

Supplementary materials

Manure acidification and air cleaners for ammonia abatement: A holistic assessment of the costs and effects on terrestrial, freshwater and marine ecosystems

Camilla Geels^{1*}, Steen Gyldenkærne¹, Tavs Nyord², Hans Estrup Andersen³, Eugenio Molina-Navarro³, Dennis Trolle³, Hans Thodsen³, Jesper L. Bak³, Maria Theresia Konrad¹, Berit Hasler¹, Kaj M. Hansen¹, Jesper H. Christensen¹ and Karen Timmermann⁴

1. Details on the atmospheric modeling

The Danish Eulerian Hemispheric Model (DEHM) is a chemistry-transport model describing the fate of air pollution in the atmosphere [1,2]. The model covers the Northern Hemisphere and includes several nests with higher resolution over specific regions. In the current setup the area of Denmark is covered by a horizontal grid with a resolution of 5.56 km x 5.56 km. Thereby the long-range transported contribution to the nitrogen load over Denmark is included at the same time as the national contribution is described in more detail. The vertical grid spans over 29 levels, with the highest resolution closest to the ground. The meteorological forcing is obtained from the numerical prediction model MM5v3.7 model [3], setup with the same domains as DEHM. The applied version of DEHM includes 67 different chemical species (gases and aerosols) and is based on a chemical scheme developed by [4]. The scheme has in DEHM been extended in order to include a better description of a number of nitrogen containing compounds. Dry deposition is based on the resistance method and is hence depending on e.g. the land-surface types and meteorology. Wet deposition is described as in-cloud and below-cloud scavenging. Emissions are typically based on a combination of several global, regional and national inventories. In the current setup the anthropogenic emissions for Denmark are based on a detailed inventory on a 1 km x 1 km resolution grid based on the SPREAD (Spatial Distribution of Emissions to Air) model [5,6]. For the European area emissions are based on the EMEP inventory, while non-European emissions are taken from the ECLIPSE5.0 dataset (<http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5.html>).

For Denmark, the temporal variation of the NH₃ emissions from the agricultural area is based on a parameterization describing the impact of meteorology, regulation, production methods etc. on the hourly emission [7-8]. The DEHM model has been extensively validated and used for a number of studies with focus on nitrogen deposition at the regional scale [9,10,11], national scale [12] and in combination with the OML-DEP model for studies at the more local scale [13,14,15,16]. The DEHM model is part of the Copernicus Atmospheric Service providing air quality forecast and analysis for Europe (<https://www.regional.atmosphere.copernicus.eu/>).

In the current study two one-year simulations have been carried out:

- The baseline with actual emissions for 2012. Here the total yearly agricultural NH₃ emission amounts to 59 kt NH₃-N for Denmark (this includes the emissions from houses, storage and manure application as given in Table 3 as well as emissions from crops and fertilizer use).
- The scenario with acidification and air cleaners. Here the total yearly agricultural amounts to NH₃ 35 kt NH₃-N for Denmark

The meteorological conditions have a significant impact on the overall deposition of N and also for the distribution between wet and dry deposition. Here we apply meteorological data for year 2009, which in Denmark can be considered close to an “average” meteorological year in terms of wind direction and precipitation, when compared to the previous 30-year period.

2. Details on SWAT calculations

The SWAT model (Soil and Water Assessment Tool, version 2009, [17]) is set up for the Northern Jutland region including the Limfjord catchment (Fig. 2). The model is based on a 32 m resample of a 1.6 m LIDAR Digital elevation model and the CORINE land use map combined with a field block map containing information on agricultural fields. A national coverage 3-layer soil map on a 250 m grid is applied [18]. The soil map is parameterized for SWAT using the Hyprss model [19]. The SWAT model is set up with 182 sub-basins of which 118 drain to the Limfjord. The sub-basins, match inlets and outflows of major lakes and monitoring stations. The SWAT model is driven by both gridded climate data (precipitation corrected for gauge under-catch [20] on a 10 km grid, solar radiation/20 km grid, wind speed/20 km grid) and by data from climate stations (temperature and relative humidity), provided by the Danish meteorological institute. The agricultural practice is included as 14 five-year crop rotations representing different forms of agriculture (pig farms, dairy farms, and arable farms at different fertilization intensities). The representation of agriculture in SWAT matches average regional statistics on crop use (area covered and relationship to farm type) and fertilizer application (chemical and/or manure applied to what crop at what time in accordance to farm type) for the year 2005 [21,22].

The SWAT model is calibrated in three steps, firstly against statistical crop yields (for all major agricultural crops), secondly against measured daily river water discharges at 10 monitoring stations and thirdly against observed daily (10 – 20 per year) river nitrogen and phosphorous loads (both mineral and organic fractions) at 8 stations. The calibration is performed using SUFI2 calibration procedure of the SWAT-CUP calibration program [23]. The calibration of river water discharge, is performed using the “sum of squared errors” objective function, while the nutrient loads are calibrated using the Nash-Sutcliffe efficiency objective function [24]. The period 2000-2005 is used for calibration, while 2006-2009 is used for validation.

The baseline and the emission scenario are simulated in the SWAT model by changing two different model inputs. The changes in atmospheric deposition as obtained from the DEHM model, are incorporated by the NO₃ concentration in rainfall. The changes in nitrogen content in slurry (organic fertilizer) are incorporated by adjusting the nitrogen concentrations in the applied organic fertilizers in the scenario.

3. Details on the calculations for the freshwater ecosystems

Results of the SWAT model were used as input to the next model in the overall system, where the effects on the freshwater ecosystems in two typical Danish lakes are quantified. The GOTM-FABM-PCLake ecosystem model [25,26] was set up to represent a shallow (max depth 2.7 m) and a deep (max depth 17 m) Danish lake, represented by the lakes Klejtrup and Tjele Langsoe. Both lakes are located within the case study catchment area, which drains into the Limfjord. The GOTM-FABM-PCLake model is a coupled one dimensional hydrodynamic and ecosystem model, which represents state-of-the-art within process-based aquatic ecosystem modelling. The model simulates a wide range of ecosystem state variables, which can help indicate the level of integrity of a freshwater ecosystem, such as total chlorophyll *a* and coverage of submerged macrophytes. We used these as indicators, when estimating potential impacts of manure acidification on freshwater aquatic ecosystems. The baseline set up for the two lakes, included river and nutrient inflow boundary conditions, based on a National model complex (DK-QNP) by [27], and local meteorological forcing based on reanalyzed ERA-Interim data [28] provided by the European Centre for Medium-Range Weather Forecasts (ECMWF).

4. Details on the calculations for the marine ecosystems

Effects on marine ecosystems as a response to the changed nutrient input following the scenario, were quantified using empirical models based on time series of marine data and nutrient loadings from the national marine monitoring program (NOVANA) as well as meteorological observations provided by the Danish Meteorological Institute. The empirical models were Partial Least Squares (PLS) regression models, with summer chlorophyll *a* concentration and light attenuation as the dependent variables and eight independent variables consisting of N-loading (ton year⁻¹), P-loading (ton year⁻¹), freshwater inflow (m³ month⁻¹), wind power (m³ s⁻³), surface irradiance (μmol photons m⁻² s⁻¹), salinity (psu), water column stability (s⁻¹) and sea surface temperature (°C). Data used for either independent or dependent variables, were linearly interpolated to obtain time weighted averages and thereafter normalized to a mean value of 1 and a standard deviation of 1, in order to minimize the effect of variation between stations. The model development procedure is described in [29]. Briefly, selection of independent variables for each model was done by an iterative process using cross-validated multiple linear regression (MLR), in combination with stepwise exclusion of the explanatory variable that gave the lowest model error in the form of "Root Mean Square Error of Cross Validation" (RMSECV). MLR regression was performed on a calibration dataset constructed as a random subset of the total dataset. The selected variables were used to produce a PLS regression and for each variables root mean squared error, RMSE, of both the calibration data and the validation data was quantified and parameters that increased the error in the prediction of the validation data were left out. The PLS regression analysis was done in MATLAB® using a PLS program package from Eigenvector® and the selection process was written for this purpose.

References

1. Christensen, J.H., 1997. The Danish Eulerian hemispheric model - A three-dimensional air pollution model used for the Arctic. *Atmos Environ* 31 (24):4169-4191
2. Brandt J., Silver J., Frohn L.M., Geels C., Gross A., Hansen A.B., Hansen K.M., Hedegaard G.B., Skjøth C.A., Villadsen H., Zare A., Christensen J.H., 2012. An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution. *Atmos Environ* 53:156-176. doi:10.1016/j.atmosenv.2012.01.011.
3. Grell, G.A., Dudhia, J., and Stauffer, D.R., 1995. A description of the fifth-generation Penn State NCAR Mesoscale Model (MM5)NCAR/TN-398+STR, 1-22, 1995.
4. Strand, A., and Hov, O.: A 2-Dimensional Global Study of Tropospheric Ozone Production, *Journal of Geophysical Research-Atmospheres*, 99, 22877-22895, Doi 10.1029/94jd01945, 1994.
5. Plejdrup, M.S. and Gyldenkerne, S. 2011. Spatial distribution of emissions to air – the SPREAD model. National Environmental Research Institute, Aarhus University, Denmark. 72 pp. – NERI Technical Report no. FR823. <http://www.dmu.dk/Pub/FR823.pdf>.
6. Plejdrup M.S., Nielsen O-K., Brandt J., 2016. Spatial emission modelling for residential wood combustion in Denmark. *Atmospheric Environment*, Volume 144, Pp 389-396. <https://doi.org/10.1016/j.atmosenv.2016.09.013>
7. Skjøth, C.A., O. Hertel, S. Gyldenkerne, and T. Ellermann, 2004. Implementing a dynamical ammonia emission parameterization in the large-scale air pollution model ACDEP, *J.Geophys.Res-Atmos.*, 109: 1-13.
8. Skjøth, C. A., Geels, C., Berge, H., Gyldenkerne, S., Fagerli, H., Ellermann, T., Frohn, L. M., Christensen, J., Hansen, K. M., Hansen, K., and Hertel, O., 2011. Spatial and temporal variations in ammonia emissions - a freely accessible model code for Europe, *Atmos Chem Phys*, 11, 5221-5236. <https://doi.org/10.5194/acp-11-5221-2011>
9. Geels, C., Hansen, K. M., Christensen, J. H., Ambelas Skjøth, C., Ellermann, T., Hedegaard, G. B., Hertel, O., Frohn, L. M., Gross, A., and Brandt, J., 2012a. Projected change in atmospheric nitrogen deposition to the Baltic Sea towards 2020, *Atmos. Chem. Phys.*, 12, 2615–2629, <https://doi.org/10.5194/acp-12-2615-2012>.
10. Simpson, D., Andersson, C., Christensen, J.H., Engardt, M., Geels, C., Nyiri, A., Posch, M., Soares, J., Sofiev, M., Wind, P., Langner, J., 2014. Impacts of climate and emission changes on nitrogen deposition in Europe: a multi-model study. *Atmos Chem Phys* 14:23. <https://doi.org/10.5194/acp-14-6995-2014>.
11. Vivanco, M. G., Theobald, M. R., García-Gómez, H., Garrido, J. L., Prank, M., Aas, W., Adani, M., Alyuz, U., Andersson, C., Bellasio, R., Bessagnet, B., Bianconi, R., Bieser, J., Brandt, J., Briganti, G., Cappelletti, A., Curci, G., Christensen, J. H., Colette, A., Couvidat, F., Cuvelier, C., D'Isidoro, M., Flemming, J., Fraser, A., Geels, C., Hansen, K. M., Hogrefe, C., Im, U., Jorba, O., Kitwiroon, N., Manders, A., Mircea, M., Otero, N., Pay, M.-T., Pozzoli, L., Solazzo, E., Tsyro, S., Unal, A., Wind, P., and Galmarini, S., 2018. Modeled deposition of nitrogen and sulfur in Europe estimated by 14 air quality model systems: evaluation, effects of changes in emissions and implications for habitat protection, *Atmos. Chem. Phys.*, 18, 10199–10218, <https://doi.org/10.5194/acp-18-10199-2018>.

12. Ellermann, T., Nygaard, J., Christensen, J. H., Lofstrom, P., Geels, C., Nielsen, I. E., Poulsen, M. B., Monies, C., Gyldenkaerne, S., Brandt, J., and Hertel, O., 2018. Nitrogen Deposition on Danish Nature, *Atmosphere-Basel*, 9, ARTN 447. 10.3390/atmos9110447.
13. Geels, C., Andersen, H. V., Ambelas Skjøth, C., Christensen, J. H., Ellermann, T., Løfstrøm, P., Gyldenkaerne, S., Brandt, J., Hansen, K. M., Frohn, L. M., and Hertel, O., 2012b. Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS, *Biogeosciences*, 9, 2625–2647, <https://doi.org/10.5194/bg-9-2625-2012>.
14. Hansen, K., Sørensen, L. L., Hertel, O., Geels, C., Skjøth, C. A., Jensen, B., and Boegh, E., 2013. Ammonia emissions from deciduous forest after leaf fall, *Biogeosciences*, 10, 4577–4589, <https://doi.org/10.5194/bg-10-4577-2013>.
15. Hertel, O., Geels, C., Frohn, L. M., Ellermann, T., Skjøth, C. A., Lofstrom, P., Christensen, J. H., Andersen, H. V., and Peel, R. G., 2013. Assessing atmospheric nitrogen deposition to natural and semi-natural ecosystems - Experience from Danish studies using the DAMOS, *Atmospheric Environment*, 66, 151-160, <https://doi.org/10.1016/j.atmosenv.2012.02.071>
16. Sommer, S.G., Østergård, H.S., Løfstrøm, P., Andersen, H.V., Jensen, L.S., 2009. Validation of model calculation of ammonia deposition in the neighbourhood of a poultry farm using measured NH₃ concentrations and N deposition. *Atmos Environ* 43 (4):915-920. <https://doi.org/10.1016/j.atmosenv.2008.10.045>.
17. Arnold, J.G. and Srinivasan, R., 1998. A continuous catchment-scale erosion model. *Modelling Soil Erosion by Water*, 55: 413-427.
18. Greve, M.H., Greve, M.B., Bøcher, P.K., Balstrøm, T., Breuning-Madsen, H., Krogh, L., 2007. Generating a Danish raster-based topsoil property map combining choropleth maps and point information. *Geografisk Tidsskrift/Danish J Geogr.* 107:1–12.
19. Wosten, J.H.M., Lilly, A., Nemes, A. and Le Bas, C., 1999. Development and use of a database of hydraulic properties of European soils. *Geoderma*, 90(3-4): 169-185.
20. Allerup, P., Madsen, H. and Vejen, F., 2000. Correction of precipitation based on off-site weather information. *Atmospheric Research*, 53(4): 231-250.
21. Thodsen, H., Andersen, H.E., Blicher-Mathiesen, G. and Trolle, D., 2015. The combined effects of fertilizer reduction on high risk areas and increased fertilization on low risk areas, investigated using the SWAT model for a Danish catchment. *Acta Agriculturae Scandinavica, Section B – Soil & Plant Science*, 65(sup2): 217-227. <https://doi.org/10.1080/09064710.2015.1010564>.
22. Trolle, D., Nielsen, A., Rolighed, J., Thodsen, H., Andersen, H.E., Karlsson, I.B., Refsgaard, J.C., Olesen, J.E., Bolding, K., Kronvang, B., Søndergaard, M. and Jeppesen, E., 2015. Projecting the future ecological state of lakes in Denmark in a 6 degree warming scenario. *Climate Research*, 64(1): 55-72. <https://doi.org/10.3354/cr01278>.
23. Abbaspour, K.C., Vajdani, M. and Haghighat, S., 2007. SWAT-CUP Calibration and Uncertainty Programs for SWAT. *Modsim 2007: International Congress on Modelling and Simulation*: 1603-1609.
24. Nash, J.E. and Sutcliffe, J.V., 1970. River flow forecasting through conceptual models part I -- A discussion of principles. *Journal of Hydrology*, 10(3): 282-290.
25. Hu, F., Bolding, K., Bruggeman, J., Jeppesen, E., Flindt, M. R., van Gerven, L., Janse, J. H., Janssen, A. B. G., Kuiper, J. J., Mooij, W. M., and Trolle, D., 2016. FABM-PCLake – linking aquatic ecology with hydrodynamics, *Geoscientific Model Development* 9: 2271-2278, <https://doi.org/10.5194/gmd-9-2271-2016>.
26. Andersen, T. K., Nielsen, A., Jeppesen, E., Hu, F., Bolding, K., Liu, Z., Søndergaard, M., Johansson, L. S., and Trolle, D., 2020. Predicting ecosystem state changes in shallow lakes using an aquatic ecosystem model: Lake Hinge, Denmark, an example. *Ecological Applications* 30(7): <https://doi.org/10.1002/eap.2160>
27. Windolf, J., Thodsen, H., Troldeborg, L., Larsen, S.E., Bogestrand, J., Ovesen, N.B., Kronvang, B., 2011. A distributed modelling system for simulation of monthly runoff and nitrogen sources, loads and sinks for ungauged catchments in Denmark. *J. Environ. Monit.*, 13: 2645-2658. <https://doi.org/10.1039/C1EM10139K>.
28. Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A.C.M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S.B., Hersbach, H., Hólm, E.V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A.P., Monge-Sanz, B.M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F. , 2011. The ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Q.J.R. Meteorol. Soc.*, 137: 553–597. doi: 10.1002/qj.828.
29. Erichsen, A.C. (Ed.), Timmermann, K. (Ed.), Christensen, J.P.A., Kaas, H., Markager, S., Møhlenberg, F., 2017. Development of models and methods to support the Danish River Basin Management Plans. Scientific documentation. Aarhus University, Department of Bioscience and DHI, Denmark. 191 pp. https://dce.au.dk/fileadmin/dce.au.dk/Udgivelser/Oevrige_udgivelser/RBMP_models_sd_2017__002_.pdf.