

We are grateful for the constructive comments from the Reviewer. We have addressed all the comments and questions raised by the Reviewer 3. 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. My main concern is the strong conclusions made in the paper which appear to be based on two measurement stations. The paper concludes that the performance is much better based on two locations, where the other 3 locations do not show improvement. Based on the time series I see only improvement in Rzecin. Hence, it seems that the evidence for an improved modelling of the ammonia budget over Poland are indicative.

We agree that the best improvement is for Rzecin, which is seen both in the time series (Fig. 6) and in the statistics (Table 5) but there is also a general improvement for the POLREGUL run in comparison to DEFAULT. FLAT simulation provides low MAE for all sites but simultaneously the results have poor correlation with the observations. If we consider both statistics MAE and R, the results from POLREGUL are better than from DEFAULT for 4 stations (instead of Diabla Góra) and the correlation coefficient for POLREGUL is better for 4 stations in comparison to FLAT. The modelling setup we used here was 1) one year of simulation with the meteorological model WRF; 2) several emission scenarios with the atmospheric transport model FRAME. This setup enables us to get an overview of three aspects: a) what is the advantage of using dynamic emission instead of constant emission, b) what is the advantage of implementation of national practice into the dynamic emission model and c) what is the sensitivity of the model on application of manure and mineral fertilizer.

We agree with the Reviewer that the conclusions provided in the text are too strong. Based on this we have modified following paragraph in the Discussion section:

Page 2037 line 19 – page 2038 line 4

Text after changes:

The monthly correlation coefficients obtained with the FRAME model for the agricultural sites are comparable to the model results that are obtained with both DEHM (Skjøth et al., 2011) and the DAMOS system (Geels et al., 2012). Application of Polish practice into the ammonia dynamic model improves the FRAME results in comparison to the European default settings of the dynamic model. This suggests that similar improvements can be obtained for other European areas. For Polish conditions, with lack of detailed information about location of the agricultural fields and the location, amount and type of livestock, a higher mean absolute error for the dynamic simulations is observed in comparison to the constant emission approach. This also suggests that spatial allocation of emission might have a greater influence on concentration results obtained from a dynamic than from a constant emission approach.

2. In addition, I think the motivation and discussion on the use of the simplified chemistry transport model needs some more attention as the validation shows that the stations are not really located in source areas. Are the assumptions of the simplified chemistry warranted? Frame was ran on a monthly time resolution. What does this mean for the ammonia emissions? Is part of the connection

between meteorological dependent ammonia emissions and meteorological dependent fate in the atmosphere lost due to this set-up?

We agree with the Reviewer that it is appropriate with a more in-depth discussion of the impact of FRAME. In our opinion the simplified chemistry model warranted in this case. We have used the model in connection with monthly mean inputs and not episodes. The emission input is therefore the mean emission during the actual month, based on hourly meteorological dependent emissions. This will mean that the part of meteorological dependent emission is not lost. Similarly, the observations of ammonia concentrations are on a monthly basis. The used chemistry-transport model will mean that the part of the meteorological dependent fate of ammonia emissions can be too simple. However, similar principles to FRAME are present in local scale models like OML (Geels et al., 2012) and OPS (Van Jaarsveld, 2004; Velders et al., 2011). It is shown with these models that in relation to ammonia and on spatial scales of 0.5-16 km it is sufficient to neglect chemical transformation and wet deposition even on a daily and weekly basis (Geels et al., 2012). OML and FRAME use similar principles on the near source domain. In relation to ammonia and the fate due to chemical conversion and wet deposition, then the FRAME methodology is more advanced than the OML method. The FRAME model does represent the important chemical reactions for ammonia (reaction with HNO_3 and H_2SO_4) as well loss through both wet and dry deposition (please see expanded description given below). As an example, OML does not include chemical conversion or wet deposition. Still the annual correlation coefficients are high (0.7-0.75) and the bias is low when OML is compared with observations. This shows, that the governing processes on ammonia on this scale is due to emissions and initial dispersion and only to a small degree conversion and deposition (dry and wet) and fully corresponds with the two latest reviews on this subject (Hertel et al., 2006, 2012).

To make this more clear we have included a new text:

Page 2037 line 16 (after "... a large computational overhead.")

Similar principles to FRAME are present in local scale models like OML (Geels et al., 2012) and OPS (Van Jaarsveld, 2004; Velders et al., 2011). It is shown with these models that in relation to ammonia and on spatial scales of 0.5-16 km it is sufficient to neglect chemical transformation and wet deposition even on daily and weekly basis (Geels et al., 2012). OML and FRAME use similar principles on the near source domain. In relation to ammonia and the fate due to chemical conversion and wet deposition, the FRAME methodology is more advanced than the OML method. Despite the OML model does not include chemical conversion or wet deposition, the annual correlation coefficients are high (0.7-0.75) and the bias is low, when compared with observations. This shows that the governing processes on ammonia concentrations on this scale are due to emissions and dispersion within the agricultural areas and only to a small degree conversion and deposition (dry and wet). These results correspond well with the two latest reviews on this subject (Hertel et al., 2006, 2012).

We have provided additional information on processes implemented in the FRAME model:

Page 2030, line 28 (after "...and frequency roses.")

Vertical diffusion of gaseous and particulate species is described with K-theory eddy diffusivity, and solved with the Finite Volume Method. The FRAME model chemistry scheme is similar to the one

used in the EMEP Lagrangian model (Barrett and Seland, 1995). The prognostic chemical variables calculated in FRAME are: NH_3 , NO , NO_2 , HNO_3 , PAN, SO_2 , H_2SO_4 , as well as NH_4^+ , NO_3^- and SO_4^- aerosol. NH_4NO_3 aerosol is formed by the equilibrium reaction between HNO_3 and NH_3 . A second category of large nitrate aerosol is presented and simulates the deposition of nitric acid on to soil dust or marine aerosol. The formation of H_2SO_4 by gas phase oxidation of SO_2 is presented by a predefined oxidation rate. H_2SO_4 then reacts with NH_3 to form ammonium sulphate aerosol. The aqueous reactions considered in the model include the oxidation of S(IV) by O_3 , H_2O_2 and the metal catalysed reaction with O_2 .

Dry deposition of SO_2 , NO_2 and NH_3 is calculated individually for five different land cover categories (arable, forest, moor-land, grassland and urban) using a canopy resistance model (Singles et al., 1998). Wet deposition is calculated with scavenging coefficients and a constant drizzle approach, using precipitation rates calculated from a map of average annual precipitation. An increased washout rate is assumed over hill areas due to the seeder-feeder effect. It is assumed that the washout rate for the orographic component of rainfall due to the seeder-feeder effect is twice that used for the non-orographic components (Dore et al., 1992).

3. The definition of the scenarios runs is not consistent throughout the paper. And sometimes 3 or 4 scenarios are mentioned. - Default (Skjøth et al. 2011) - No application emissions - Is the existing emission method in WRF-CHEM a constant emission over time as it is termed FLAT in section 2.2? If so, this is not common practice in European chemistry transport models. I would call it “constant emissions” - Polish regulation and practice: Often regulation is mentioned but practice could be a better word for this simulation.

We agree with the Reviewer. We clarified the definition of the scenarios throughout the text. We removed the reference to WRF-Chem where it was used in the general context of the constant emissions. The “the existing method used in WRF-Chem” is the same as “flat emission” and it was unified in the text. The “flat emission” term was change to “constant emission”. “Polish regulation and practice” term was changed to “Polish practice”.

We have modified the text:

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).

4. In our modelling system we found that the change in diurnal cycle of the emissions can induce large changes in modelled annual mean ammonia levels (using the same emission total). You have

changed both the day to day variability as the diurnal cycle. Do you have an idea how much this effects your results?

FRAME is not sensitive to this kind of variation as it by definition calculates monthly mean values. This kind of experiment requires a more advanced atmospheric transport model like the DEHM (Brandt et al., 2012), LOTUS-EUROS (Mues et al., 2014) or WRF-Chem modelling systems (Grell et al., 2005). Having said this, the emission model alone can also have an impact on the total emission and thus also on the annual mean ammonia levels.

Specific comments:

5. P2020,L8-9 It is stated the model is robust with respect to stable and storage emissions. What do you mean?

Our scenario run without fertilizer shows that the model output is very sensitive to the timing of manure application. Additionally, the calibration of the temperature functions that are used inside buildings are used on a data-rich and European-wide data set by Seedorf et al. (1998a, 1998b).

6. P2026, L13: Default values for the contribution of the total ammonia emission to each activity i.

Changed.

7. P2026, L23: In equation 1 and 2 I miss the consequent use of the index for the hour/time of the year. The explanation of the equations in the lines below is not really understandable without the original publication. Please provide the calculation of E_{pot} as well.

The explanation of the equations has been expanded:

Page 2027, lines 15-22

Here, μ_i is the mean value for the parameterized distribution. This means that μ_i (given in days or hours) corresponds to the time of the year when the Gaussian function obtains its maximum value. This is the optimal time for the farmer to apply manure according to crop growth. Therefore, the value of μ_i depends on the results from the crop growth model which vary from cell to cell over the entire model grid. σ_i is the spread of the Gauss function, which here parameterizes the amount of time that all farmers carry out this specific activity in each grid cell. A large σ_i means that the emission from the corresponding activity takes place during most of the year, while a small σ_i means that emission takes place during a few weeks. Here t is the actual time of the year. The temperature correction T_{corr} and the emission potential $E_{pot_i}(x,y)$ (calculated in the preprocessing) is given in eq. (3).

$$T_{corr} = e^{(0.0223*t(x,y))} \quad \text{for } i= 8, 9, 10, 11, 12, 13$$

$$T_{corr} = 1 \quad \text{otherwise}$$

$$E_{pot_i}(x,y) \neq 1 \quad \text{for } i= 8, 9, 10, 11, 12, 13$$

$$E_{pot_i}(x,y) = 1 \quad \text{otherwise}$$

8. P2027, L3: refer to section 2.2

Changed.

9. P2028, L6: I assume from the text that all fields in a province get the same amounts of fertilizer and manure. Or is the manure application performed per commune? The provinces are rather large. Do you think this affects the results?

Yes, all the fields in a province get the same amount of fertilizer and manure. This simplified information is used because of data availability.

10. P2029, L10 Are the Poland default settings in Table 2 consistent with the Polish emission inventory?

Yes, the sum of dynamic ammonia emission from DEFAULT is consistent with the Polish emission inventory.

11. P2033 L17: Figure 3 shows large emissions for FKt(15) although this emission source accounts only 1% of the annual emission total. Please explain. Is Jarczew the best location to show this plot for?

We are sorry; we have uploaded a wrong figure. Large emission on Jarczew station in summer is related to application of manure (Fct 10), not to ammonia treated straw as it was showed in the previous (incorrect) figure. The location was selected because an air quality station working within the EMEP network is operating there.

12. P2034, L6: In the description of Figure 5 it is mentioned that there is a large variability between day and night. This variability is only 25 %. I would remove the word large and insert the quantification. 25% is rather small compared to traditional estimates in variability as commonly used in other modelling studies.

We agree. The sentence has been changed:

Due to diurnal variability in air temperature and wind speed there is a day-night variation in emission. The mean for the entire year diurnal variation is equal to 20% (Wrocław) - 25% (Leszno), with the lowest values during winter (about 10%) and highest in spring and summer (about 30%).

13. P2034, L20 is much higher than

Corrected.

14. P2035. The discussion in 3.3 and the figure highlights the need for hour-by-hour calculations.

We agree that hour-by-hour calculations are relevant and are state of the art. Please see a reply to the comment 1, where we explained the reason we used the setup presented in this study.

15. P2037. L17-29. The conclusions here are based on two sites that compare favorably, whereas the other sites seem to say something different. The evaluation at more remote locations seem to show that in Poland the atmospheric transport and transformation are important processes. The conclusion that it is only emission driven seems not warranted. In my opinion it is not possible to conclude for Poland that this study obtained as good results as for Denmark with DEHM and

DAMOS. The evaluation basis is completely different to support this statement. The study is a step forward in ammonia modelling over Poland, but maybe these statements are a bit too enthusiastic.

We agree with the Reviewer that the statements were too enthusiastic. We have provided several modifications to the text (as given in reply to comment 1 and 2). Modifications in the text are recalled below.

Page 2037 line 19 – page 2038 line 4

Text after changes:

The monthly correlation coefficients obtained with the FRAME model for the agricultural sites are comparable to the model results that are obtained with both DEHM (Skjøth et al., 2011) and the DAMOS system (Geels et al., 2012). Application of Polish practice into the ammonia dynamic model improves the FRAME results in comparison to the European default settings of the dynamic model. This suggests that similar improvements can be obtained for other European areas. For Polish conditions, with lack of detailed information about location of the agricultural fields and the location, amount and type of livestock, a higher mean absolute error for the dynamic simulations is observed in comparison to the constant emission approach. This also suggests that spatial allocation of emission might have a greater influence on concentration results obtained from a dynamic than from a constant emission approach.

Page 2037 line 16 (after “... a large computational overhead.”)

Similar principles to FRAME are present in local scale models like OML (Geels et al., 2012) and OPS (Van Jaarsveld, 2004, Velders et al. 2011). It is shown with these models that in relation to ammonia and on spatial scales of 0.5-16 km it is sufficient to neglect chemical transformation and wet deposition even on daily and weekly basis (Geels et al., 2012). OML and FRAME use similar principles on the near source domain. In relation to ammonia and the fate due to chemical conversion and wet deposition, the FRAME methodology is more advanced than the OML method. Despite the OML model does not include chemical conversion or wet deposition, the annual correlation coefficients are high (0.7-0.75) and the bias is low, when compared with observations. This shows that the governing processes on ammonia concentrations on this scale are due to emissions and dispersion within the agricultural areas and only to a small degree conversion and deposition (dry and wet). These results correspond well with the two latest reviews on this subject (Hertel et al., 2013, Hertel et al., 2006).

16. P.2038. The results for Diabla Gora show clearly that the our understanding or modelling approach is not sufficient to explain the measured concentrations. Assuming that in Poland temperatures are well below zero and snow cover and frozen open water are often present I wonder if the presented explanation is more than speculation. Are these conditions represented well in the model system? Why are so many references being made to WRF-CHEM? There are more models available to study this issues on higher temporal resolutions that are further concerning ammonia modelling and easier to handle.

We agree with the comment. The meteorological conditions were based on the WRF model and also observations data from 210 rainfall sites in Poland. The meteorological data suggest that the natural emission could take place over the Diabla Góra station up to the late autumn, because after that

period (December and January) mean daily temperatures are below zero. The long-term evaluation of the WRF model (Kryza et al., 2015) shows, that the model resolves these conditions well.

The text has been modified:

Page 2038, line 16

Natural emission could explain the high ammonia concentrations at the Diabla Góra station in autumn and late autumn (until beginning of December), when mean daily temperature is above zero and no snow cover present. Based on the emission and measurements data as well as model results it is difficult to explain the high ammonia concentrations in the mid-winter period. These could be more efficiently studied with chemistry transport models which are connected online with both meteorology like e.g. GATOR-MMTD (Jacobson et al., 1996), WRF-Chem (Grell et al., 2005), GEM-AQ (Kaminski et al., 2007), and a dynamic ammonia emission model.

We have changed the reference from WRF-CHEM to general on-line coupled meteorology and chemistry models in the entire text.

17. A multi-year simulation which is easily performed with FRAME could have made a stronger case.

A multi-year simulation would also require simulations with the dynamic ammonia emission model and the WRF meteorological model. With the approach used in this study, we have minimised the calculation costs by running FRAME for the same metrological conditions (one year) but showed the importance of application of different emission approaches and implementation of national practice to ammonia model. The results show that the dynamic approach to ammonia emission is important for this area, therefore we propose further studies to apply the dynamic emission in a more complex atmospheric transport model and for a longer study period.

References:

- Barrett, K. and Seland, O.: European Transboundary Acidifying Air Pollution: Ten Years Calculated Fields and Budgets to the End of the First Sulphur Protocol, European transboundary air pollution report, 1- 150,1995.
- Brandt, J., Silver, J. D., Frohn, L. M., Geels, C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Skjøth, C. A., Villadsen, H., Zare, A. and Christensen, J. H.: 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, 2012.
- Dore, A. J., Choularton, T. W. and Fowler, D.: An improved wet deposition map of the United Kingdom incorporating the seeder-feeder effect over mountainous terrain., *Atmos. Environ.*, doi:10.1016/0960-1686(92)90122-2, 1992.
- Geels, C., Andersen, H. V., Ambelas Skjøth, C., Christensen, J. H., Ellermann, T., Løfstrøm, P., Gyldenkerne, S., Brandt, J., Hansen, K. M., Frohn, L. M. and Hertel, O.: Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS, *Biogeosciences*, 9(7), 2625–2647, doi:10.5194/bg-9-2625-2012, 2012.

- Grell, G., Peckham, S. E., Schmitz, R., McKeen, S., Frost, G., Skamarock, W. C. and Eder, B.: Fully coupled “online” chemistry within the WRF model, *Atmos. Environ.*, 39(37), 6957–6975, doi:10.1016/j.atmosenv.2005.04.027, 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.
- Hertel, O., Skjøth, C. A., Løfstrøm, P., Geels, C., Frohn, L. M., Ellermann, T. and Madsen, P. V.: Modelling Nitrogen Deposition on a Local Scale—A Review of the Current State of the Art, *Environ. Chem.*, 3(5), 317, doi:10.1071/EN06038, 2006.
- Hertel, O., Skjøth, C. A., Reis, S., Bleeker, A., Harrison, R. M., Cape, J. N., Fowler, D., Skiba, U., Simpson, D., Jickells, T., Kulmala, M., Gyldenkerne, S., Sørensen, L. L., Erisman, J. W. and Sutton, M. A.: Governing processes for reactive nitrogen compounds in the European atmosphere, *Biogeosciences*, 9(12), 4921–4954, doi:10.5194/bg-9-4921-2012, 2012.
- Van Jaarsveld, H.: The Operational Priority Substances model: Description and validation of OPS-Pro 4.1., National Institute for Public Health & Environment, 1-156, 2004.
- Jacobson, M. Z., Lu, R., Turco, R. P. and Toon, O.: Development and application of a new air pollution model system e Part I: Gas-phase simulations, *Atmos. Env*, 30, 1939–1963, 1996.
- Kaminski, J., Neary, L., Lupu, A., McConnell, J., Struzewska, J., Zdunek, M. and Lobocki, L.: High Resolution Air Quality Simulations with MC2-AQ and GEM-AQ, in *Air Pollution Modeling and Its Application XVII SE - 86*, edited by C. Borrego and A.-L. Norman, pp. 714–720, Springer US., 2007.
- Kryza, M., Watzek, K., Ojrzyńska, H., Szymanowski, M., Werner, M. and Dore, A. J.: High resolution dynamical downscaling of ERA-Interim using the WRF regional climate model (Part 1) – model configuration and statistical evaluation for the 1981-2010 period, *Pure Appl. Geophys.*, In revision, 2015.
- Mues, A., Kuenen, J., Hendriks, C., Manders, A., Segers, A., Scholz, Y., Hueglin, C., Builtjes, P. and Schaap, M.: Sensitivity of air pollution simulations with LOTOS-EUROS to the temporal distribution of anthropogenic emissions, *Atmos. Chem. Phys.*, 14(2), 939–955, doi:10.5194/acp-14-939-2014, 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.
- Singles, R., Sutton, M. A. and Weston, K. J.: A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain, *Atmos. Environ.*, 32(3), 393–399, doi:10.1016/S1352-2310(97)83467-X, 1998.
- 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.

Velders, G. J. M., Snijder, A. and Hoogerbrugge, R.: Recent decreases in observed atmospheric concentrations of SO₂ in the Netherlands in line with emission reductions, *Atmos. Environ.*, 45(31), 5647–5651, doi:10.1016/j.atmosenv.2011.07.009, 2011.