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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.

Anonymous Referee #1

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General comments

This paper presents model (EMEP) simulations of atmospheric nitrogen deposition to the Baltic Sea basin for a 12-yr period (1995-2006) over which N emissions (as reported by HELCOM contracting parties) are supposed to have declined by 11%. The main objectives of the paper are i) to assess the reduction in N deposition over the same period in relation to the reduction in emissions, ii) to study the controls of the interannual variability in N deposition and iii) to identify the main sources areas contributing to the total deposition. The paper is written clearly and structured logically and the overall reasoning is sound, starting with a description of the work undertaken jointly by HELCOM and EMEP and the specific objectives of this paper, followed by trends in reported N emissions, and then a presentation of model deposition results using first

actual emissions to derive actual trends in deposition, and then using fixed emissions to study the meteorological control and interannual variability. Source allocation of N deposition is discussed at the end. Some figures in the paper could be improved and harmonized to facilitate the reading of the paper (see comments below).

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 (eg 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 1992-2001and 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?

The manuscript may be published subject to adressing the following minor revisions.

Specific comments

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?

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

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.

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

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.

Technical corrections

p1804, l18: ...threat to THE unique and fragile...

p1804, l24: total nitrogen 'input' (rather than 'load')

p1805, l14: 'Altogether' thirteen joint EMEP... (instead of 'All together')

p1805, l16: change 'load' to 'input'

p1805, l16-17: remove comma between 'both' and 'measurements'

p1805, l27: ...318 GgN yr-1 for the basin area...

p1805, l28: change 'algae' to 'algal'

p1806, l15-16: ...to avoid these kinds of problems...

p1806, l19: ...in the centre of THE reduced model domain...

p1807, l13: ...different kindS of pollutants...

p1807, l20: ...50% of total N deposition... (rather than 'to the')

p1807, l23 and Table 1: for Germany, the oxidised N emission reported for 2006 is the same as in 1995 (2131.1 Gg); this is likely an error, as emissions decrease steadily down to 1443 Gg in 2005. Is this just an error in the table, or was this figure of 2131 Gg for 2006 actually used in the model runs?

p 1819, Table 1: the title is given as annual emissions of 'oxidised nitrogen', while I believe this table refers to the emissions of NOx (NO+NO2). Throughout the text, the term 'oxidised nitrogen' is also used for deposition but presumably refers not only to NOx but also to NO3-, HNO3, HONO and possibly other forms (eg organic nitrate?) of oxidised N. Table 1's title should be changed to NOx, as well as all references to table 1 in the text (in the emission context), while there should be elsewhere in the text a short statement describing what is meant by 'oxidised' N (which chemical species are included) in the deposition context. This is best done p1808, l24-25, at the start of the section on N deposition.

p1807, l29: Lithuania (not Latvia!) is the other country together with Finland where NH3 emission was higher in 2006 than in 1995 (see Table 2)

p1808, l4 and Figure 2: as it stands in the MS, Figure 2 does not have part a and part b, but 'left' and 'right' panels. Please change either text, or Figure 2 caption.

p1808, l19: remove question mark '?' at the end.

p1809, l11: Time series OF oxidised-dry...

p1826, Fig 5, p1830, Fig 8 and p1831, Fig 10: Please harmonise legends (Oxidised dry vs Dry oxidised, etc...), and please harmonise colour codes and symbols between the three figures to facilitate the visual comparison. In Fig 10 caption, the period is

1995-2006 (not 1997-2006)

p1810, l15: insert 'in HELCOM contracting parties' between 'total nitrogen emissions' and 'annual total nitrogen deposition'

p1810, l22-23: remove 'deposition' between 'increased deposition of nitrogen' and 'after the reduction of nitrogen emission'

p1811, l4: '...in THE case of annual deposition...

p1811, l6: '...oxidised wet AND monthly reduced...

p1812, l7-8: remove this section from the sentence: "i.e., using constant emissions of the year 2006 and meteorology of the corresponding years from 1995–2006", this is a straight repeat of what was said 5 line above.

p1813, l1: ...maximum OCCURRED in the year 2000...

p1813, l7-14: the standard deviation normalised to the mean is called the coefficient of variation; please use this terminology

p1814, l5-7: please rephrase: "Over the 1997–2006 period, the average contributions by Germany, the United Kingdom and Poland to oxidised nitrogen deposition into the Baltic Sea basin were 16%, 11% and 10%, respectively."

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