

We are grateful for the constructive comments from the Reviewer. We have addressed all the comments and questions raised by the reviewer 1. In our response the comments have been marked in black and our responses have been marked in blue. Furthermore, the manuscript has been checked by a native speaker.

1. I agree with the other referees that the biggest problem with the paper is the overinterpretation of rather few measurements. I will not repeat their points, but this aspect needs to be toned down.

We agree with the Reviewers that the conclusions provided in the text are too strong. We have included several changes in the text. Please see reply to comment 1 and 2 from the Reviewer 1.

2. Abstract (and related discussions)

My only real objection here concerns line 25, which claims that dynamical approaches are a 'viable objective' for all CTMs. I am not convinced that this is really true. What are data requirements and shortages? Do the authors expect data on fertilizer practices, irrigation, soil characteristics, and legislation and farming traditions to be available in the near (or foreseeable) future?

We do not expect detailed data on fertilizer practices, irrigation and so on in the near future but it was shown by Skjøth et al. (2011) that even with scarce and rather uncertain information about agricultural practice and production methods, improvements in CTM modelling may be obtained from applying a dynamic NH₃ emission model. Our study suggests that results could be further improved by incorporation of national practices into the model. However, an application of a dynamic approach requires more computer power and lengthens a simulation time which is a disadvantage of this method. Recently it has also been shown that the concept of a sector based emission inventory (e.g. separating emission from fertilizer and buildings) and simulating the fertilizer application using a Gaussian Model with Growing Degree Hours was a viable approach for the global model Geos-Chem that was run on 2.5 x 2.5 degree resolution (Paulot et al., 2014). With this approach and reasonable assumptions it was possible to create global data with sufficient high quality that could be used in Geos-Chem and it was shown that this approach was better than fixed profiles for Europe, China and USA, respectively.

We have modified the sentence:

Page 2022, line 25

Implementing a dynamical approach for simulation of ammonia emission is a reliable but challenging objective for CTM models that continue to use fixed emission profiles. Such models could handle ammonia emissions in a similar way to other climate-dependant emissions (e.g. biogenic volatile organic compounds).

3. Other points on the abstract:

- could be shorter

The abstract has been shortened.

- omit or define NWP

Omitted.

4. P2024,L8. The cited Riddick paper is for tropical seabird colonies, which is a bit exotic for a paper dealing with Poland. The paper by Simpson et al. (1999) suggested that in Europe the NH₃ emissions from 'natural' sources were almost negligible compared to agricultural.

We have change the citation from (Riddick et al., 2014) to (Andersen et al., 1999), (Hansen et al., 2013) and (Sutton et al., 1997), which concern: ammonia emission from a spruce forest in Denmark, ammonia emission from a deciduous forest in Denmark and emission from hill surface (grass moorland and blanket bog) in the UK, respectively. We have included also the study of (Simpson et al., 1999).

Modified test:

Page 2023, line 7-8

Ammonia is mainly emitted to the atmosphere from agricultural operations (Bouwman et al., 1997), but also from natural sources (e.g. Andersen et al., 1999; Hansen et al., 2013; Sutton et al., 1997). Agriculture's share in total ammonia emission in European Union was 94% in 2010 (EEA, 2014) and is largely from animal excreta and fertilizers. The contribution of natural emission is negligible compared to agricultural for the most of European areas (Simpson et al., 1999; NATAIR 2007).

5. P2024. First paragraph - explain which regions are being discussed by the cited studies.

The focus is on European areas. We have clarified this in the text:

Page 2024, line 3

Ammonia affects the acidification of European soils that arises from the deposition of N from the atmosphere (Sutton et al., 2009; Theobald et al., 2009).

6. P2025 and elsewhere. There is no such thing as the 'WRF-Chem model for Poland'. WRF-Chem was not built for Poland, and there is no unique model version; there may even be several groups running WRF-Chem for Poland. Please state whose implementation of WRF-Chem you are referring to, and give this a name.

We agree with the comment. We have removed the reference to WRF-Chem throughout the text, where it was used in the context of the constant emission. Please see also reply to point 10 (below).

7. P2026,L14 - 'default values were implemented...'. Who, where? (In this study, or in Skjøth?)

We have clarified this in the text:

Page 2026, line 14

Default values were therefore implemented by Skjøth et al. (2011) for many European countries.

8. P2028,L3 refers to Sect. 2.1.1, but no such section exists.

Correct reference is 2.2 – this has been corrected.

9. P2077,L5 how and when is W as ventilation used and estimated?

We have clarified this in the text:

Page 2027, line 5

Ventilation is parameterised by using a large European data set from Seedorf et al. (1998a, 1998b). The derivation is fully described in Gyldenkærne et al. (2005) and uses outside temperatures and management practice in open and closed barns.

10. P2028,L11. I found these scenarios and their explanation confusing. Usually one begins to explain the 1st scenario and then develop explanations for the following ones. Here the authors begin with the last. And as noted by referee #3, the names change at different points in the paper. I miss also an explanation of the motives and thinking behind NOFERT. Please itemize better and explain each scenario, and then stick to the chosen naming convention throughout. As a minor point, it seemed odd to put scenario 3 (FLAT) in the middle of the non-WRF scenarios.

We agree with the Reviewer. We have changed the order of scenarios and keep it clear throughout the text (changed all figures and tables related to the scenarios). We have clarified the definition of the scenarios. Please see the modifications given below.

Page 2025, line 10-18

With this we will compare a constant emission approach (FLAT, scenario 1) against: 2) a dynamic approach based on the European-wide default settings (Skjøth et al., 2011, scenario DEFAULT), 3) a dynamic approach that takes into account Polish practice and less regulation compared to Denmark (POLREGUL), 4) a scenario that focuses on emissions from agricultural buildings (NOFERT). We will test all four scenarios for a full year with a simplified chemical transport model (CTM) in order to minimize the computational penalty and discuss the results from our four scenarios against related results that have been obtained for Denmark (Skjøth et al., 2011), Germany (Skjøth et al., 2011) and France (Hamaoui-Laguel et al., 2014).

Page 2028, line 10

The annual gridded NH₃ emissions were then used to construct 4 scenarios termed FLAT (1), DEFAULT (2), POLREGUL (3) and NOFERT (4) (Table 2). Applying the scenarios DEFAULT and FLAT shows the advantage of implementation of the dynamic emission model (DEFAULT) instead of using a constant emission profile (FLAT). This step is especially important for the area of Poland, as the dynamic approach at high spatial and temporal resolution has not been used before and because Poland is a large country where the variations in the climate cause changes in crop growth throughout the country, thereby affecting agricultural activity. Then, by replacing the default setup in the dynamic model with Polish regulations (POLREGUL) we wanted to provide some outlines for the users of this or similar models concerning the expected range of changes in ammonia emission.

This is considered particularly important due to the expanding use of this open-source model. These differences in emissions are caused by variations in agricultural practice in different countries, which are caused by both climate (thus affecting agricultural activity) and national regulations. A detailed description of the POLREGUL approach is provided below. In the fourth scenario (NOFERT) we wanted to show the sensitivity of the dynamic model to application of manure and fertilizers, mainly in respect of spring ammonia emission peak, thereby demonstrating that the implementation of the method should carefully assess national regulations on manure application for optimal performance of the model.

11. P2031,L18. What are 'specific' geographical areas.

Specific geographical area concerns location of stations listed in the bracket. We have modified the text to make it more clear:

Page 2031, line 18-19

Three of these EMEP stations are located in specific geographical areas, e.g. sea coast in the north (Łeba), the highest peak in the Sudety Mountains (Śnieżka), and a large forestland in NE Poland (Diabla Góra).

12. P2032,L12. Why 250m and 750m?

We have explained this in the text.

Page 2032, line 11-13

6 trajectories were run for each day with an episode from group 1, once every 6 hours. The trajectories were run for the receiving heights of 250 m and 750 m, as it was suggested by Hernández-Ceballos et al. (2014) that trajectories between 300 and 700 m do not show large differences in transport path within the first 12-24 hours.

13. P2038,L18. I assume you mean dissociation, not evaporation? You should give a reference for that process also (eg Fowler et al, 2009 for a recent review).

We meant evaporation, here. It is explained below:

Page 2038, line 17-19

Another factor that can cause an increase of ammonia concentrations within a plant canopy coupled with altered microclimate could be evaporation of ammonium containing aerosol (Fowler et al., 2009; Nemitz et al., 2004).

14. P2056, Fig. 3. The legend gives function names, but the axis says emissions. These are different things. Also, the yellow Fct10 line is very hard to see in my copy. Different line styles, bolder, and maybe some markers would help.

We have clarified in the figure caption that description in the legend concerns emission from given functions. We have improved the figure.

Page 2056, figure 3 caption

Fig 3. Time series of the seasonal variation in emission (POLREGUL run) for various agricultural emission categories in Jarczew. Description in the legend concerns emission from functions (Fct) described in Table1.

15. P2057, Fig. 4. Why compare one day's 3 hour period of emission with a monthly mean from FRAME? Compare like with like.

We agree with the comment. The emission has been aggregated into monthly values.

16. P2060, Fig 7. Which scenario is this - be explicit in the captions.

Clarified in the caption:

Fig 7. Modelled emission (POLREGUL) and measured concentration for the Jarczew station

17. P2061, Fig 8. It would be easier to see the trajectories with bolder lines. Also, are these 250m or 750m trajectories.

We have have changed the line style to bold. These are 250 m (upper row) and 750 m (lower row) – we have marked this in upper-right corner.

References:

- Andersen, H. V, Hovmand, M. F., Hummelshøj, P. and Jensen, N. O.: Measurements of ammonia concentrations, fluxes and dry deposition velocities to a spruce forest 1991-1995, *Atmos. Environ.*, 33(9), 1367–1383, 1999.
- Bouwman, A. F., Lee, D. S., Asman, W. A. H., Dentener, F. J., Van Der Hoek, K. W. and Olivier, J. G. J.: A global high-resolution emission inventory for ammonia, *Global Biogeochem. Cycles*, 11(4), 561–587, doi:10.1029/97GB02266, 1997.
- Fowler, D., Pilegaard, K., Sutton, M. A., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., Schjoerring, J. K., Granier, C., Neftel, A., Isaksen, I. S. A., Laj, P., Maione, M., Monks, P. S., Burkhardt, J., Daemmgen, U., Neiryneck, J., Personne, E., Wichink-Kruit, R., Butterbach-Bahl, K., Flechard, C., Tuovinen, J. P., Coyle, M., Gerosa, G., Loubet, B., Altimir, N., Gruenhage, L., Ammann, C., Cieslik, S., Paoletti, E., Mikkelsen, T. N., Ro-Poulsen, H., Cellier, P., Cape, J. N., Horváth, L., Loreto, F., Niinemets, Ü., Palmer, P. I., Rinne, J., Misztal, P., Nemitz, E., Nilsson, D., Pryor, S., Gallagher, M. W., Vesala, T., Skiba, U., Brüggemann, N., Zechmeister-Boltenstern, S., Williams, J., O'Dowd, C., Facchini, M. C., de Leeuw, G., Flossman, A., Chaumerliac, N. and Erisman, J. W.: Atmospheric composition change: Ecosystems–Atmosphere interactions, *Atmos. Environ.*, 43(33), 5193–5267, doi:10.1016/j.atmosenv.2009.07.068, 2009.
- Gyldenkerne, S., Skjøth, C. A., Hertel, O. and Ellermann, T.: A dynamical ammonia emission parameterization for use in air pollution models, *J. Geophys. Res.*, 110(D7), D07108, doi:10.1029/2004JD005459, 2005.

- Hamaoui-Laguel, L., Meleux, F., Beekmann, M., Bessagnet, B., Générumont, S., Cellier, P. and Létinois, L.: Improving ammonia emissions in air quality modelling for France, *Atmos. Environ.*, 92, 584–595, doi:10.1016/j.atmosenv.2012.08.002, 2014.
- Hansen, K., Sørensen, L. L., Hertel, O., Geels, C., Skjøth, C. A., Jensen, B. and Boegh, E.: Ammonia emissions from deciduous forest after leaf fall, *Biogeosciences*, 10(7), 4577–4589, doi:10.5194/bg-10-4577-2013, 2013.
- Hernández-Ceballos, M. A., Skjøth, C. A., García-Mozo, H., Bolívar, J. P. and Galán, C.: Improvement in the accuracy of back trajectories using WRF to identify pollen sources in southern Iberian Peninsula, *Int. J. Biometeorol.*, 58, 2031–2043, doi:10.1007/s00484-014-0804-x, 2014.
- Nemitz, E., Sutton, M. A., Wyers, G. P., Otjes, R. P., Mennen, M. G., van Putten, E. M. and Gallagher, M. W.: Gas-particle interactions above a Dutch heathland: II. Concentrations and surface exchange fluxes of atmospheric particles, *Atmos. Chem. Phys. Discuss.*, 4, 1519–1565, doi:10.5194/acpd-4-1519-2004, 2004.
- Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K. and Henze, D. K.: Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3), *J. Geophys. Res. Atmos.*, 119(7), 4343–4364, doi:10.1002/2013JD021130, 2014.
- Riddick, S. N., Blackall, T. D., Dragosits, U., Daunt, F., Braban, C. F., Tang, Y. S., MacFarlane, W., Taylor, S., Wanless, S. and Sutton, M. A.: Measurement of ammonia emissions from tropical seabird colonies, *Atmos. Environ.*, 89, 35–42, doi:10.1016/j.atmosenv.2014.02.012, 2014.
- Seedorf, J., Hartung, J., Schröder, M., Linkert, K. H., Pedersen, S., Takai, H., Johnsen, J. O., Metz, J. H. M., Groot Koerkamp, P. W. G., Uenk, G. H., Phillips, V. R., Holden, M. R., Sneath, R. W., Short, J. L. L., White, R. P. and Wathes, C. M.: A Survey of Ventilation Rates in Livestock Buildings in Northern Europe, *J. Agric. Eng. Res.*, 70(1), 39–47, doi:10.1006/jaer.1997.0274, 1998a.
- Seedorf, J., Hartung, J., Schröder, M., Linkert, K. H., Pedersen, S., Takai, H., Johnsen, J. O., Metz, J. H. M., Groot Koerkamp, P. W. G., Uenk, G. H., Phillips, V. R., Holden, M. R., Sneath, R. W., Short, J. L., White, R. P. and Wathes, C. M.: Temperature and Moisture Conditions in Livestock Buildings in Northern Europe, *J. Agric. Eng. Res.*, 70(1), 49–57, doi:10.1006/jaer.1997.0284, 1998b.
- Simpson, D., Winiwarter, W., Borjesson, G., Cinderby, S., Ferreiro, A., Guenther, A., Hewitt, C. N., Janson, R., Khalil, M. A. K., Owen, S., Pierce, T. E. and Puxbaum, H.: Inventorying emissions from nature in Europe, *J. Geophys. Res.*, 104(98), 8113–8152, 1999.
- 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.: Spatial and temporal variations in ammonia emissions – a freely accessible model code for Europe, *Atmos. Chem. Phys.*, 11(11), 5221–5236, doi:10.5194/acp-11-5221-2011, 2011.
- Sutton, M. A., Nemitz, E., Theobald, M. R., Milford, C., Dorsey, J. R., Gallagher, M. W., Hensen, A., Jongejan, P. A. C., Erisman, J. W., Mattsson, M., Schjoerring, J. K., Cellier, P., Loubet, B., Roche, R., Neftel, A., Hermann, B., Jones, S. K., Lehman, B. E., Horvath, L., Weidinger, T., Rajkai, K., Burkhardt, J., Löpmeier, F. J. and Daemmgen, U.: Dynamics of ammonia exchange with cut grassland: strategy and implementation of the GRAMINAE Integrated Experiment, *Biogeosciences*, 6(3), 309–331, doi:10.5194/bg-6-309-2009, 2009.

- Sutton, M. A., Perthue, E., Fowler, D., Storeton-West, R. L., Cape, J. N., Arends, G. G. and Mols, J. J.: Vertical distribution and fluxes of ammonia at Great Dun Fell, , 31(16), doi:10.1016/S1352-2310(96)00180-X, 1997.
- Theobald, M. R., Bealey, W. J., Tang, Y. S., Vallejo, A. and Sutton, M. A.: A simple model for screening the local impacts of atmospheric ammonia., *Sci. Total Environ.*, 407(23), 6024–33, doi:10.1016/j.scitotenv.2009.08.025, 2009.