We are grateful for the constructive comments from the Reviewers. We have addressed all the comments and questions. 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.

## Reviewer 1

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 3.

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 forseeable) future?

   We do not expect detailed data on fertilizer practices, irrigation and so on in the near feature 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 $NH_3$ 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 (European Environment Agency 2014, www.eea.europa.eu) and is largely from animal excreta and fertilizers. The contribution of natural emission is negligible compared to agricultural for the most European area (Simpson et al., 1999; Friedrich 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 elswhere. 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 Skjoth?)

   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 $NH_3$ 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 spatial 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.

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## Reviewer 2

Some attention to the English is required throughout – I have not identified all errors in the specific comments below so suggest a final revision by a native English speaker.

The language has been carefully checked by a native speaker.

Specific comments:

P1 L15 Define CTM

Defined.

P1 L22 Define NWP

Defined.

P1 L30 change 'was compared' to 'were compared'

Changed.

P4 L1 What is the WRF model (and subsequently WRF-Chem and WRF-ARW)?

WRF (Weather Research and Forecasting model) and WRF-ARW (The Advanced Research WRF) was used interchangeably in the text. We have clarified this and now only WRF is used. WRF-Chem is a chemical transport model (WRF coupled with chemistry). Reference to WRF-Chem was removed from this sentence.

Modified text:

Page 2025, lines 8-11

We will connect the model directly with the meteorological calculations from the Weather Research and Forecasting model (WRF, Skamarock and Klemp, 2008) according to the vision of Sutton et al. (2013).

P5 L5 'stables' is often used as a term for livestock housing by non-native English speakers in Europe; however, in English, stables is normally understood to refer specifically to housing for horses. Please change the term here and elsewhere in the manuscript to 'livestock housing' or similar.

The term was changed to “livestock housing”.

P5 L7 What are the units for the various parameters in Eq. 1 (and 2 on following page). I have to admit to not fully understanding the subsequent description of the derivation of the functions – is the function an emission value itself, or a multiplier to be applied to the emission input data. Perhaps this description could be expanded slightly to aid understanding.

The units have been explained:

$E_i(x,y)$ [kg ha$^{-1}$ year$^{-1}$]

$Epot_i(x,y)$ [unitless]

$T_i(x,y)$ [ºC]

$W_i(x,y)$ [m s$^{-1}$]

The description has been expanded for clarity:

Page 2026, line 11; new text:

The individual functions are distributed into two groups: Gaussian functions for short term emission sources and annual functions. Both groups respond to the environmental variables wind speed and temperature. The Gaussian functions are linked to a crop growth model developed by Olesen & Plauborg (1995). The crop growth model uses accumulated temperature sums to determine the timing of the maximum value of the individual gauss functions.

2027, lines 15-17 (modified and expanded text)

Here, $\mu_i$ is the mean value for the parameterized distribution. This means that $\mu_i$ (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 $\mu_i$ depends on the results from the crop growth model which vary from cell to cell over the entire model grid. $\sigma_i$ 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 $\sigma_i$ means that the emission from the corresponding activity takes place during most of the year, while a small $\sigma_i$ means that emission takes place during a few weeks. Here *t* is the actual time of the year. The temperature correction Tcorr and the emission potential $Epot_i(x,y)$ (calculated in the preprocessing) is given in eq. (3).

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

$T_{corr} = 1$ otherwise

$Epot_i(x,y) \neq 1$ for i= 8, 9, 10, 11, 12, 13

$Epot_i(x,y) = 1$ otherwise

P6-7 It would be good to include some introduction as to why these 4 specific scenarios are being modelled.

We agree with the Reviewer. We have included some introduction (please see below). We have also changed the order of scenarios as it was suggested by Reviewer 1 (It concerns all figures and tables related to the scenarios).

Page 2028, line 10

The annual gridded $NH_3$ 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 spatial 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 in respect 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.

P7 L8 It is not clear here whether you mean 20% of all manure, which equates with all slurry, or 20% of slurry bein applied to grassland. Please clarify.

We have clarified the sentence:

Page 2028, lines 17-19

In Poland the solid and slurry fractions of the manure is applied differently due to national regulations. Solid manure goes into annual crops as only slurry is allowed on grasslands. Between 10% and 20% of the slurry fraction is applied to grassland, which covers about 25% of the entire agricultural area.

P11 L9-10 Values are presented for the grid square (5x5km) containing the Jarczew station?

Yes, values are presented for the grid square. It has been clarified in the text:

Page 2033, line 12-13

The seasonal variation of emission (POLREGUL run) for different agricultural categories for the grid representing Jarczew station is shown in Fig. 3.

P11 L15-17 Does this sentence apply generally for Poland or specifically for this grid square containing the Jarczew station? If it is a general statement for Poland, can anything be said about the spatial variation in large pig farms and cattle farming?

It is a general statement for Poland. It was clarified in the text. Pig and cattle farming in Poland is highly fragmented. There are many small farms in southern part of the country with a low number of cattle, between 2 and 10 (Litwińczuk and Grodzki, 2014). Dairy farms are, located in the north-eastern and central Poland, where the specialisation in milk production results in high animal numbers at each farm. Cattle kept for meat production are usually kept in herds of 25 and are farmed in north-eastern Poland. The highest concentration of pig farming as well as the largest farms are in central part of Poland (GUS, 2011).

P13 L11 I don't see any Fig. 8 – is it missing? And what is the RIP tool?

Figure 8 is attached in the Biogeosciences Discussion paper. Please see page 2061. RIP (which stands for Read/Interpolate/Plot) is a Fortran program used for visualizing output from gridded meteorological data sets.

We have expanded a description of the RIP tool:

Page 2032, lines 9-10

RIP version 4.5 (Stoelinga, 2009), which is a is a Fortran program used for visualizing output from gridded meteorological data sets, was implemented to get 36 h backward trajectories for the Jarczew station.

P14 L6-8 But data presented here show the opposite to what is being said in this sentence i.e. the data here show moving from the DEFAULT to POLREGUL gives a decrease in spring emissions.

We agree with the comment. We wanted to emphasize here the range of changes, which could be expected due to an application of national practice into the dynamic model. A scale and character of changes will vary between countries and depend on local infrastructure and practice.

We clarified it in the text:

Page 2036, line 27

The scale and character of changes between the POLREGUL and DEFAULT simulations with the dynamic ammonia model will vary between countries and depend on local agricultural infrastructure and practice.

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## Reviewer 3

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 no 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 for 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 chemical 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 a daily and weekly basis (Geels et al., 2012). OML and FRAME use similar principles for 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. Although 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 chemical 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 represented 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 (Skjoth 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 Epot as well.

   The explanation of the equations has been expanded:

   Page 2027, lines 15-22

   Here, $\mu_i$ is the mean value for the parameterized distribution. This means that $\mu_i$ (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 $\mu_i$ depends on the results from the crop growth model which vary from cell to cell over the entire model grid. $\sigma_i$ 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 $\sigma_i$ means that the emission from the corresponding activity takes place during most of the year, while a small $\sigma_i$ means that emission takes place during a few weeks. Here *t* is the actual time of the year. The temperature correction Tcorr and the emission potential $Epot_i(x,y)$ (calculated in the preprocessing) is given in eq. (3).

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

$T_{corr} = 1$ otherwise

$Epot_i(x,y) \neq 1$ for i= 8, 9, 10, 11, 12, 13

$Epot_i(x,y) = 1$ 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 enthousiastic.

    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 for 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. Although 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 chemical 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.

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# Understanding emissions of ammonia from buildings and application of fertilizers: an example from Poland

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## Abstract

A Europe-wide dynamic ammonia ($NH_3$) emissions model has been applied for one of the large agricultural countries in Europe, and its sensitivity on the distribution of emissions among different agricultural functions was analyzed by comparing with observed ammonia concentrations and by implementing all scenarios in a Chemical Transport Model (CTM). The results suggest that the dynamic emission model is most sensitive to emission from animal manure, in particular how animal manure and its application on fields is connected to national regulations. To incorporate the national regulations, we obtained activity information on agricultural operations at the sub-national level for Poland, information about infrastructure on storages and current regulations on manure practice from Polish authorities. The information was implemented in the existing emission model and was connected directly with ~~the NWP~~ calculations from the Weather Research and Forecasting model (WRF~~-ARW~~). The model was used to calculate four emission scenarios with high spatial (5 km x 5 km) and temporal resolution (3h) for the entire year 2010. In the four scenarios, we have compared ~~the European-wide default model settings against: 1) a scenario that focuses on emission from agricultural buildings, 2) the existing emission method used in WRF-Chem in Poland, and 3) a scenario that takes into account Polish infrastructure and agricultural regulations.~~ a constant emission approach (FLAT, scenario 1) against: 2) a dynamic approach based on the Europe-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). The ammonia

emission was implemented into the chemical transport model FRAME and modelled ammonia concentrations ~~was~~ were compared with measurements. The results for an agricultural area suggest that the default setting in the dynamic model is an improvement compared to a non-dynamical emission profile. The results also show that further improvements can be obtained at a~~on the~~ national scale by replacing the default information on manure practice with information that is connected with local practice and national regulations. ~~Implementing a dynamical approach for simulation of ammonia emission is a viable objective for all CTM models that continue to use fixed emission profiles.~~ 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 should handle ammonia emissions in a similar way to other climate dependent emissions (e.g. Biogenic Volatile Organic Compounds). Our results, compared with previous results from the DEHM and the GEOS-CHEM models, suggest that implementing dynamical approaches improves simulations in general even in areas with limited information about location of the agricultural fields, livestock and agricultural production methods such as Poland.~~

**Keywords:** $NH_3$, dynamic emission modelling, application of fertilizers, Poland

## 1 Introduction

~~Ammonia is emitted to the atmosphere, mainly from agricultural operations (Bouwman et al., 1997), but also from natural sources (Riddick et al., 2014).~~ 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 (European Environment Agency 2014, www.eea.europa.eu) and is largely from animal excreta and fertilizers. The contribution of natural emission is negligible compared to agricultural for the most European area (Simpson et al., 1999; Friedrich 2007). Ammonia is the main alkaline gas in the atmosphere (Hertel et al., 2012) and is responsible for neutralizing acids (sulphuric and nitric acid) formed through the oxidation of sulphur dioxide ($SO_2$) and nitrogen oxides ($NO_x$) (Seinfeld and Pandis, 2006). This leads to creation of ammonium ($NH_4^+$) salts, which are incorporated in atmospheric aerosols (Banzhaf et al., 2013; Reis et al., 2009). The emission of

$NH_3$ makes a major contribution to the formations of particulate matter, $PM_{10}$ and $PM_{2.5}$ (de Meij et al., 2009; Werner et al., 2014), accounting for up to 50% of the total mass of $PM_{2.5}$ (Anderson et al., 2003). As such, ammonia-containing aerosols are a very important component in regional and global aerosols processes (Xu and Penner, 2012). There is a direct climate penalty on ammonia emission (Skjøth and Geels, 2013), mainly because the volatilization potential of ammonia nearly doubles for every 5ºC temperature increase (Sutton et al., 2013). In the fifth report of IPCC, ammonia emission is highlighted as an important component with a considerable feedback effect on climate and air quality that remains to be understood (IPCC, September 2013). There is therefore a need to improve the descriptions of ammonia emission models and advance the level of input data to these models (Flechard et al., 2013; Guevara et al., 2013; Wichink Kruit et al., 2012) and correspondingly use them with chemistry transport models. Ideally, this improved approach should directly use results from climate or numeric weather prediction models (Sutton et al., 2013) because the fluxes of ammonia with the surface are directly and non-linear related to meteorology (Baklanov et al., 2014).

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). The two governing processes for nitrogen deposition are wet deposition of ammonium-containing aerosols and dry deposition of ammonia (Bash et al., 2013; Hertel et al., 2012). Ammonia also contributes to the eutrophication of terrestrial ecosystems and surface waters and the development of a lower tolerance to stress in woodland and forests (Sutton et al., 1998, 2009). This eutrophication leads to loss of plant diversity in a wide range of habitats (Emmett, 2007; Jones et al., 2011; Stevens et al., 2004). Nitrogen deposition exceeds the critical loads in most European countries, such as France (van Grinsven et al., 2012), the Netherlands (Jones et al., 2011), Belgium (Jones et al., 2011), Germany (Nagel and Gregor, 2001) and Poland (Hettelingh et al., 2009; Kryza et al., 2013a). The regions with the highest nitrogen deposition are the areas with intense agricultural production, high ammonia emission and corresponding high deposition of ammonia containing compounds (Hertel et al., 2012; Wichink Kruit et al., 2012). The calculation of maps of critical load exceedance require Chemical Transport Models (CTMs) to generate estimates of nitrogen deposition (Flechard et al., 2013). These exceedance maps generally require high spatial and temporal resolution in the atmospheric models (Geels et al., 2012; Mues et al., 2014) and it has been shown that this requires detailed information on emission from different agricultural operations (e.g. Skjøth et al., 2011). These operations also rely on national legislations on manure management (e.g. Gyldenkærne et al., 2005), regional husbandry methods (e.g. Skjøth et al., 2011), as well as prevailing crops and use of mineral fertilizer (Gyldenkærne et al., 2005; Misselbrook et al., 2006). This information can be obtained from agricultural databases in countries like Denmark (e.g. Gyldenkærne et al., 2005), the Netherlands (van Pul et al., 2008) and the UK (Hellsten et al., 2008), but has so far not been available in countries with substantial ammonia emissions such as France, Italy and Poland. Simplified approaches to agricultural production methods (activity data) have therefore been applied in existing models that aim at making Europe-scale calculations (Skjøth et al., 2011), which will decrease the quality of the results. It has therefore been highlighted that there is a need to obtain national and detailed activity data and integrate this information into models (Flechard et al., 2013).

The aim of this paper is to obtain activity information on agricultural operations at the subnational level for Poland, one of the largest agricultural countries in Europe, ~~Poland,~~ and implement these data in an existing ammonia emission model (Skjøth et al., 2004, 2011). We will connect the model directly with the meteorological calculations from the Weather Research and Forecasting model (WRF, Skamarock and Klemp, 2008) according to the suggestion of Sutton et al. (2013). ~~We will connect the model directly with the NWP calculations from the WRF model (Skamarock and Klemp, 2008) according to the vision of Sutton et al. (2013)~~. ~~on a model grid that is identical to the WRF-Chem model for Poland (Werner et al., 2014). With this we will compare the Europe-wide default settings (Skjøth et al., 2011) against: 1) a scenario that focuses on emissions from agricultural buildings 2) the existing method used in WRF-Chem over Poland and 3) and a scenario that takes into account Polish infrastructure and less regulation compared to Denmark. We will test all four scenarios for a full year with a simplified chemical transport model (CTM) in order to minimize the computational penalty with WRF-Chem 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).~~

With this we will compare a constant emission approach (FLAT, scenario 1) against: 2) a dynamic approach based on the Europe-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 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).

## 2 Methodology

### 2.1 Emission model

$NH_3$ emissions have been calculated with a dynamic model originally developed for Denmark. The fundamentals of the model are provided by Gyldenkærne (2005), Skjøth et al. (2004) and Skjøth et al. (2011). The general idea behind the emission model is to use the gridded annual total $NH_3$ emissions (data described in the next section) and to use available activity data to make a disaggregation of the gridded annual totals into specific agricultural sectors with a similar emission pattern. The emission from each sector then uses a parameterization that depends on both the volatilization as a function of meteorology and the temporal pattern of the activity. This creates a set of additive continuous emission functions, denoted as $Fct_i$, typically with a time resolution of 1 or 3 h. The methodology allows for either normalization to full agreement with national annual official emissions (Skjøth et al., 2011) or freely fluctuating emissions due to meteorology, where the freely fluctuating emissions can be either larger or smaller compared to official estimates (Skjøth and Geels, 2013). The emission parameterization consists of 16 additive continuous functions (Table 1), describing emission from animal houses and storage (3 functions), application of manure and mineral fertilizer (7 functions), emission from crops (4 functions), grazing animals, ammonia treatment of straw and road traffic. The individual functions are distributed into two groups: Gaussian functions for short term emission sources and annual functions. Both groups respond to the environmental variables wind speed and temperature. The Gaussian functions are linked to a crop growth model developed by Olesen & Plauborg (1995). The crop growth model uses accumulated temperature sums to determine the timing of the maximum value of the individual Gauss functions. The applied functions were originally derived for Danish conditions and presented in Skjøth et al. (2004) but Skjøth et al. (2011) suggest that a majority of the functions may be directly applicable for a large part of Europe. Default values were therefore implemented by Skjøth et al. (2011) for many European countries. Several of the underlying studies for producing parameterizations, such as the applied growth model (Olesen and Plauborg, 1995) and the farm surveys by Seedorf et al. (1998a, 1998b), are based on Europe-wide studies and are considered appropriate for large geographical regions (Skjøth et al., 2011), while the parameterizations for manure application may need adaptation to national regulation, which is known to change over time (Skjøth et al., 2008).

The functions for emission from ~~stables~~ livestock housing and manure storage are defined in Eq. (1), and the temporal profile of emission depends on air temperature and wind speed in a given grid cell:

$$Fct_i = \frac{E_i(x,y)}{Epot_i(x,y)} * (T_i(x,y))^{0.89} * (W_i(x,y))^{0.26} \quad i=[1;\,3]. \tag{1}$$

$E_i(x,y)$ [kg $ha^{-1}$ $year^{-1}$]

$Epot_i(x,y)$ [unitless]

$T_i(x,y)$ [ºC]

$W_i(x,y)$ [m $s^{-1}$]

Index i refers to functions 1-3 and x and y refer to the coordinate in the east-west direction and south-north direction. $Fct_1$ refers to animal houses with forced ventilation, $Fct_2$ refers to open animal houses, and $Fct_3$ to manure store. $E_i$(x, y) is the emission input into the model and $Epot_i$(x, y) is the emission potential scaling factor for a given grid cell. The emission potential is used to scale the annual emission up/down in accordance with the officially reported value. Input emission data for the Poland domain was obtained according to the procedure described in section ~~2.1.1~~2.2. $T_i$(x, y) is the temperature in either animal houses or at the surface of the manure storage, and W is either the ventilation inside the building or the 10 m wind speed above the storages. 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. The emission potential is approximated by the 2 m air temperature, provided by the WRF~~-ARW~~ model and a simple parameterization for temperatures and ventilation in livestock housing~~stable~~ systems (Gyldenkærne et al., 2005). The WRF~~-ARW~~ model configuration and evaluation is provided in the following sections.

Table 1

Functions $Fct_4$ – $Fct_{15}$ are related to plant growth and include emissions from plants and emissions due to applications of fertilizer and manure (Table 1). Functions 4 to 15 depend on both air temperature and wind speed. The temporal variations for these activities have therefore been parameterized by the Gauss functions (Eq. 2).

$$Fct_i = \left(W_{corr} * T_{corr} \frac{E_i(x,y)}{Epot_i(x,y)}\right) * \frac{e^{\left(\frac{(t-\mu_i(x,y))^2}{-2\sigma_i^2(x,y)}\right)}}{\sigma_i\sqrt{2\pi}} \qquad i = [4;\ 15] \qquad (2)$$

~~Here, $\mu_i$ is the mean value for the parameterized distribution understood as the time of the year when the Gauss function obtains its maximum value. $\sigma_i$ is the spread of the Gauss function. $W_{corr}$ and $T_{corr}$, which are related with meteorological parameters – wind speed and temperature, are given in Gyldenkærne (2005).~~ Here, $\mu_i$ is the mean value for the parameterized distribution. This means that $\mu_i$ (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 $\mu_i$ depends on the results from the crop growth model which vary from cell to cell over the entire model grid. $\sigma_i$ 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 $\sigma_i$ means that the emission from the corresponding activity takes place during most of the year, while a small $\sigma_i$ means that emission takes place during a few weeks. Here *t* is the actual time of the year. The temperature correction Tcorr and the emission potential $Epot_i(x,y)$ (calculated in the preprocessing) is given in eq. (3) and eq. (4).

$$T_{corr} = e^{(0.0223*t(x,y))} \qquad \text{for i= 8, 9, 10, 11, 12, 13} \qquad (3)$$

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

$$Epot_i(x,y) \neq 1 \qquad \text{for i= 8, 9, 10, 11, 12, 13} \qquad (4)$$

$$Epot_i(x,y) = 1 \qquad \text{otherwise}$$

The emission from plants is only included in the inventories for a few countries (e.g. Gyldenkærne et al., 2005) and can in principle be calculated on-line in a chemical weather forecast model (e.g. Sutton et al., 2013) by using a mechanism that describes the bi-directional flux (Massad et al., 2010). Emissions from plants were therefore not included here.

## 2.2 Emissions input data and scenarios

The spatial pattern of $NH_3$ agricultural emission for Poland for the year 2010 was prepared using the methodology proposed by Dragosits et al. (1998), which is implemented in several atmospheric model systems over the UK (e.g. Oxley et al., 2013). Data on the animal number and fertilizer consumption, provided by the Polish National Statistical Office, were combined with the national emission estimates (KOBIZE 2013) and spatially allocated with using gridded data from the Corine Land Cover map (European Commission, 2005). Data on animal numbers were available at commune level and fertilizer consumption at province level. Detailed information about the calculation methodology used for Poland is described in Kryza et al. (2011). The annual $NH_3$ emissions were gridded to a spatial resolution of 5 km x 5 km to be in accordance with the mesh in the meteorological model (Fig. 1).

Table 2

~~The annual gridded $NH_3$ emissions were then used to construct 4 scenarios termed DEFAULT (1), NOFERT(2), FLAT(3), and POLREGUL(4), respectively (Table~~).

The annual gridded $NH_3$ emissions were then used to construct four 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 spatial 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 practice and 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 in respect 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.

For the POLREGUL scenario the information on Polish infrastructure and management methods was obtained from the IIASA review for the Danish and Polish area (Klimont and Brink, 2004). Firstly, both countries have a ban on application of manure and mineral fertilizer before 1 March. Secondly, the manure storage capacity in Poland is about 3 months, compared to 7-9 months in Denmark. This means that farmers in Poland need to apply manure during spring, summer and autumn. ~~In Poland between 10-20% of husbandry manure (only slurry) are applied to grassland, while they cover about 25% of the agricultural area.~~ In

Poland the solid and slurry fractions of the manure is applied differently due to national regulations. Solid manure goes into annual crops as only slurry is allowed on grasslands. Between 10% and 20% of the slurry fraction is applied to grassland, which covers about 25% of the entire agricultural area. Poland does not have a detailed nitrogen quota system at the field level like Denmark does, and the Polish regulations do not contain definitions of manure-N efficiency. The Danish regulations force farmers to apply most of the mineral fertilizer and husbandry manure into growing crops, and there is a strict limit on how much manure and mineral fertilizer is allowed to be added to each field in Denmark (Skjøth et al., 2008). A consequence is that a limited amount of mineral fertilizer is used in Denmark and that the majority (90%) is applied to growing crops (April-May) and the remaining part to grassland (summer). This is not the case in Poland, where there is a larger consumption of mineral fertilizer. Assuming that all fields in Poland receive sufficient fertilizer (manure and mineral) without an upper limit forced by regulation, a consequence is that as much manure as possible will be used early in the season and that the majority of the mineral fertilizer will be used on grasslands during summer (especially June, July and August) as there is a ban on applying mineral fertilizer to meadows and pasture after 15 August. Therefore the simple assumption therefore is that all fields will have equal amounts of manure and mineral fertilizers during spring and summer (Table 3, Poland scenario). Finally, the regulations in Poland allow farmers to apply manure to fields throughout October, which is not allowed in Denmark. A consequence is that the timing of this autumn application, when the farms empty their storages, has its peak 2-4 weeks later than in Denmark. We have therefore chosen ordinal day number 290 (counted from the beginning of January each year, in our study 2010) as the default peak time for this activity in Poland.

Table 3

## 2.3 Meteorological input data – WRF~~-ARW~~ model configuration and model performance

The Advanced Research WRF model was used with three one-way nested domains (Skamarock and Klemp, 2008). The outer domain (131 x 131 gridpoints) covers Europe with a horizontal resolution of 45 km x 45 km. The intermediate domain covers the area of central Europe with a resolution of 15 km x 15 km (94 x 94 grid points). The innermost domain (194 x 194 gridpoints) covers the area of Poland at 5 km x 5 km resolution. Meteorological data from the innermost domain are used in this study. Vertically, the domains are composed of 35 terrain-following hydrostatic-pressure coordinates, with the top fixed at 10 hPa. The simulation was driven by the NCEP final analysis available every 6 h with 1.0$^o$ x 1.0$^o$ spatial resolution. Analysis nudging was applied for the first two domains.

The model uses the same configuration of physics as presented by Kryza et al. (2013b), including the Goddard microphysics scheme (Tao et al., 1989), Yonsei University planetary boundary layer scheme (Hong et al., 2006), MM5 similarity surface layer and RRTMG and RRTM schemes for short- and longwave radiation (Iacono et al., 2008; Mlawer et al., 1997). The Kain-Fritsch cumulus scheme is applied for the first two domains (Kain, 2004). For the innermost domain, cumulus convection is explicitly resolved.

Because the WRF~~-ARW~~ derived spatial information on air temperature and wind speed is a key input for the emission model, the modelled meteorological data were extensively evaluated by comparison with the measurements. The measurements were available every 6 h from 69 meteorological stations located in Poland. The model error was calculated for each station and summarized using domain wide error~~. The domain wide error statistics were calculated and summarized with three error statistics~~: mean error (ME), mean absolute error (MAE) and index of agreement (IOA, unitless). The definitions of the aforementioned error measures are listed in the Supplement (Table 1). Air temperature at 2 m (T2) and wind speed at 10 m a.g.l. (W10), which are used by the dynamic model of ammonia emission, show good agreement with the measurements (Table 4). The air temperature is slightly underestimated, but the IOA is very close to 1.0. The wind speed is slightly overestimated, with the ME >0.

Table 4

## 2.4 The FRAME model

The standard version of the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) provides information on the annual mean oxidized sulphur and oxidized and reduced nitrogen atmospheric air concentrations and deposition. A detailed description of the FRAME model is given in Singles et al. (1998), Fournier et al. (2004), Dore et al. (2006) and Vieno et al. (2010). Details on the model configuration for Poland can be found in Kryza et al. (2010), Kryza et al. (2012) and Werner et al. (2014). FRAME is a Lagrangian model which describes the main atmospheric processes in a column of air moving along straight-line trajectories following specified wind directions. The model consists of 33 vertical layers of varying thickness, ranging from 1 m at the surface to 100 m at the top of the domain. As such the FRAME model is designed for studies where processes on local scale and landscape scale will be governing (e.g. ammonia emissions) and have a simplified treatment of long-distance transport and associated chemistry. Trajectories are advected with different starting angles at a 1° resolution using directionally dependent wind speed 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 represented 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)

Concentrations at the boundary of the model domain are calculated with the FRAME-Europe model, which is a model similar to FRAME but which runs for the whole of Europe on the EMEP grid at 50 km x 50 km resolution. For this study the model was adapted to run and provide results at monthly resolution. Monthly wind roses were developed from the WRF data using a method similar to that described by Dore et al. (2006). Information on rainfall for FRAME was calculated by using observed data from 210 rainfall sites in Poland. Geographically weighted regression kriging, with elevation used as an independent explanatory variable (Szymanowski et al., 2013), was used here to produce a 5 km x 5 km gridded data set that matches the meteorological grid from the WRF model.

FRAME was run four times for each month. Simulations for 1 month differ in the emission scenario, which are described in Table 2 (section 2.2).

### 2.5 Measurements of ammonia ($NH_3$) and ammonium ($NH_4^+$) air concentrations & backward trajectories from the WRF~~-ARW~~ model

Verifying observations are obtained from stations within the EMEP network (Aas et al., 2012). Four EMEP stations that measure daily air concentrations of gaseous ammonia and aerosol ammonium ($NH_3$+$NH_4^+$) and $NH_4^+$ are available for Poland: PL02 Jarczew, PL03 Śnieżka, PL04 Łeba, PL05 Diabla Góra (Fig. 1).~~ Three of these EMEP stations are located in specific geographical areas (Łeba – sea coast in the north, Śnieżka – the highest peak in the Sudety Mountains, SW Poland, Diabla Góra – in a large forestland, NE Poland).~~ 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). These areas contain limited or even no agricultural activity. Only Jarczew station, located in central-eastern Poland, is located in an agriculture area, and therefore best suited for validation of the model results. One additional site from the NitroEurope network provided measured monthly ammonia concentration. This site, Rzecin, is located in a wetland area, which is surrounded by forests with full coverage of woodland within the nearest 1 km. Land cover outside this woodland is mainly agricultural, and with the highest ammonia emissions in Poland.

Error statistics ME, MAE and R for modelled and measured $NH_3$ concentrations were presented for each site individually, and mean statistics based on five stations were calculated for the entire year and for the periods with (March-October) and without application of manure (January, February, November, December). The definitions of the error measures are listed in the supplementary material (Table 1).

Additionally, for Jarczew, the 3-hourly emissions from the dynamic model were aggregated into daily values and plotted with average daily concentrations from the station. The daily observations and aggregated model calculations were then sorted in two groups: (1) a group with high concentrations of $NH_3$ that were not simulated by the emissions model, and (2) the remaining days. Group 1 was then investigated in detail with air mass trajectories calculated with WRF~~-ARW~~ data~~. RIP version 4.5 (Stoelinga., 2008) was used to get 36 hours backward trajectories for the Jarczew station~~ RIP version 4.5 (Stoelinga, 2009), which is a is a Fortran program used for visualizing output from gridded meteorological data sets, was implemented to get 36 h backward trajectories for the Jarczew station.

~~. 6 trajectories were run for each day with an episode from group 1, once every 6 hours and for the receiving heights 250 m and 750 m.~~ 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

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between 300 and 700 m do not show large differences in transport path within the first 12-24 hours. Each episode was then analyzed with respect to potential atmospheric transport from neighborhood regions with high ammonia emissions.

# 3 Results

The results are organized as follows: first the annual ammonia emission and results from the POLREGUL option of the dynamic model for Poland are described. In the second subsection FRAME model concentrations from four runs (FLAT, DEFAULT, POLREGUL, NOFERT) ~~(DEFAULT, NOFERT, FLAT and POLREGUL)~~ are presented and compared with measurements. Finally, the relationship between the dynamically modelled emissions and measured concentrations for one selected station was presented.

## 3.1 $NH_3$ emission in Poland in 2010

Total ammonia emission (sum for the total country area) in Poland in 2010 was 270 Gg. The highest annual emissions are in the central part of the country, and locally exceed 35 kg ha$^{-1}$ year$^{-1}$ (maximum 45 kg ha$^{-1}$ year$^{-1}$, Fig. 2). These are areas with agricultural activity contributing to the majority of $NH_3$ emissions in Poland. The $NH_3$ emissions are in the range of 1 to 10 kg ha$^{-1}$ year$^{-1}$ over 70% area of the country. The lowest emissions are in the west, north-west and south-east, where agricultural activity is less intense and large areas are covered with forests.

Figure 1

From analysis of the monthly total (Fig. 2), it can be seen April is the month of the highest emission for both the DEFAULT and POLREGUL model run. In the case of the DEFAULT run about 40% of the annual emission is related to this month and minor emission peaks appear in March, July and September. For the POLREGUL scenario the April peak is lower by about 40% in comparison to DEFAULT, and ~~the~~ increased emission also appears in July and October (Fig. 2). Generally, there was higher emission in the period with average monthly temperature above 5.0ºC.

The seasonal variation of emission (POLREGUL run) for different agricultural categories for the grid representing Jarczew station is shown in Fig. 3. In April, which is also the month with the highest ammonia emission for the total area of Poland, three functions have their highest values. At this time, the peaks are observed for applications of manure on bare soils, application of fertilizers and manure on crops. ~~Ammonia treated straw (Fct15) is responsible for high ammonia emission in summer, whereas the autumn peak of emission (end of~~

~~September, October) is related mainly to application of manure.~~ Application of manure (Fct10) is responsible for peak of emission in summer and autumn. Emission related to livestock is dominated in Poland by Fct1 because of large-scale farming of pigs ~~in Poland~~ (37 cattle and 99 pigs per 100 ha of agricultural land, ~~GUS~~ Central Statistical Office of Poland 2010, stat.gov.pl). Due to meteorological conditions (temperature), their contribution is doubled in the summer season in comparison to winter.

Figure 2

Figure 3

~~The spatial distribution of ammonia emission for selected days (sum of emission between hours 12-15 UTC for the 15th day of a month) is presented in Fig. 4. The three hourly averages for the total area of Poland are equal to 0.5, 8.0, 3.0, 2.7 g ha$^{-1}$, respectively, in February, April, June and September.~~ The spatial distribution of ammonia emission for selected months of each season (February, April, June and September) is presented in Fig. 4. The monthly averages for the total area of Poland are equal to 0.11, 2.56, 0.42, 0.70 kg ha$^{-1}$ in February, April, June and September, respectively. The maximum values are observed in April in the central part of the country, where they reach ~~50~~ 10-12 kg ha$^{-1}$.

Figure 4

For the three selected locations (names of the locations are taken after the nearest towns, marked in Fig. 1) in Poland – Wrocław (south-west), Suwałki (north-east) and close to Leszno (middle-west) - hourly emissions for the selected period (from March to May) are shown in Fig. 5. Two of these locations represent the areas of the longest (Wrocław) and the shortest (Suwałki) growing season in Poland. The spring increase in emission appears first in Wroclaw (middle of March) and then almost four weeks later in Suwałki. Leszno is located in the area with the highest ammonia emissions in Poland. ~~There is a large day to night emission variability due to diurnal variability in air temperature and wind speed.~~ 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%).

Figure 5

### 3.2 $NH_3$ concentration calculated with the FRAME model

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The spatial distribution of modelled $NH_3$ concentration from the POLREGUL scenario for February, April, June and September is illustrated in Fig. 4. The highest concentrations are in the agricultural areas in the central part of Poland, with maximum values equal to 1.32, 26.0, 16.5 and 9.2 µg $m^{-3}$ for February, April, June and September, respectively. High spatial correlation (≥0.9) between the modelled ammonia emission and FRAME ammonia concentration (Fig 4.) was calculated for each month.

Time series and error statistics of modelled (FLAT, DEFAULT, POLREGUL, NOFERT)~~(DEFAULT, NOFERT, FLAT, POLREGUL)~~ and measured $NH_3$ concentrations are presented in Fig. 6 and Table 5. For most sites (Rzecin, Jarczew, Łeba and Śnieżka) R and MAE are best for the NOFERT and POLREGUL runs.  The best performance was obtained for Jarczew and Rzecin. For each station the DEFAULT run calculates that the concentrations peak in April which is not present in the measurements or is much ~~lower~~ higher than observed (Jarczew, Rzecin). Application of Polish ~~regulations~~ practice in the dynamic model has improved the results most significantly in comparison to the DEFAULT for Rzecin and Jarczew. Jarczew is the only station located directly in an agricultural area, whereas Rzecin is under the influence of an agricultural region with the highest ammonia emission in Poland. The poorest performance for each model run is for Diabla Góra, for which the measured time series has a ~~totally~~ different pattern in comparison to the other sites.  High measured concentrations for this station are obtained in late autumn and the winter months.

For three model runs (FLAT, DEFAULT, NOFERT) ~~(DEFAULT, NOFERT, FLAT)~~ correlation coefficients are lowest for the summer period in comparison to the entire year (Table 6), whereas the summer period has the highest correlation coefficients for the POLREGUL scenario. The POLREGUL scenario therefore improved the results significantly in comparison to DEFAULT for summer period – the correlation coefficient increased from 0.21 to 0.73 and MAE decreased from 0.83 to 0.68.

Figure 6

Table 5

Table 6

### 3.3 Comparison of daily emissions with measured concentrations and backward trajectories case study

Due to the high spatial correlation between ammonia emission and concentration (Fig 4) we looked for the relationship between the dynamically modelled emissions and measured concentrations for Jarczew station (Fig. 7). The main peaks in emissions (April, September) are reflected in the concentration data. There are also some peaks in concentrations (e.g. end of February, beginning of June and end of October) which are not resolved by the emission model. These could suggest the limitations of the emission model, or could be related to meteorology which has resulted in the transport of ammonia from neighboring areas. Backward trajectories, for the mentioned high concentration episodes (end of February, beginning of June and end of October), were calculated with the RIP tool (Fig. 8) in order to check whether it is possible to connect these observed peaks in concentrations with atmospheric transport of ammonia. We have found that for these episodes the trajectories have a similar pattern - transport from the south or south-west sector. The air masses that reached Jarczew have during these episodes passed areas with high ammonia emissions in comparison to the local area surrounding the station.

Figure 7

Figure 8

## 4 Discussion and conclusion

The temporal and spatial variability of ammonia emission has been analysed over Poland with four scenarios: FLAT (no temporal variation), DEFAULT (matches the Europe-wide default settings in the ammonia emission model, Skjøth et al. 2011), POLREGUL (takes into account Polish infrastructure and less regulation compared to DEFAULT) and NOFERT (excludes application of manure and mineral fertilizer).

~~DEFAULT (matches the Europe-wide default settings in the ammonia emission model, Skjøth et al. 2011), NOFERT (excludes application of manure and mineral fertilizer), FLAT (the existing emission method (no temporal variations) used in WRF-Chem over Poland) and POLREGUL (takes into account Polish infrastructure and current and less regulation compared to DEFAULT).~~ The emissions were then been implemented in the FRAME model for a fast response on simulating the effect of the scenarios in relation to atmospheric chemistry. The results show that, in general, the model ~~simulations~~ results for the agricultural areas were improved by applying a dynamical model by using Europe-wide (default) settings instead of using a fixed emission profile. ~~However,~~ If Polish ~~infrastructure~~ practice and national regulation is incorporated into the emission model, the FRAME model performance is further improved ~~much better results are achieved for agricultural areas~~.

The model results show large difference in emissions between months, as well as between day and night. This is due to increased volatilization of ammonia caused by increased temperatures (Eq. 1 and 2) and emission from animal and mineral fertilizer that are applied over short time periods during spring, summer and autumn (Eq. 2). Taking into account the entire area of the country, the highest emission is obtained during spring (especially in April). The spring emission peak (and corresponding concentrations) is mainly related to the application of fertilizers and manure, which is clearly illustrated by comparing the POLREG and NOFERT simulations (Fig. 6). The sensitivity of the model to the application of manure is highlighted by the large difference between the DEFAULT and POLREGUL scenarios. In April the emission is 40% lower in the POLREGUL scenario than in the default scenario. This is not surprising, as previous results have shown that national regulation can change emissions from manure to increase by more than 100% in spring and decrease during summer to less than 10% (Skjøth et al., 2008). The scale and character of changes between the POLREGUL and DEFAULT simulations with the dynamic ammonia model will vary between countries and depend on local agricultural infrastructure and practice. The dynamic model predicts the spring peak in emission to start in south-west Poland and then progressing to the rest of the country (Fig. 5). South-west Poland has the longest growing season (Żmudzka, 2012) and is the area where farmers initiate their field activities in Poland. In this region, field work, including application of fertilizers or manure, can start earlier than in other regions of the country. This aspect of a ”northward progressing ammonia plume” due to spring application is therefore very well captured by the model and has also been implemented in the GEOS-CHEM model (Paulot et al., 2014) and DEHM models (Skjøth et al., 2011).

Major $NH_3$ emission peaks modelled for the Jarczew agricultural station are also observed in $NH_3$ concentration measurements. However, some peaks in concentrations are not reflected in the emission data. As suggested by Asman et al. (1998) and Fowler et al. (1998) atmospheric ammonia can be transported up to 100 km. According to Geels et al. (2012) the fraction of locally emitted $NH_3$ depositing locally is ~~on~~ of the order of 15-30% for a grid of 16 km x 16 km. In our study, the analysis of backward trajectories showed that increased concentrations can be related to transport of ammonia from neighboring areas with high emission. A more thorough investigation on this scale requires more sophisticated modelling tools than FRAME ~~such as WRF-Chem~~.

FRAME is a relatively simple Lagrangian model and the results were found to be in good agreement with measurements for Poland (Kryza et al., 2011, 2012; Werner et al., 2014) and for the UK (Dore et al., 2015). This enables us to run several scenarios for the entire year without 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 a daily and weekly basis (Geels et al., 2012). OML and FRAME use similar principles for 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. Although 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 chemical conversion and deposition (dry and wet). These results correspond well with the two latest reviews on this subject (Hertel et al., 2006, 2012).

~~Furthermore, the results with FRAME are considered good enough for this study on the emission patterns, as the obtained monthly correlation coefficients on the POLREGUL scenario are similar to the model results that are obtained both with DEHM (Skjøth et al., 2011) and the DAMOS system (Geels et al., 2012). In fact, the very high spatial correlation between ammonia emission and ammonia concentration (Fig. 4) suggests, that on a monthly basis, the governing process for ammonia concentrations is emission and only to a smaller degree atmospheric transport and transformation. The four scenarios show that focus on the agricultural practice, national regulations and the infrastructure (e.g. storage facilities) is a key challenge but very important for obtaining the best results. For Poland it was possible to obtain almost as good results as DEHM for Danish and German sites, despite the lack of detail about location of the agricultural fields and the location, amount and type of livestock in Poland. This suggests that similar improvements can be obtained for other European areas.~~

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.

One of the sites (Diabla Góra) has an inverted time series in comparison to all other stations - the highest ammonia concentration appears in late autumn and in the winter months. Our calculations, which took into account only agricultural sources, were not able to catch peaks in this period. The Diabla Góra station is located in a large forested area called “Borecka Forest”, surrounded by lakeland, with a small contribution of arable land in the region. ~~Due to this location, high ammonia concentrations in this period may be related with natural sources.~~ Open water areas (Barrett, 1998; Sørensen et al., 2003) and natural land areas (Duyzer, 1994) have been shown to emit $NH_3$. Emission of $NH_3$ from ecosystems are found to take place when the atmospheric $NH_3$ concentration is lower than the stomatal $NH_3$ compensation point (Mattsson et al., 2009), as a result of decomposing leaf litter and due to cuticular desorption (David et al., 2009; Hansen et al., 2013). As suggested by Hansen et al. (2013), natural ammonia emission from deciduous forests should be considered as an emissions source which could be dynamically simulated with atmospheric transport models. 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).~~. Another factor that can cause an increase of ammonia concentrations could be evaporation from ammonium containing aerosols.~~ Ammonium-chloride, ammonium-nitrate and ammonium-bi-sulphate are all formed from reversible processes in the atmosphere. ~~Such processes can more efficiently be studied with models like WRF-Chem, once they have been connected with a dynamical ammonia emission model.~~ 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.

The dynamical approach has consistently provided good results for agricultural regions during the winter months, which is due to the large response on ammonia emission from agricultural buildings caused by outside temperatures. An implementation of this type of emission model into ~~WRF-Chem~~ CTM online coupled with meteorology will be a direct response to the suggestion by Sutton et al. (2013) and a direct coupling between ammonia emission, meteorology and chemistry and can address some of the challenges in the modelling of air pollution that have been highlighted (Baklanov et al., 2014).

For regional modelling of ammonia in Europe, the overall results suggest that it will be an advantage to move from a static to a dynamic approach. The Europe-wide default setting in the model given by Skjøth et al. (2011) can be expected to improve the results over large areas, but a better picture over Poland will be obtained if the values from Table 2 in Skjøth et al. (2011) are replaced with the values from our POLREGUL scenario. Further improvement on ammonia emissions is likely to be related to natural sources (Hansen et al., 2013; Riddick et al., 2014) as well as the dependence on emission from fertilizer on soil type as shown by the CHIMERE model (Hamaoui-Laguel et al., 2014). These initiatives are currently beeing adressed by the ECLAIRE project (http://www.eclaire-fp7.eu/), which focuses on climate driven emissions (BVOCs and ammonia) as suggested by the latest IPCC report (2013), that calls for more studies on the feedback mechanisms between climate and air quality.

**Field Code Changed**

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Table 1. The functions describing the temporal variation in $NH_3$ emissions from various activities*.

| Function | Description | Required meteorological parameters* |
|---|---|---|
| Fct1 | Animal houses with forced ventilation | $W_i$, $T_i$ |
| Fct2 | Open animal houses | $W_i$, $T_i$ |
| Fct3 | Manure storage | $W_i$, $T_i$ |
| Fct8 | Spring application of manure on bare soil | $W_{corr,}$ $T_{corr}$ |
| Fct9 | Application of manure on crops | $W_{corr,}$ $T_{corr}$ |
| Fct10 | Summer application of manure | $W_{corr,}$ $T_{corr}$ |
| Fct11 | Autumn application of manure | $W_{corr,}$ $T_{corr}$ |
| Fct12 | Spring application of fertilizers | $W_{corr,}$ $T_{corr}$ |
| Fct13 | Summer application of fertilizers | $W_{corr,}$ $T_{corr}$ |
| Fct14 | Emission related to grassing cattle | $W_{corr,}$ $T_{corr}$ |
| Fct15 | Emission related to ammonia treated straw | $W_{corr,}$ $T_{corr}$ |

*Functions Fct4-Fct7 have not been simulated in this study (Fct4- Winter crops, Fct5-Spring crops, Fct6-Later spring crops, Fct7- Grass)

Table 2. The emission scenarios used in this study.

| ~~Scenario*~~ | ~~Description~~ |
| --- | --- |
| ~~DEFAULT (1)~~ | ~~A default emission distribution that matches the European-wide default settings in the ammonia emission model, based on the original Danish model (Skjøth et al., 2011).~~ |
| ~~NOFERT (2)~~ | ~~An emission scenario that excludes application of manure and mineral fertilizer.~~ |
| ~~FLAT (3)~~ | ~~The existing emission method (no temporal variations) used in WRF-Chem over Poland (Werner et al., 2014; Werner, Kryza, & Dore, 2013).~~ |
| ~~POLREGUL (4)~~ | ~~A scenario that takes into account Polish infrastructure and current and less regulation compared to Denmark (Klimont & Brink, 2004).~~ |

~~* Scenarios DEFAULT, NOFERT and POLREGUL were prepared with the ammonia emission model (Skjøth et al., 2011) described in section 2.1.~~

| Scenario* | Description |
| --- | --- |
| FLAT (1) | No temporal variations. |
| DEFAULT (2) | A default emission distribution that matches the Europe-wide default settings in the ammonia emission model, based on the original Danish model (Skjøth et al., 2011). |
| POLREGUL (3) | A scenario that takes into account Polish practice and current and less regulation compared to Denmark (Klimont and Brink, 2004). |
| NOFERT (4) | An emission scenario that excludes application of manure and mineral fertilizer. |

* Scenarios DEFAULT, POLREGUL and NOFERT were prepared with the ammonia emission model (Skjøth et al., 2011) described in section 2.1.

Table 3. Relative distribution of the total $NH_3$ emission from the agricultural activities in Poland as defined by $Fct_1$-$Fct_{15}$. Poland default – distribution based on Europe-wide default settings, Poland scenario – distribution based on Polish ~~infrastructure~~ practice and regulations.

| Name | Fct1 | Fct2 | Fct3 | Fct8 | Fct9 | Fct10 | Fct11 | Fct11a | Fct12 | Fct13 | Fct14 | Fct15 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Poland default | 0.20 | 0.09 | 0.07 | 0.09 | 0.09 | 0.00 | 0.05 | 0.05 | 0.28 | 0.03 | 0.05 | 0.01 |
| Poland scenario | 0.20 | 0.09 | 0.07 | 0.07 | 0.05 | 0.02 | 0.07 | 0.07 | 0.10 | 0.20 | 0.05 | 0.01 |

Table 4. Domain wide error statistics for 2 m temperature (T2) and 10 m wind speed (W10) over Poland for 2010.

| | ME | MAE | IOA |
|---|---|---|---|
| T2 | -0.68 K | 1.79 K | 0.99 |
| W10 | 0.16 m $s^{-1}$ | 1.29 m $s^{-1}$ | 0.84 |

Table 5. FRAME model results - error statistics for the individual sites (mean from 12 months).

| Statistic | Run | Rzecin | Jarczew | Łeba | Śnieżka | Diabla Góra |
|---|---|---|---|---|---|---|
| ME | DEFAULT | -0.32 | 1.44 | 0.26 | 0.56 | -0.02 |
| | NOFERT | -0.76 | -0.50 | -0.14 | 0.21 | -0.47 |
| | FLAT | -0.34 | 0.31 | 0.00 | 0.13 | -0.13 |
| | POLREGUL | -0.36 | 1.30 | 0.24 | 0.51 | -0.07 |
| MAE | DEFAULT | 0.68 | 1.75 | 0.48 | 0.56 | 0.74 |
| | NOFERT | 0.76 | 0.55 | 0.18 | 0.21 | 0.49 |
| | FLAT | 0.63 | 0.62 | 0.24 | 0.16 | 0.25 |
| | POLREGUL | 0.39 | 1.66 | 0.33 | 0.51 | 0.68 |
| R | DEFAULT | 0.48 | 0.55 | 0.06 | 0.38 | -0.28 |
| | NOFERT | 0.92 | 0.81 | 0.64 | 0.43 | -0.80 |
| | FLAT | 0.02 | 0.72 | -0.18 | 0.14 | 0.06 |
| | POLREGUL | 0.85 | 0.84 | 0.65 | 0.65 | -0.55 |

| Statistic | Run | Rzecin | Jarczew | Łeba | Śnieżka | Diabla Góra |
|---|---|---|---|---|---|---|
| ME ($\mu g\ m^{-3}$) | FLAT | -0.34 | 0.31 | 0 | 0.13 | -0.13 |
| | DEFAULT | -0.32 | 1.44 | 0.26 | 0.56 | -0.02 |
| | POLREGUL | -0.36 | 1.3 | 0.24 | 0.51 | -0.07 |
| | NOFERT | -0.76 | -0.5 | -0.14 | 0.21 | -0.47 |
| MAE ($\mu g\ m^{-3}$) | FLAT | 0.63 | 0.62 | 0.24 | 0.16 | 0.25 |
| | DEFAULT | 0.68 | 1.75 | 0.48 | 0.56 | 0.74 |
| | POLREGUL | 0.39 | 1.66 | 0.33 | 0.51 | 0.68 |
| | NOFERT | 0.76 | 0.55 | 0.18 | 0.21 | 0.49 |
| R (unitless) | FLAT | 0.02 | 0.72 | -0.18 | 0.14 | 0.06 |
| | DEFAULT | 0.48 | 0.55 | 0.06 | 0.38 | -0.28 |
| | POLREGUL | 0.85 | 0.84 | 0.65 | 0.65 | -0.55 |
| | NOFERT | 0.92 | 0.81 | 0.64 | 0.43 | -0.8 |

Table 6. FRAME error results - error statistics from all sites for summer (III-X) and winter (XI-II) period. Unit for ME and MAE is µg m$^{-3}$, R is unitless.

| ~~-~~ | ~~DEFAULT~~ | | | ~~NOFERT~~ | | | ~~FLAT~~ | | | ~~POLREGUL~~ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ~~-~~ | ~~year~~ | ~~III-X~~ | ~~XI-II~~ | ~~year~~ | ~~III-X~~ | ~~XI-II~~ | ~~year~~ | ~~III-X~~ | ~~XI-II~~ | ~~year~~ | ~~III-X~~ | ~~XI-II~~ |
| ~~ME~~ | ~~0.23~~ | ~~0.70~~ | ~~-0.25~~ | ~~-0.31~~ | ~~-0.36~~ | ~~-0.25~~ | ~~0.07~~ | ~~-0.12~~ | ~~0.25~~ | ~~0.16~~ | ~~0.65~~ | ~~-0.32~~ |
| ~~MAE~~ | ~~0.54~~ | ~~0.83~~ | ~~0.25~~ | ~~0.31~~ | ~~0.36~~ | ~~0.25~~ | ~~0.23~~ | ~~0.21~~ | ~~0.25~~ | ~~0.50~~ | ~~0.68~~ | ~~0.32~~ |
| ~~R~~ | ~~0.48~~ | ~~0.21~~ | ~~0.75~~ | ~~0.72~~ | ~~0.70~~ | ~~0.75~~ | ~~0.26~~ | ~~0.04~~ | ~~0.48~~ | ~~0.01~~ | ~~0.73~~ | ~~-0.71~~ |

| - | FLAT | | | DEFAULT | | | POLREGUL | | | NOFERT | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| - | year | III-X | XI-II | year | III-X | XI-II | year | III-X | XI-II | year | III-X | XI-II |
| ME | 0.07 | -0.12 | 0.25 | 0.23 | 0.7 | -0.25 | 0.16 | 0.65 | -0.32 | -0.31 | -0.36 | -0.25 |
| MAE | 0.23 | 0.21 | 0.25 | 0.54 | 0.83 | 0.25 | 0.5 | 0.68 | 0.32 | 0.31 | 0.36 | 0.25 |
| R | 0.26 | 0.04 | 0.48 | 0.48 | 0.21 | 0.75 | 0.01 | 0.73 | -0.71 | 0.72 | 0.7 | 0.75 |

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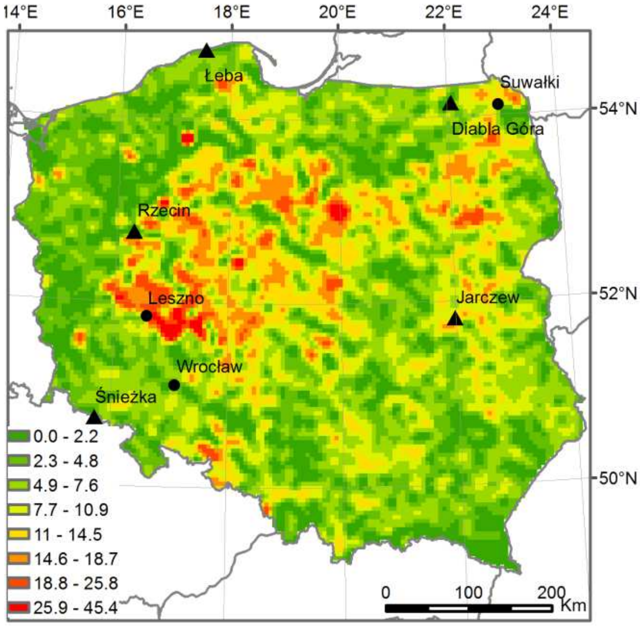


Fig 1.Total annual emission of $NH_3$ in 2010 [kg ha$^{-1}$ year$^{-1}$]. $NH_3$ measurement sites indicated by triangles. Additional locations discussed in the paper indicated by dots (Wrocław, Leszno, Suwałki).

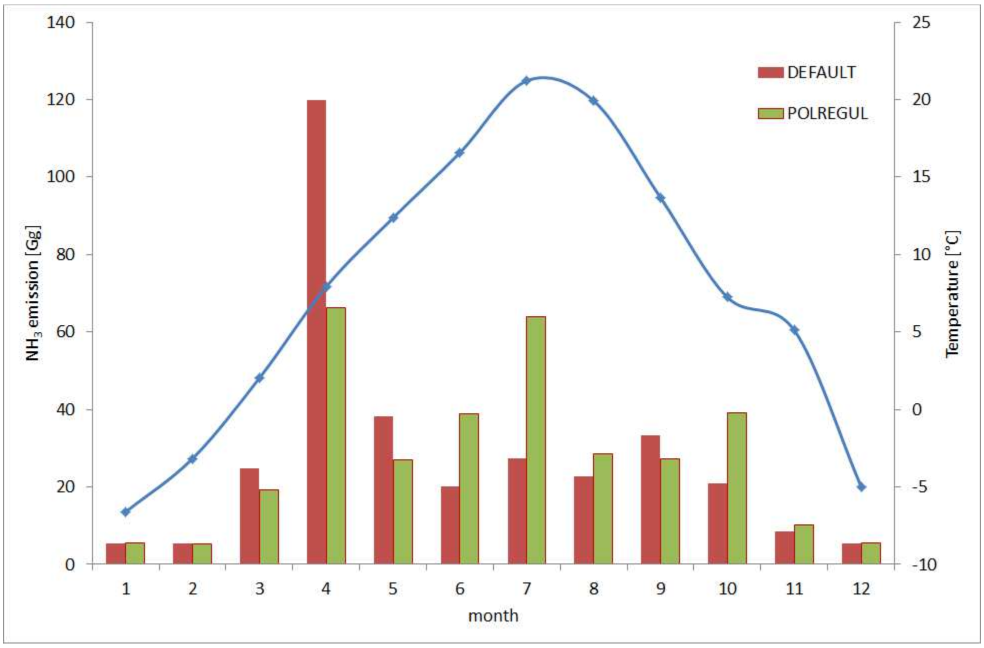


Fig 2. Monthly emission of $NH_3$ for DEFAULT and POLREGUL run and average temperature in 2010.

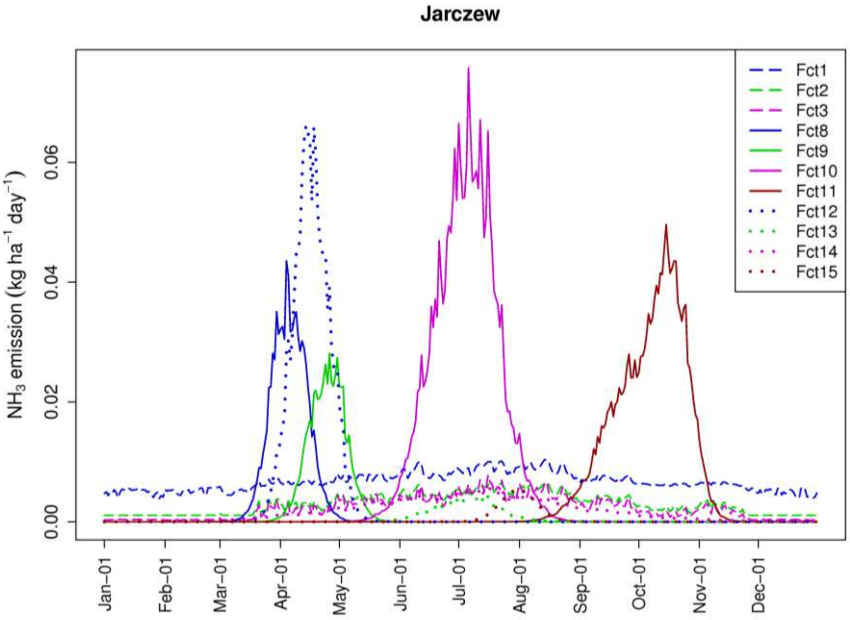


Fig 3. Time series of the seasonal variation in emission (POLREGUL run) for various agricultural emission categories in Jarczew. Description in the legend concerns emission from functions (Fct) described in Table1.

~~Time series of the seasonal variation in emission (POLREGUL run) for various agricultural emission categories in Jarczew (Functions (Fct) described in Table1).~~

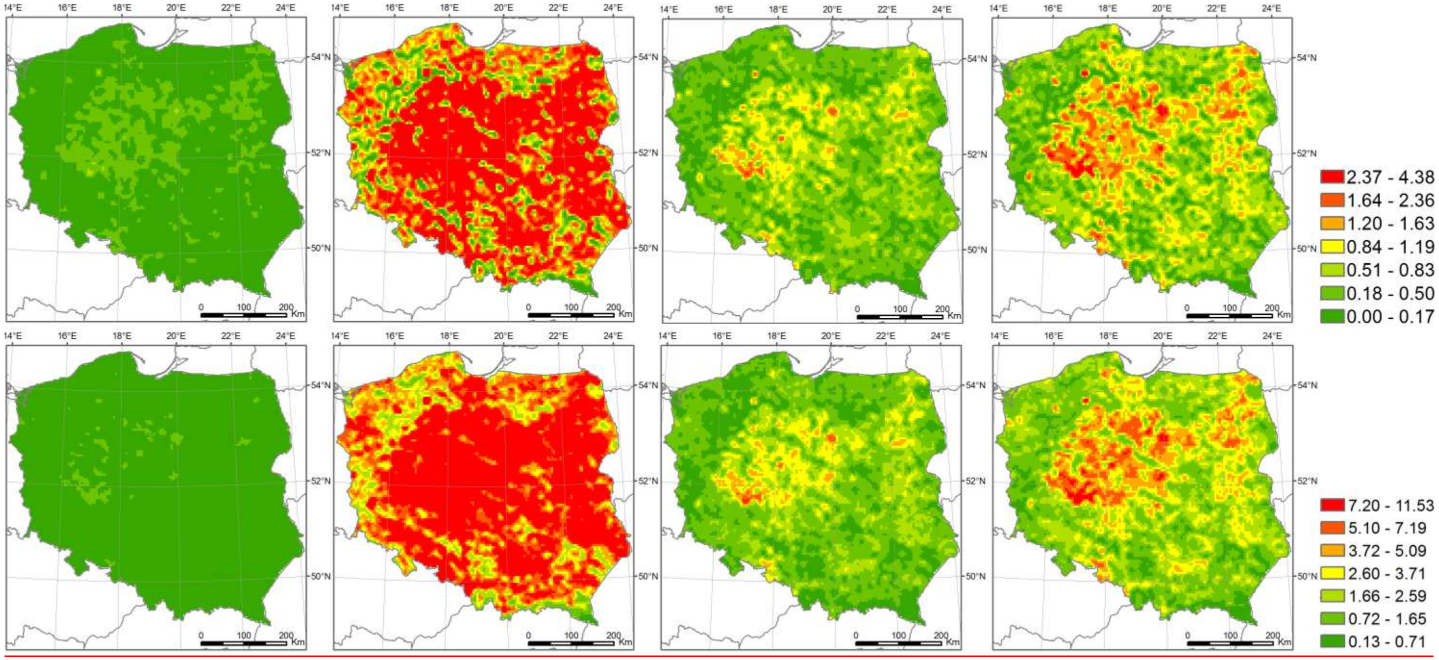


Fig 4. Top: Spatial distribution of $NH_3$ emissions over Poland for ~~15th~~ February, ~~15th~~ April, ~~15th~~ June and ~~15th~~ September 2010 ~~at 12-15 UTC~~ [kg $ha^{-1}$ ~~3 hours~~month$^{-1}$]. Bottom: Monthly mean ammonia concentrations calculated with the FRAME model (POLREGUL) for February, April, June and September 2010 [µg $m^{-3}$].

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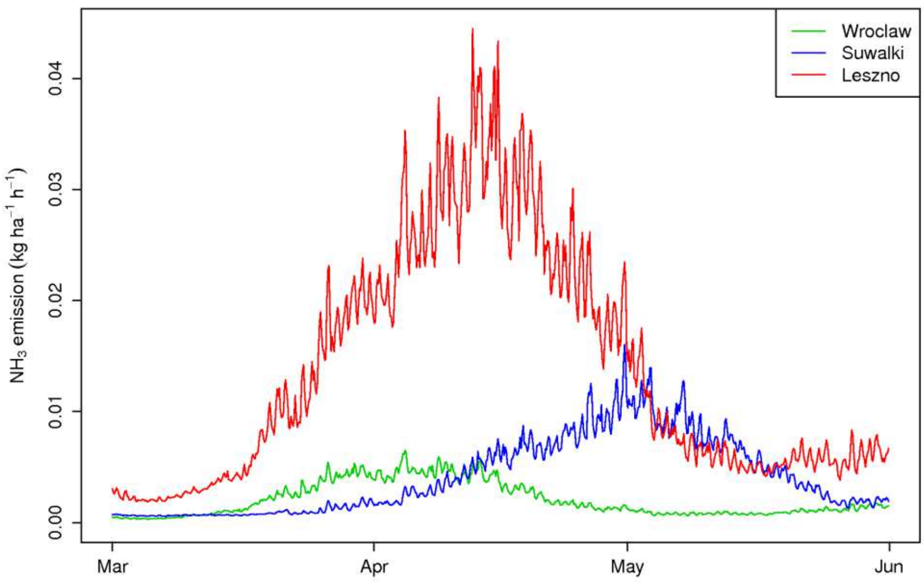


Fig 5. The hourly variation in simulated $NH_3$ emissions for POLREGUL scenario. Data from March to May for three locations in Poland (Wrocław, Suwałki and Leszno).

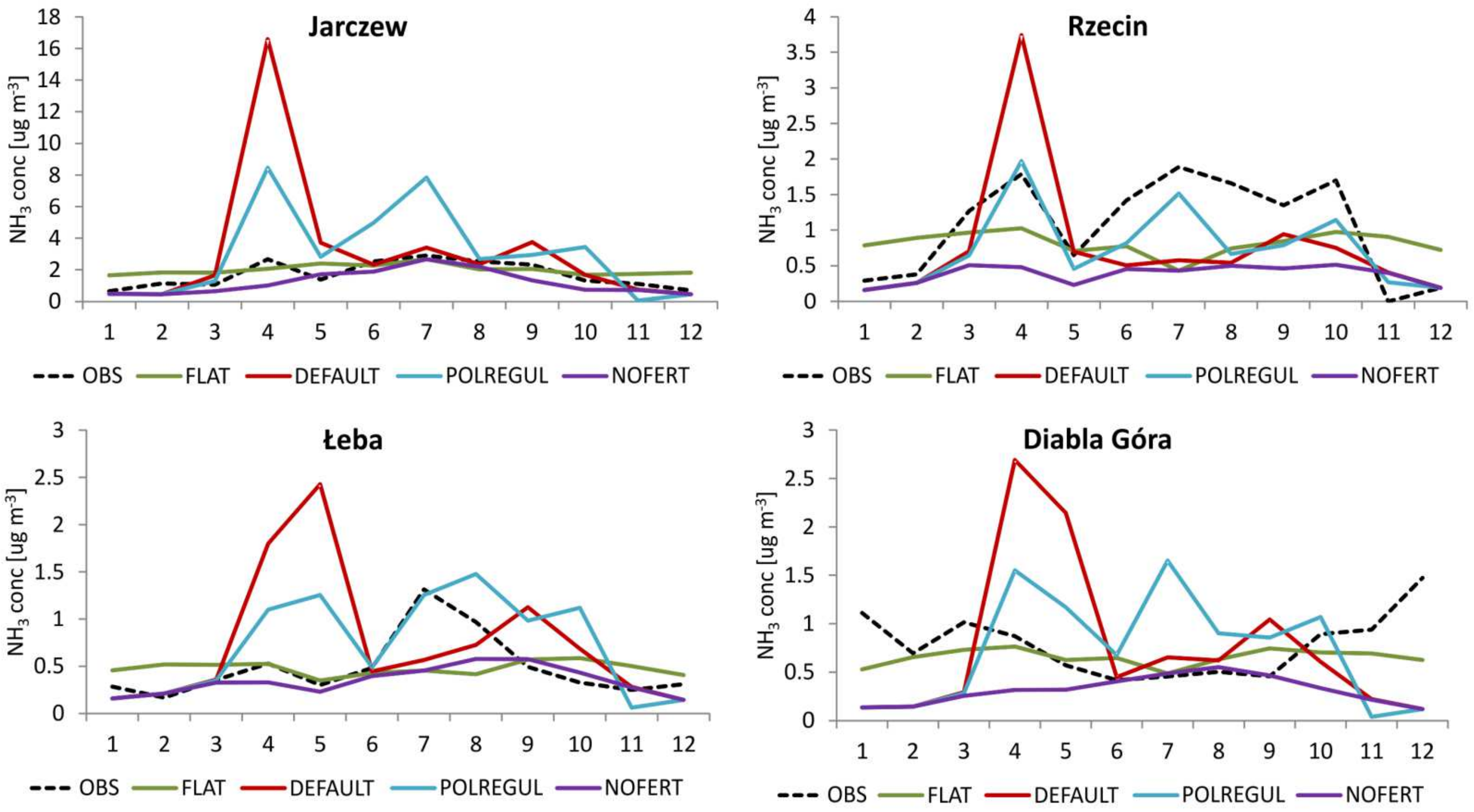


Fig 6. Time series of modelled and measured $NH_3$ concentrations for 2010.

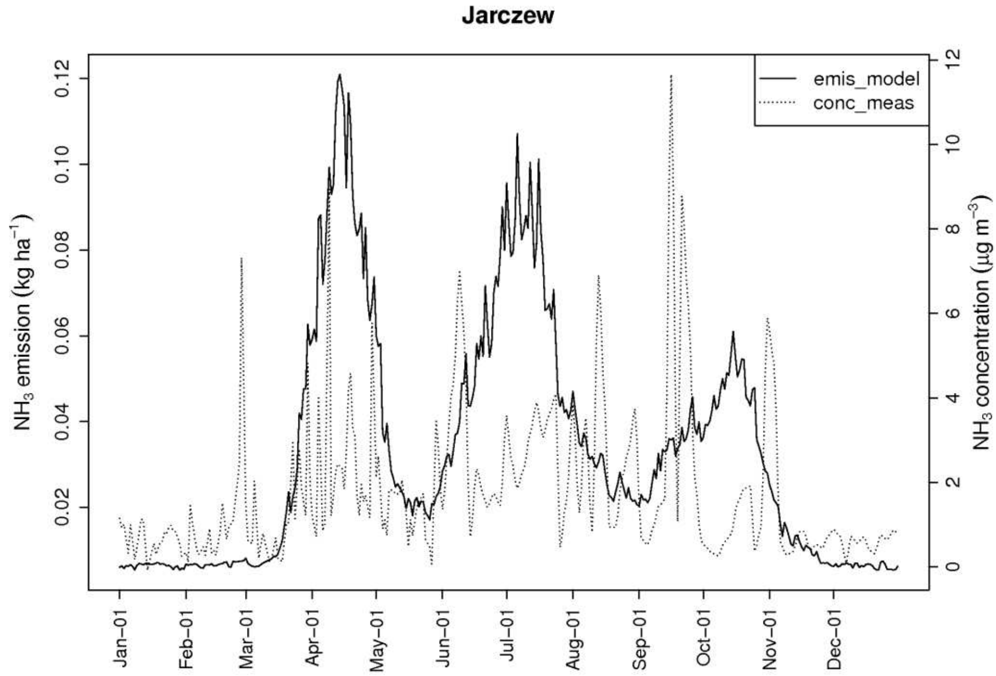


Fig 7. Modelled emission (POLREGUL) and measured concentration for the Jarczew station

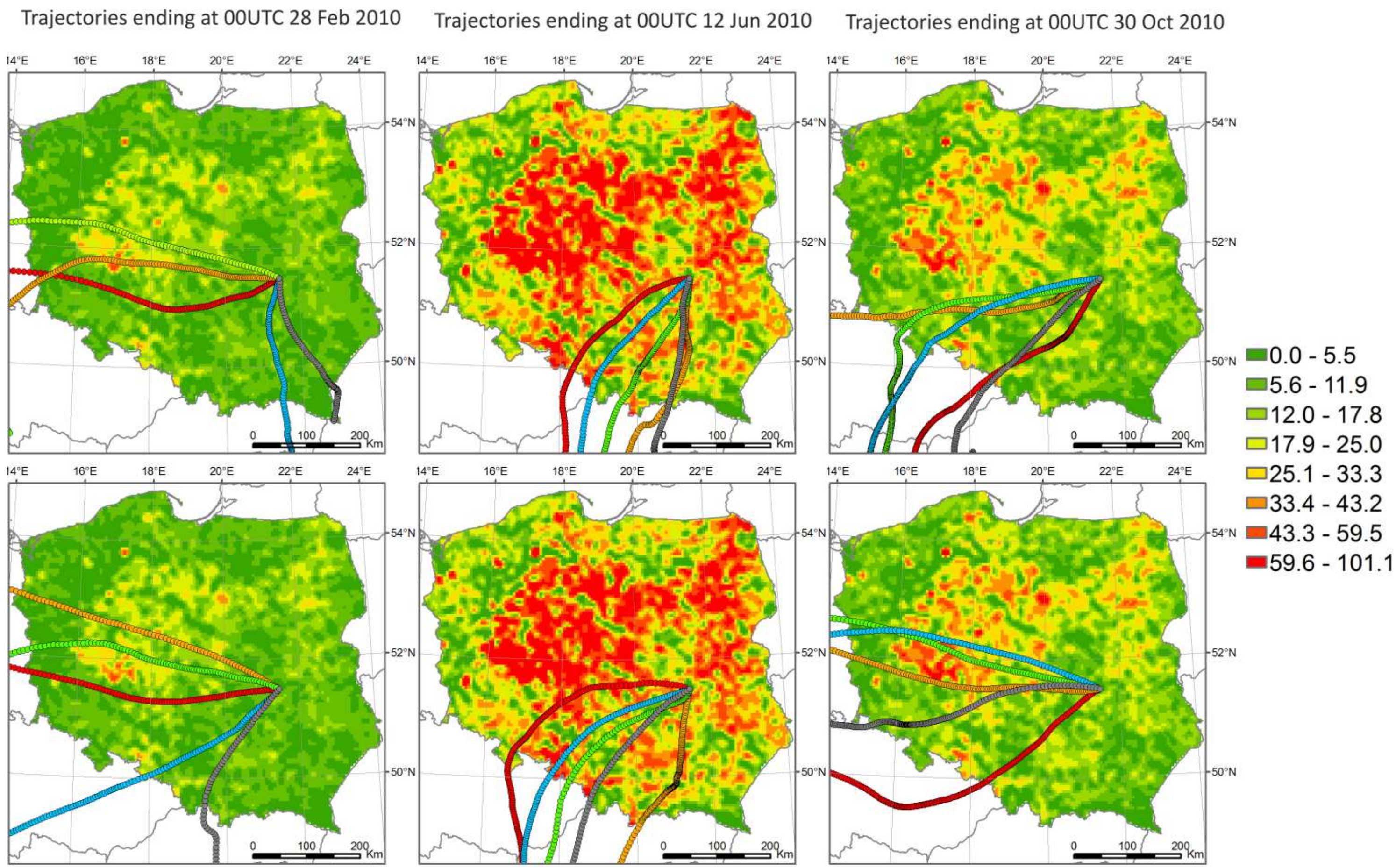


Fig 8. 36-hours backward trajectories ending in Jarczew during episodes (25-27.02.2010; 09-11.06.2010; 10-29.10.2010) with high $NH_3$ measured concentrations. The first trajectory (gray) starts at 12.00 of the first day of each episode, and then starts every 6 hours, and are presented in the following colours: blue, green, orange, red. Spatial distribution of modelled ammonia emission during the episodes (unit: g $ha^{-1}$ $48hours^{-1}$).

1