remarks are included in the revised manuscript.

Technical corrections All suggested technical corrections are included in the revised version of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 1803, 2011.

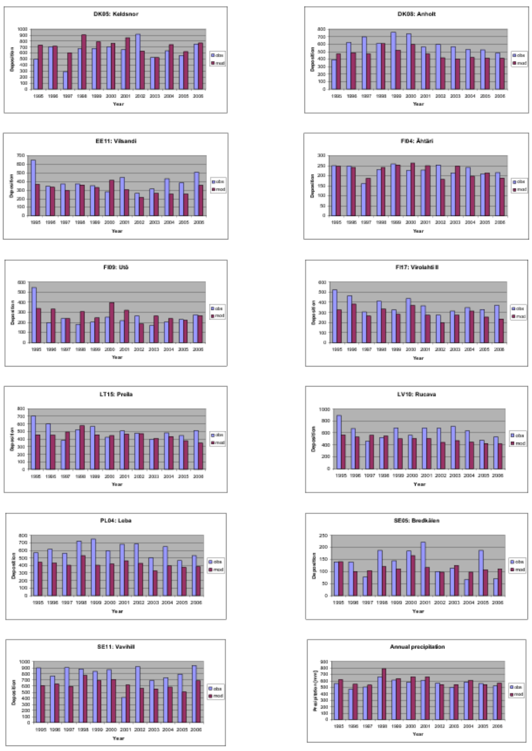

Fig. 1. Comparison of annual wet deposition based on observations (obs) at HELCOM stations and calculated by the EMEP model (mod). Units: mg N/m2. Observed and modelled precipitation.

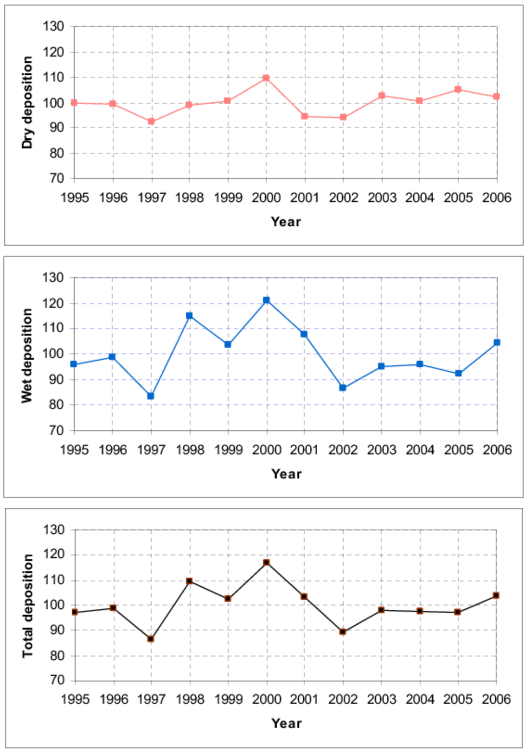

Fig. 2. Annual deposition of dry, wet and total nitrogen to the Baltic Sea basin in % of the average value over the 1995-2006 period. Constant 2006 emissions and variable meteorology.

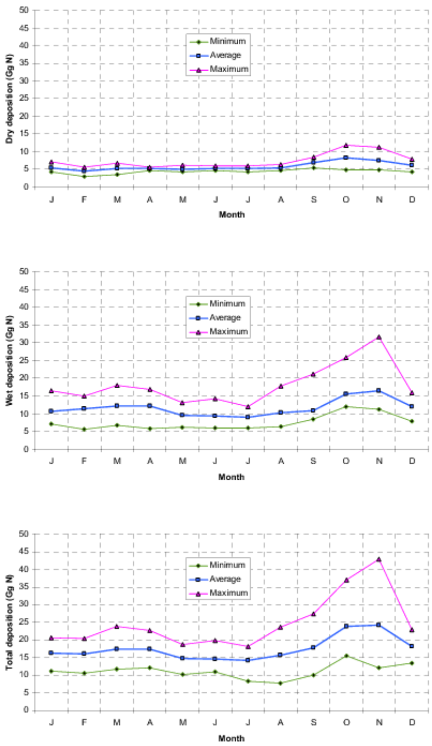

Fig. 3. Monthly minimum, maximum and monthly average (over 12 years) deposition of dry, wet and total nitrogen to the Baltic Sea basin in the period 1995 - 2006.