Biogeosciences Discuss., 9, C888–C900, 2012
www.biogeosciences-discuss.net/9/C888/2012/

Biogeosciences
Discussions

Interactive comment on "Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS" *by* C. Geels et al.

C. Geels et al.

cag@dmu.dk

Received and published: 1 May 2012

Response to review by Addo van Pul of the Biogeosciences Discuss paper: Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS" by C. Geels et al.

We appreciate the detailed review by Addo van Pul and the revised manuscript is clearly improved due to the suggested modifications and corrections. In the following we go through the comments by the reviewer point by point:

Reviewer: In General: The paper is clear and well written. The subject is not very new but still very relevant especially in the light of Natura2000. The main scope of

the paper is to show that more detail is needed in assessing the N-deposition. Again that as such is not so new. I'm more curious in how well the system works. The validation is only again concentrations in air. Why not use other components as well, i.e. wet deposition, ammonium aerosol. Besides how well does the model calculate the other N-components? Main concern I have is about uncertainties. There is hardly any information on uncertainty in modelling and more specific on how well the depositions are modelled. Particularly in comparing the depositions to critical loads this plays a crucial role. So can the authors give an estimate on that? This should be indicated in Figure 9 as well. Since the parametrization of the dry deposition is quite uncertain I specifically would have liked to see a discussion on that and specifically how the authors think their results will change comparing the current parameterization to the one with bi-directional fluxes included? I have indicated the paper to be reconsidered after major revisions. In fact this is too strong but there is no rating in between. The main revision I want to see and review is on the uncertainties.

Answer: First of all we appreciate that the reviewer finds the paper clear, well written and relevant. We agree that the subject is not completely new, but we believe that we through our research demonstrate that it is possible to design and apply a comprehensive coupled model system not only for research but also for environmental management. By including measurements of ammonia from a dense network covering different background conditions and ecosystem types we show that it is possible to improve the modelling of atmospheric ammonia in a landscape with a mix of agricultural and natural areas – an important step towards a better assessment of the total N load.

As the title of the paper implies, our aim has been to develop a system that can simulate the concentration of ammonia in a region like Denmark where the natural and semi-natural ecosystems are located as a patchwork in an agricultural landscape. As ammonia is the most heterogeneous of the N-components, this will be an important input to the estimation of the total N deposition. We therefore focus on the comparison with measured concentrations of NH3 and the local-scale model only includes NH3. In

previous work we have of course validated the regional model against measurements of wet depositions, ammonium etc. In a recent paper (Geels et al. 2012) we show a validation of a 20 year model run against measured values of the N-components at the five sites in Denmark and in other papers we have validated against data from the EMEP network. In order not to replicate these validations in the current manuscript, we have now included the appropriate references in Section 2.3.1, where the DEHM model is described. The following text has been included:

"In previous studies DEHM has been validate against measured depositions and concentrations of various nitrogen components across the EMEP measuring sites in Europe and the model is seen to captures the overall measured patterns (Brandt et al., 2012; Geels et al., 2005; Pul et al., 2009). In a recent study, measurements of air concentrations and wet depositions of nitrogen components covering the years 1990-2009 at the five main monitoring sites in Denmark have been used for validation of DEHM with a resolution of 16.67 km x 16.67 km over Northern Europe (Geels et al., 2012). Dry deposition fluxes have not been measured, but by applying the dry deposition velocities from the model, the measured air concentrations are converted to dry deposition fluxes and the estimates of the yearly total nitrogen deposition to land areas and marine areas have been compared to the model results for the same areas. As an average over the full period the model tends to overestimate the estimated deposition to land areas by 20% and underestimate the deposition with approximately 10% at the marine sites. "

It is true that a more detailed discussion of the uncertainties related to modelling of NH3 deposition is important and should be included. Regarding bi-directional flux and the possible impact of including this in the model; it is mentioned in the discussion in Section 4, with references to the experience from other studies. In the Discussion section we had some bullits on possible explanations to why the model overestimates the measured NH3 levels. We have now rewritten this to include a discussion of the uncertainties related to the dry deposition process and more details on the effects of

including bi-directional fluxes.

The intro to this section has been changed to: "The current validation at Danish sites shows a general overestimation by the DEHM model and the coupled system. The reason for this is not yet fully explored. In general the modelling of ammonia will be effected by uncertainties connected to the emission, the subsequent transport and conversion in the atmosphere as well as to the removal processes. In the following we discuss the possible reasons for the seen overestimation and the main uncertainties related to the involved processes: " And the following points have been added: " - in general parameterizations of dry deposition of ammonia are regarded as highly uncertain (Simpson et al., 2011) as they are based on very few field flux data and the involved exchange processes are poorly understood (Flechard et al., 2011). In this recent study by Flechard et al. inferential modelling with four different dry deposition modules showed large differences (up to a factor of 3) in estimated dry deposition velocities among the models. The main difference is related to how the non-stomatal resistance is described in the models. Only one of the inferential models included a non-zero compensation point for e.g. croplands (leading to a negative deposition velocity here), but otherwise the deposition module from the EMEP model gave the lowest deposition velocities (Flechard et al., 2011). As the description of the dry deposition in the DEHM and OML-DEP is very similar to the module in the EMEP model, this indicates that the modelled deposition in our study lies in the low range of such estimates. A possible underestimation of the deposition could hence be part of the explanation for an overestimation of the ammonia concentration at the Danish sites. - the omission of a full description of bi-directional fluxes (as discussed in a section above) over land and marine areas adds to the overall uncertainty related to the dry deposition process in DAMOS. As discussed in e.g. (2010) the inclusion of a dry deposition module with a description of the bi-directional exchange will lead to lower depositions, especially in canopies with high N status. Wichink Kruit (2012) found an increase in the ammonia concentration nearly everywhere across Europe (especially over agricultural fields), when including the bi-directional exchange in a CTM. For our simulations this

indicates that the models possibly overestimates the deposition over agricultural surfaces, leading to a too fast decrease in the atmospheric concentration and hence a too small transport of ammonia to more remote areas. However, part of this is indirectly accounted as the Danish ammonia emission inventory already includes an emission from grown crops."

More Specific comments by the reviewer:

Reviewer: p 1589, ln 23: footprint: Be more precise here: the deposition pattern? Answer: Changed to "deposition footprint"

Reviewer: P 1590, ln 25: Vd on what time scale?; as is indicated above the flux is bi-directional so episodes of negative Vd will occur. So the indicated range is not complete. Answer: True. We have changed the text to: "…. and the dry deposition (with typical dry deposition velocities in the order of 0.03 to 5.0 cm/s depending on surface and meteorological conditions) (Hertel et al., 2006). The bi-directional flux mentioned above, can also lead to negative deposition velocities over saturated ecosystems."

Reviewer: p 1591; ln 22-25: I suppose it is meant: the goal of this paper; the hypothesis as such is not so new and has been proven to be true. Answer: True, we have reformulated this to: "It is our goal to combine high resolution emission inventories with local scale and regional scale CTMs in a coupled system, in order to improve the model performance with respect to assessing ambient NH3 levels and resulting depositions to this patchwork of ecosystems."

Reviewer: P 1594; ln 3: area sources: how high are these crop emissions and how are these incorporated in the model calculations. That is: how do you parameterize deposition to the crops while meantime you have emission from the crops? Answer: Out of the national totals, the emission from crops is on the order of 7%. There is no link between the area emissions and the deposition. A compensation point is not included and the models deposit NH3 also to the areas with crops. We now include a note on this in the section where these emissions are mentioned.

Reviewer: P 1596; ln 26; what is meant there? Diffuse sources on an resolution of 16.67x16.67 km grid? Answer: In order to make the text more clear, it has been reformulated to: "Also other anthropogenic emissions are for the Danish area included with a higher resolution where possible. Emissions from road traffic are included on resolution of 1 km x 1 km, while other emissions sources like industry, power plants etc. are included on a 16.67 km x 16.67 km grid, based on results from the HYSCENE project (Jensen et al., 2010). "

Reviewer: P 1597; ln 14-18; what does this validation say? If the concentration is so well modelled, does this mean the background contribution (>20 km) is not important? This is not in line with the conclusions of the paper where it is stated that over 50% is from non-local sources. Answer: In the referred previous work, background concentrations from DEHM were included. We have now moved this small section to the end of section 2.3.2 where the coupled system DAMOS is described. It is more appropriate to include the text here.

Reviewer: P 1597; ln 26; this conversion is dependent on the SO2-concentrations. Is the factor corrected for the difference between the SO2 concentrations between 1989 and now. Answer: No we use the original transformation rate from Asmann et al. (1989) in OML-DEP. Now the NH3 concentration is reduced by ca. 10% within the domain. Due to the present day low acid levels a better first-order approximation would be to disregard this transformation. Another solution would be to input an hourly chemical transformation rate from the DEHM model based on the excess acid at the boundary of the local scale model. We have now included a comment on this in the bullits, where the uncertainties are discussed: "In OML-DEP the conversion from NH3 to NH4 is probably too large as it is based on the acidity level in the 1980s. The acidity level has decreased significantly since then. However, this will only have a minor effect on the ammonia level in the OML-DEP model."

Reviewer: P 1598; ln: just to get it right; OML is only used around monitoring sites or nature reserves and as a one way coupling? So from DEHM to OML and not back?

Answer: Yes, the two models are one-way coupled and OML-DEP is only setup for 16 km x 16 km domains aground the sites of interest. We have added this information to Section 2.3.2.

Reviewer: P 1600: ln.23-26: what would be a possible explanation for the overestimation in spring and summer? Answer: We believe that it linked to the fact that individual fields in reality are smaller than our 400 m x 400 m grid. So in the model the impact of areas sources will due to spatial averaging become to large. The measuring sites are located typically a few hundred meters from the nearest field and will therefore not be so directly influenced by the area emissions (that peaks in spring). We have added the following text to the discussion section: "The overestimation of the concentration in spring for DAMOS is likely caused by the spatial averaging of the area sources into the model grid. While the measuring sites are located at some distance from individual fields, the model results will be directly influenced by area sources with in the 400 m x 400 m central grid cell. "

Reviewer: P1602: section 3.4: Runs are made with and wthout local emissions. How are nonlinair processes treated in those runs (like wet deposition). (Or: how is the split made between local and non-local wet deposition?). Answer: It is only the local scale model OML-DEP that has been run with and without local emissions. Wet deposition is not included in the local scale model, so we do not really understand this question, sorry.

Reviewer: P1603: ln 15-20: you conclude a bit contra intuitively that the large scale model DEHM overestimated ammonia concentration. Since long it is found that CTMs underestimate ammonia concentrations because of their large horizontal scale. I think you should add to your discussion that DEHM already overcomes this problem by calculating at a much higher resolution. Answer: Good point. We have now included the following text in the discussion: "In the past regional scale CTMs have been found to underestimate the observed concentration of NH3 due to the relative coarse resolution of e.g. 50 km x 50 km (Pul et al., 2009b). In the current setup a much higher resolution

is applied in the CTM over Denmark and the regional model tends to overestimate the NH3 levels. In the Danish area with many agricultural sources the direct impact of local emissions becomes too high when applying a 5.56 km x 5.56 km grid resolution in the model. However, by coupling the regional model to. . .. ."

Reviewer: P1605: ln 26: what are large computational costs? What will be the run time for evaluating 100 N2000 areas on a 400x400m grid on a domain of say 16x16 km? Answer: ca. 12 CPU hours pr site on a standard PC. I.e. on a PC with two CPUs it will take approximately 25 days for 100 N2000 areas. As part of NOVANA we make calculations for ca. 130 sites every year. We have added this information to the manuscript.

Reviewer: P1606; ln 1 and further: since dry deposition is a very important term in the mass balance of ammonia and so plays a very important role in the comparison to measurements, what do the authors think the uncertainty is in the dry deposition velocity and what effect it has on the modelled concentration and local deposition? Answer: We have now included a discussion of the uncertainty related to the dry deposition process in the bullit point on page 1607 (see above).

Reviewer: P1606: ln 14; references are not in list. Answer: Have been included now.

Reviewer: P1606: ln 25; why haven't the authors checked their calculations of ammonia aerosol and ammonia concentrations in precipitation to measurements? That also gives a very good insight whether the ammonia balance is well modelled. Answer: We completely agree and have of course also made such comparisons with DEHM results and data from both the Danish sites and EMEP sites. However, in the current study where we use the coupled model for simulations of NH3 we have chosen to focus on the results for the NH3 concentration and then only refers to the comparisons with the other N components.

Reviewer: P1607: ln 3: not nicely formulated; I think models refer to DEHM and DAMOS and not the 4 models mentioned above? Answer: True. We have reformulated

to: “The current validation at Danish sites shows a general overestimation by the DEHM model and the coupled system. The reason for this is not yet fully explored. . .. .”

Reviewer: P1607; second bullit: do the authors mean the same effect as is found for DEHM, i.e. emissions further away from a measurement site do have a too large effect at the measurement site because of the emissions spread over the grid? Be more clear. Answer: Yes that is exactly what we suggest. We have reformulated to make more it clear: “in the regional model the spatial averaging of emissions leads to an overestimation of concentrations in high emission areas. It is possible that even the 400 m x 400 m resolution in the local scale model at some sites will lead to a too high impact of nearby sources. A newer version of the model will hopefully be able to run with a higher resolution (100 m x 100 m) and the same domain size of 16 km x 16 km. Thereby the impact by local emissions can be handled more realistically in the model.”

Reviewer:P1607: ln 9, 10: which not witch; what is an up-concentration? What would be the reason for an overestimation of the emissions during nighttime? Answer: Our parameterization of the emission is driven by the temperature at 2 m , which is ok for daytime conditions. Most likely the surface T will be lower than the 2 m T during night, so the real emission will be lower than we estimate. We have added the following to the manuscript:

“- the parameterisation of the emission does not reflect the true diurnal cycle, but e.g. overestimates the night-time emission, which combined with typical limited night-time atmospheric mixing leads to higher concentrations of ammonia near the surface. The temporal variation of the emission is currently driven by the air temperature at 2 m (Skjøth et al., 2004) as provided by the meteorological model. In reality the emission is most likely driven by the surface temperature, which especially during night time will be lower than the applied temperature at 2 m above the surface. Hence the true emissions will be lower during night than estimated by this parameterization.”

Reviewer:P1607: fourth bullit: why should there be an underestimation? Where is this

suggestion based on? Answer: We base this on a analysis of NH4 concentrations (underestimated by the model) and NHx (well captured). We have now reformulated and moved some text: “- a potential underestimation of the chemical conversion from ammonia to ammonium in DEHM, which is directly linked to the content of acid components in the ambient air and the parameterisation of the exchange between gas phase and aerosol component. When looking at the other nitrogen components simulated by DEHM it is for example seen that the results show a tendency for underestimating the ammonium concentrations at the Danish sites, while the sum of NH3 and NH4 (NHx) is well captured (Ellermann et al., 2010). This is likely linked to a general underestimation of sulphate in the model. In OML-DEP the conversion from NH3 to NH4 is probably too large as it is based on the acidity level in the 1980s. The acidity level has decreased significantly since then. However, this will only have a minor effect on the ammonia level in the OML-DEP model.”

Reviewer:P1607: ln 21: borders not boarders Answer: Ok

Reviewer:P1608: ln 1: do not does Answer: Ok

Reviewer: P1608: ln 1-3: this effect is in my opinion rather far-fetched. If this is not underpinned by some estimates it has no value. Answer: We have now included more details on this issue and references to the literature where it is described. So this bullit point now includes the following:

“- the models do not take an increased deposition at the edges of woods into account. Up to fourfold increases in atmospheric deposition at the forest edges have been reported (De Schrijver et al., 2007). Increased dry deposition seems to be the reason for this so-called edge effect, which can continue up to distances of 50-150 m within the forest (Spangenberg and Kolling, 2004). Omission of this in grid cells with forest might lead to an underestimation of the deposition and hence an overestimation of the concentration.”

Reviewer:P1608; ln 15: formulation is a bit strange. In general: line 4-15: are elabo-

rations of the bullits above and should be merged there. Is more clear. Answer: As suggested we have merged this text into the bullits in the text above.

Reviewer:P1608: ln 22: how many sites? Answer: Sorry a word was missing. It should be “plan for each of the ca. 250 NATURA 2000 sites”

Reviewer:P1609 and Figure 9; crucial in this assessment is how accurate are the modelled depositions? If firm conclusions are drawn about exceedences of Natura2000 areas it should be indicated what the uncertainties are? So it should be indicated in Figure 9 with bars what the uncertainty in deposition is as is done for critical loads. And than again draw conclusions. Answer: True – this a very important point raised by the reviewer, however, it is not a trivial task to asses the uncertainties related to the modelled depositions. We have now included an uncertainty range in Fig. 9. We have not been able to find uncertainty estimates for other regional models applied by various research groups across Europe to compare with. The text have been changed to:

“Based on experience from the Danish Background Air Quality Monitoring Program where measured and modelled nitrogen components at the five main Danish stations are analysed each year, we approximate the uncertainty related to the annual total nitrogen deposition to land areas to be on the order of +/- 40% (Ellermann et al., 2011). In Fig. 9. the simulated total N load at 26 specific nature areas in the Aarhus region in Denmark are displayed together with the critical loads related to the general nature types within each nature area. The critical load is given as an interval and the simulated values are show as a central estimate with an uncertainty interval of +/- 40%. The central estimate is at 13 areas seen to be above the upper limit of the critical load and for nine of these sites the DAMOS results based on different scenarios showed that it is not possible to get below this upper limit even if the local sources are reduced. At none of the sites the central estimate of the N deposition was below the critical load, but at nine sites the lower end of the uncertainty interval is below the lower limit of the critical load.”

And in the final conclusions we now include the following text:

“. . ...However, there are still large uncertainties related to modelling of the nitrogen load and theses should be taken into account when the model system is used in environmental management.

- A better representation of the surface exchange of ammonia in the next version of DAMOS can potentially reduce the uncertainty. Nevertheless, the surface exchanges of nitrogen components are highly parameterized in current CTMs and dedicated measurements of both the gas and particle components are needed in order to constrain the applied model parameters.”

Reviewer:Ln 15: I do not agree that the annual variation in deposition due to meteorology should be taken into account. It is doubtfull if this is ecologically relevant and the issue of the total uncertainty in the deposition calculation is far more important. Answer: We agree that the total uncertainty is more important, but we do not think that it is fair just to disregard the impact of inter-annual variations in meteorology. We have modified the text to: “However, the current analysis of the inter-annual variations (Sect. 3.2.1) show that year-to-year variation in the meteorology alone can lead to variations in the annual nitrogen load on the order of approximately 10-20 %. These variations are well below the overall uncertainty of such estimates, but should if possible be taken into account when the estimated nitrogen loads are used for management. Alternatively, the estimated nitrogen load should be based on model simulations covering several years e.g. in order to avoid the impact of a single extreme year. ”

Reviewer:P1610: ln 13: overestimates Answer: Ok.

Reviewer: Fifth bullit: is not very well elaborated in the paper. Where is this the case? Answer: As described in Section 3.1. the modelled spatial patterns of dry deposition of NH3, show potentially very high loads close to the sources. In table 4 the % of locally emitted NH3 that deposit within the 16 km x 16 km domain is given for the main sites. We have modified the statement somewhat to: “The fraction of locally emitted NH3

depositing locally (here in a 16 km x 16 km domain) is on the order of 15% to 30%."

Reviewer:Sixth bullit: signal = deposition? Answer: Yes, we have changed that accordingly.

Interactive comment on Biogeosciences Discuss., 9, 1587, 2012.