**Biogeosciences
Discussions**

Interactive comment on "The influence of model grid resolution on estimation of national scale nitrogen deposition and exceedance of critical levels" *by* A. J. Dore et al.

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Response to Referee #1

General comments:

With respect to methodology, then the authors have used the FRAME model at 1km with 1km input in for meteorology and emission. Results from these model results are then aggregated to two coarser grid resolutions, 5km and 50km that are compared with the 1km model results. These two coarser grid resolutions are identical to grid resolutions used by common CTM model that have been used for either European scale calculations such as the EMEP model (Simpson et al., 2003) or specific UK calculations

Full Screen / Esc

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Interactive Discussion

Discussion Paper

such as the EMEP4UK model (Vieno et al., 2010) and are as such very relevant. However, the results would have been stronger, if the authors also had chosen to aggregate input data to 5km (neglecting the 50 km resolution) and then also made calculations on a 5km grid in a similar way as in Hallworth et al (2010) as a comparison to the presented calculations. An alternative is however, if sufficient model results from the FRAME model from previous publications (e.g. Hallsworth et al., 2010; Kryza et al., 2011; Zhang et al., 2011) provides sufficient material to provide an in depth discussion in relation to the three scenarios. This issue is directly related to the aim of the paper. The authors write on page 12084-85: "The aim of this work is to investigate the influence of spatial averaging of modelled air concentrations and deposition (effect 1 above) on the exceedance of critical loads for nitrogen deposition.". From an endusers point of view, this result is very interesting as there is a recommendation to use a 1km reference data set compared to a 5km data set. But from an atmospheric modelling point of view, which is the topic in this manuscript, then in many cases it will make less sense to aggregate this type of model data (based on 1km input data) to a coarser resolution (e.g. reducing the data set which limit the amount of meaningful analysis).

The model was run using a 5 km resolution simulation for the model grid and all the input data. A comment has been added on the comparison of the results from this simulation and the 1 km resolution simulation.

"The data presented here compares concentrations and deposition from a 1 km resolution simulation with those re-gridded at resolutions of 5 and 50 km. This ensures conservation of mass when comparing the exceedance of critical loads. However it is also of interest to compare the results of the 1 km resolution simulation with those from a simulation with a model grid resolution of 5 km (effect 2, as discussed in the introduction). The comparison showed that the chemical transformation rate from gas to aerosol was somewhat more rapid with the 5 km resolution simulation. This can be attributed to the increased instantaneous mixing of ammonia with acid gases in the coarser resolution simulation. This resulted in lower dry deposition of nitrogen

(mostly from HNO3, NO2 and NH3 gas) and higher wet deposition (predominantly due to washout of ammonium and nitrate aerosol). National deposition of reduced and oxidised nitrogen was higher by 2% and 7% respectively with the 5 km resolution simulation."

The authors use relatively few citations from peer reviewed journals (15-16) and in comparison to that, a relatively high number of citations from different types of sources such as reports (13-14). Additonally, the manuscript contains a number of unsupported statements, especially in the introduction. I will therefore recommend a thorough investigation of the manuscript with respect to extended use of citations from peer reviewed scientific journals. I have a listed a number of possible locations in a list below this review.

In response to different reviewers' comments, the number of citations has generally been increased and the balance changed to 9 reports & books and 31 journal publications. However some reports have been retained where they represent the most appropriate reference. In addition, it should be noted that many of these reports have been peer-reviewed and are easily accessible via the reported web links.

There are a few unclear sections in the manuscript and the paper could benefit from a more in depth discussion concerning uncertainty of the applied methodology and a comparison with similar results, most likely from the UK, Netherlands and Denmark.

References to other relevant studies have been included. A discussion on uncertainty due to the difference between various model parameterisations has been included in the introduction: "Significant differences in estimation of the exceedance of critical loads may be obtained with different modelling techniques. Choice of chemical parameterisation, calculation of deposition velocities and estimation of precipitation are all factors which can lead to a divergence amongst models in their estimation of nitrogen deposition. The seeder-feeder effect is explicitly represented in FRAME (Fournier et al., 2005) with an enhanced washout coefficient applied to orographic remain. This

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

simulates the efficient removal of particulate nitrogen incorporated in cloud droplets and washed out from precipitation from above. Eulerian models, i.e. EMEP (Fagerli et al., 2011; Vieno et al., 2010a) and CMAQ: Chemel et al., 2010) have more complex microphysical schemes for formation of rain and cloud but do not explicitly include a parameterisation of the seeder-feeder effect. The representation of land cover and deposition to different vegetation types may vary between models and their applications. Whilst some applications use a dominant land category to derive deposition in each model grid square, others calculate deposition explicitly to different land categories. For ammonia gas in particular, the dry deposition velocity may be approximately an order of magnitude higher for forest and acid grassland than for improved grassland. Five different land classes are represented in FRAME (forest, semi-natural grassland, improved grassland, arable and urban). In the EMEP model 16 land classes are used, including sub-divisions of the arable and forest classes for detailed ecosystem effects studies."âĂČ Minor comments to the text in the manuscript:

Page 12081 line 8-10: This sentence only concerns in-cloud scavenging. The authors must also consider below cloud scavenging. A sentence was added: "Gases and particles may also be washed out of the boundary layer below cloud by falling rain drops."

Page 12083, line 2.4: The sentence "Models have the added advantage that calculations are made at a large number of model grid cells, invariably with much higher spatial density than that which can be achieved by measurement alone." This statement is likely too general. A good example of high resolution observations with large geographical coverage is satellite based observations (e.g. Clarisse et al., 2009).

This section was modified: "Models have the added advantage that calculations are made at a large number of model grid cells, invariably with much higher spatial density than that which can be achieved by surface monitoring of concentrations in air and precipitation. Remote sensing from satellites has however been demonstrated to be an effective technique to measure gaseous nitrogen compounds in air with global

coverage (Clarisse et al., 2009)."

Page 12083, line 23. Please change "The model operates" to "the long range transport model" and add also the scales for Gaussian local scale model

The text was changed accordingly

Page 12083, line 26-29. Please extend the sentence" Vogt (2011) calculated ammonia concentrations and deposition at a 25m resolution in an agricultural landscape. Fine resolution model simulation was demonstrated to be necessary to reproduce measured ammonia concentrations." The extensions could include a very short description on the tools and methods in a similar manner as the authors describe the OPS and DAMOS systems.

Further detail of the study has been added: "Vogt et al (2011) calculated ammonia concentrations and deposition at a 25 m resolution in an agricultural landscape. A landscape-resolved emissions map was input to a Lagrangian local dispersion model and the modelled concentrations were compared with measurements from a dense network of passive diffusion and denuder samplers. Fine resolution model simulation was demonstrated to be necessary to reproduce the measured ammonia concentrations."

Page 12084, line 24. Please add (FRAME) and the reference as this is the first place in the text the model name is given.

The acronym description and references have been added

Page 12084. The last section. Here the authors state that they want to use the FRAME model results at 1km, 5km and 50km. A few more lines why exactly it is interesting to study model results at these resolutions and not use the results from previous studies.

The text was expanded to provide an explanation of the choice of data resolution used in the study. Further discussion of the 1 km version of the model is included at the end of the 'Model Description section

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

"The results of a 1 km model simulation have been mapped at resolutions of 1 km, 5 km and 50 km. The 1 km resolution data represents a recent development in the capability of the model (Hallsworth et al., 2010) whereas previous studies were undertaken using 5 km resolution data. 50 km resolution data is of interest as this is the resolution of the EMEP model (Fagerli et al., 2011) used at a European scale to provide support for the Convention on Long Range Transport of Air Pollutants."

Page 12085. The use of an annual precipitation map and its actual resolution is likely to affect the calculated wet deposition. What is the underlying quality of that map, especially with respect to spatial resolution? Additionally, then it is likely an advantage with respect to wet depositions, that the authors use a high quality estimate of the annual precipitation to calculate the wet deposition. Precipitation from numerical models are one of the most uncertain factors. This could go into a specific sub-section or a general topic that discuss uncertainties.

A paragraph was included in the precipitation section: As the measurement of rainfall with standard rain gauges is a simple and accurate process, this technique can produce a relatively precise precipitation map. Standard measurement error due to under-capture is typically about 5% (Keller et al.). Measurements in upland regions, with more highly spatially variable precipitation, are less concentrated. This will therefore result in greater uncertainty in precipitation and therefore nitrogen deposition in the high rainfall regions, which also contain sensitive ecosystems. More complex Eulerian models (Vieno et al., 2010 ; Chemel et al., 2010) dynamically generate precipitation with a meteorological model. However, modelled estimates of precipitation are known to be subject to considerable uncertainty.

Page 12085, line 18. please correct to the year 2011 in the reference to "after Poland (Kryza et al., 2011)". I will also suggest that the authors use recent studies that have been published in Biogeosciences (Zhang et al., 2011)

The reference was corrected and reference to Zhang et al. (2011) was added.

Page 12085, line 26. The authors name the model FRAME-Europe. This version of the FRAME model is new in the manuscript. Please provide a reference to that model or describe what it is.

The use of the term 'FRAME-Europe' was removed and paragraph clarified:

âĂć Input gas and aerosol concentrations at the edge of the UK model domain are calculated with FRAME using a European-scale simulation run on the EMEP 50 km scale grid and emissions provided from the EMEP data base (Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe http://www.emep.int/UniDoc/index.html).

Page 12088: About emissions in general. It is unclear what type of emissions the authors use for ammonia, except that they originate from the agricultural sector. Do the authors use one uniform emission factor for cattle for the entire model domain? Or do the emission factors depend on both production methods and geographical area (e.g. variations in annual emission due to overall variations in climate)? Do the authors have a hourly, daily or season emission profile (e.g. Hellsten et al., 2008; Skjøth et al., 2011).

Ammonia Emissions are not dependent on geographical region or climate but emissions factors are calculated according to sub-livestock category (i.e. dairy, heifers, heifers in calf, beef cows in calf, bulls, other cattle older than 2 years, other cattle 1-2 years, calves etc ..). A diurnal profile in ammonia emissions is included in the model. The text has been modified to include these factors

"Emissions of ammonia are predominantly from agriculture and in particular due to direct emissions from livestock. Total annual emissions are estimated based on census data of farm animals and estimates of emissions factors per animal at a sub-livestock category level. The spatial distribution of agricultural ammonia emissions uses census data at a parish level and distributes15 livestock emission categories within each parish according to land use category and its suitability for agriculture. Emissions of ammonia

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Interactive Discussion

Discussion Paper

from a wide range of non-agricultural sources are also included (Dragosits et al, 2000). "

12090, line 9. The geographical areas "The Pennines" and "The Lake District" are probably well known by readers with good knowledge of English geography, but less known by others readers. Please be a bit more general (e.g. adding North West England) to The Lake District and similar for The Pennines.

Geographical locations of these regions have been added to the text

Page 12091, line 13-16. The authors argue that there is a clear improvement in model calculations with increasing resolution. It is partly unclear what the statistics actually cover. The particular section would also improve if the authors discuss the rural and the AURN network model results separately in relation to the purpose of the monitoring sites. The AURN network shows as expected improvements of all parameters down to 1km but the rural stations simulations does not. The authors could highlight that and discuss on more depth the cause to these differences.

Further details of the AURN and rural monitoring networks have been added:

"Here we consider model correlation with measurements of NO2 gas concentrations. In the UK, monitoring of NO2 concentrations is undertaken with both the rural monitoring network using diffusion tubes (20 sites) and the Automated Urban and Rural Network (AURN) using chemiluminescence. The 12 rural sites in the AURN are considered here for validation of the model. The AURN is intended to check if statutory air quality standards form EC Directives and targets are met and to identify long-term trends in air pollution concentrations. The rural monitoring network provides valuable information regarding background concentrations of NO2."

It is difficult to speculate on reasons for the precise details of the correlation statistics. In reality these will depend on local conditions at each individual site. The aim here is really to show an overall improvement with use of the 1 km resolution data making use

Interactive
Comment

Full Screen / Esc

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Interactive Discussion

Discussion Paper

of data from both networks.

Page 12092, line 2-12: The depositions estimates are a bit unclear how the have been made and what the main uncertainties are. Is it right, that the authors calculate depositions to each of the land cover types in the FRAME model (6 land cover types)? And is true that the authors afterwards associate the all the sensitive ecosystems to one of these 6 land cover types, thereby assuming that the deposition to these ecosystem is what have been calculated in the FRAME model (to one of the 6 land cover types)? If so, then there will be a potential systematic error for some ecosystems. If heath and bog use the mechanistic structure of woodlands (e.g. higher roughness) then dry depositions might be overestimated to heath and bog compared to reality as these ecosystems in reality are less rough than woodlands. Please comment on that, and if it is relevant, then please discuss this in a final version of the manuscript.

The description of the calculation of exceedance of critical loads has been simplified: "The calculation of the exceedance of critical loads for nitrogen deposition is described in detail in Hall et al. (2006) and RoTAP (2011). Exceedances were calculated separately for each habitat type using 1km critical loads data and ecosystem-specific deposition (ie, moorland deposition for the grassland, heath, bog and montane habitats, and woodland deposition for the woodland habitats). However, rather than include separate exceedance maps for each habitat, the exceedance maps presented here (Figures 6(a) and 6(b)) are based on 5th-percentile critical loads that combine data for all habitats. The 5th-percentile critical load is set to protect 95% of the total sensitive habitat area in each 1km square."

Yes, the model separately calculates deposition to different land types using land-cover specific roughness and canopy resistance. This explanation has been modified in the model description:

âĂć "The parameterisation employed to calculate dry deposition of SO2, NOx and NH3 includes vegetation specific canopy resistance and surface roughness."

Page 12093, line 16-23: This paragraph seems unsupported by scientific arguments. Please explain why, as there are no model simulations that shows both scenarios. Only the scenario in Fig 3 is presented. I need a scenario that presents model results that represents: "In regions where emissions are densely concentrated in areas of intense agriculture and urban agglomerations which are distinctly separated from natural ecosystems, high grid resolution is of lower importance."

The statement was removed.

Page 12093, line 29: What is the unit keq Ha-1? This is also seen on Fig 4a,b. The unit is kilo-equivalents per hectare which is commonly used for calculations of the exceedance of critical loads.

Page 12095, line 1-2: "Less high resolution data (i.e. 5 km) was found to be adequate for calculating national scale summary statistics on the exceedance of critical loads." I am very uncertain if this conclusion holds. They have aggregated 1 km resolution data that are based on 1km 1 emission input and 1km precipitation estimates to 5km. This is not the same as using 5km input in emission and precipitation and 5km receptor points. This topic is directly related to the second paragraph in this review.

The statement was removed from the text

Page 12095, line 3-6: Input data seems to be among the most sensitive things. The authors conclude that improvement on livestock number and agricultural practice is the key. How about dependence on climatic variables (e.g. temperature, precipitation)? This is directly related to the specific question concerning ammonia emissions on page 12088.

A comment on the dependence of emissions on meteorology was added:

"Improvements in information on the spatial distribution of livestock numbers and on agricultural practice and the dependence of emissions on meteorological variables (i.e. temperature, precipitation, wind speed) are necessary to achieve better estimates of

Interactive
Comment

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Interactive Discussion

Discussion Paper

nitrogen dry deposition."

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Minor comments related to figures and tables in the manuscript:

The number of decimals and accuracy in all figure legends could be re-assessed. As an example:

Fig 1a: 100-200 etc would here be more appropriate.

Additionally, is part of the green areas (0-100m) actually sea water or just low elevation terrain? Water areas must be without any color.

Fig 1b: <1000, 1000-1400 etc would here be more appropriate. Note that the figure has three cells without colour in the upper left part of the figure. Is this because the legend does not cover the lowest precipitation values?

Fig 2: 0, 12, 24, 36,48,60 would here be more appropriate

Fig 3a: 0.55-3.68. 0.5-3.7 would here be more appropriate Etc

About Fig 5. The are no units on the axis in the scatter plots.

Figure 3a, 3b. Needs coordinates/distances along the figure sides as on Fig 4a and

4B Figure 4a, 4b. The legend to the right needs a title as in figure 3a,b (NOx air concentrations). List with possible locations that could benefit from a citation in this manuscript:

The recommended changes to the plots have been included

About Fig 1a. Where do the data about the topography come from and what is the resolution? Please provide a reference.

The digital elevation model used here comes from the SRTM (Shuttle Radar Topography Mission) database. The original resolution of the model is 3 arc seconds. The

Interactive
Comment

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Interactive Discussion

Discussion Paper

model was resampled to 250 m x 250m grid and reprojected from geographical coordinate system (lat/long) to Ordnance Survey National Grid for presentation. The reviewer recommends various additional references in addition to those requested by other reviewers. Whilst many of these have been included, a reference for the topography map is considered a lower priority.

Table 2. This table is unclear with respect to what it shows. Maximum value and minimum values. Are these model estimates and of what? The table needs units and a description of what the quantities are a summary of.

The maximum and minimum values are for the difference between the measured and modelled values. This has been clarified in the table and the units decribed.

About Fig 5 (left). It looks as there are fewer red circles in Fig 5 (about 12) compared to blue triangles and green crosses (about 20). Please explain why.

The number of points are identical for each resolution data set. However at certain sites the concentrations of 1 km and 5 km data are nearly identical so some degree of over-plotting of symbols is inevitable.

About the Standford Park Site. There is no description of where this site is. Please add coordinates

The co-ordinates have been added to the plot

References :

The introduction has been re-written and a substantial number of appropriate references have now been added, some of which were those suggested by the reviewer and others were selected by the authors.

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