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Spatial, temporal and vertical distribution of ammonia concentrations over Europe – comparing a static and dynamic approach with WRF-Chem

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NH$_3$ concentrations over Europe – a static and dynamic approach with WRF-Chem

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Abstract

The study focuses on the application of a dynamic ammonia emission into the Weather Research and Forecasting Chemistry model (WRF-Chem) and the influence on the simulated ammonia concentrations and the overall model performance. We have focused on agricultural ammonia sources and have analysed both hourly and daily patterns of ammonia emissions and concentrations at measurement sites located in agricultural areas or influenced by this activity. For selected episodes, we have also investigated the 3-D patterns of the ammonia concentrations in the atmosphere. The application of the dynamic ammonia emission into the WRF-Chem model (the "DYNAMIC" simulation) results in an improvement of the modelled daily ammonia concentrations in comparison to a static approach (the "BASE" simulation), which is currently widely used in chemical transport models. In the case of hourly resolution, we have observed an improvement for the DYNAMIC approach for the winter and autumn seasons, but for the entire year the modelled hourly ammonia peaks are shifted toward the afternoon hours if compared with measurements. This study indicates that the current description of the diurnal cycle of the ammonia concentration from fields is not accurate and more research is needed in order to improve the processes that describe the emission from fertilised fields. The results suggest that the governing processes in relation to the diurnal cycle are the atmospheric mixing and the emission strength. Therefore, an improved description of the diurnal profile of ammonia concentrations within atmospheric models requires a better description of the planetary boundary layer height and a stronger daily pattern of ammonia emission, e.g. through increased evaporation or increased fluxes from the surface.

1 Introduction

Ammonia (NH$_3$) is the most abundant form of reduced nitrogen in the gas-phase within the atmosphere (Behera et al., 2013). Ammonia contributes to both formation of partic-

ulate matter (PM) and deposition of reactive nitrogen to the environment (Banzhaf et al., 2013; Hertel et al., 2012; Reis et al., 2009). Ammonia plays a decisive role in particulate matter formation chemistry by determining the amount of ammonium sulphate and ammonium nitrate as PM constituents (Bessagnet et al., 2014). Erisman et al. (2008) estimate that NH_3 emissions from agriculture give a substantial contribution (13 %) to the PM concentration in Europe and thereby adds significantly to the external costs related to air pollution in Europe (Brandt et al., 2013). The deposition of nitrogen in the form of ammonia can result in eutrophication of sensitive ecosystems and to acidification of the soil (Bouwman et al., 1997). An enhanced load of nitrogen in terrestrial ecosystems has been found to correlate with loss of biodiversity (Stevens et al., 2004) and can increase ecosystem vulnerability to extreme weather and insect attacks (Bobbink et al., 2010). Despite the importance, there are still many knowledge gaps on ammonia. Long term observational data series are in general scarce compared to e.g. ozone (Tørseth et al., 2012), and hourly observations of ammonia are even more rare due to limitations in existing monitoring networks such as EMEP (Tørseth et al., 2012).

Agriculture was responsible for 94 % of the total NH_3 emissions in the EU in 2010 (European Environment Agency, http://www.eea.europa.eu/). Agricultural emissions are related to farm buildings, manure, fertilisers and grazing animals, and are strongly influenced by climate and weather (Skjøth and Geels, 2013). NH_3 emission varies primarily with temperature and air velocity (Monteny and Erisman, 1998). The volatilization potential nearly doubles for every 5 °C and varies significantly through the day and season (Gyldenkærne et al., 2005; Sutton et al., 2013). NH_3 emission is also controlled by water availability, which allows nitrogen compounds to dissolve, be taken up by organisms and be released through decomposition (Sutton et al., 2013). The regional variation reflects local production methods and agricultural practice, which to a large extend is governed by regional scale climate conditions (Skjøth et al., 2011). Despite these well-known dependencies of climate and meteorology on ammonia emissions, the emissions are handled in a very simplified manner in most atmospheric models (Skjøth et al., 2011). Many integrated effects of meteorology and climate on ammonia

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remain to be studied and this has been highlighted by IPCC (Kirtman et al., 2013) as an area that is poorly understood. Improvement of representation of processes that lead to ammonia emission in atmospheric models has therefore frequently been highlighted as an area that needs scientific attention (Flechard et al., 2013; Sutton et al., 2013).

Ammonia emission inventories for regional air-quality models in Europe have mainly been based on a bottom-up approach, where activity statistics are combined with standard emission factors to estimate annual emissions. The temporal variation is typically included as a standardised seasonal variation with a monthly profile (Schaap et al., 2005). Recently, Sutton et al. (2013) suggested two long-term goals on ammonia modelling within CTMs. Firstly, the same meteorological data should be used to drive the emission, the chemistry-transport and the bi-directional exchange. Secondly, the emission should be calculated online in the CTM's whereby the feedbacks between the ammonia emissions and climate can be included. Recently, Zhu et al. (2015) indicated that updates to the governing processes on both dynamics and physics concerning NH$_3$ need improvements. Previous studies have shown significant improvements in model performance by replacing static seasonal variations by a dynamic approach which accounts for physical processes like volatilization of NH$_3$ (Skjøth et al., 2011). Similarly, other modelling studies have shown that some atmospheric components are sensitive to the formulation of the ammonia emission (Gilliland et al., 2003). This highlights the need for a better understanding of ammonia emissions and how this is implemented in CTM models. Results presented by Werner et al. (2015) suggest that implementing this dynamical approach improves simulations even in areas with limited information about location of the agricultural fields, livestock and agricultural production methods.

Recent studies on modelling atmospheric ammonia with CTMs have focused on the implementation of the bidirectional NH$_3$ exchange between atmosphere and surface (Bash et al., 2013; Wichink Kruit et al., 2012; Zhu et al., 2015), impact of ammonia emissions on concentrations of secondary inorganic aerosols (Banzhaf et al., 2013; Bessagnet et al., 2014; Hamaoui-Laguel et al., 2014), investigations of the role of different natural emission sources of ammonia (Hansen et al., 2013; Riddick et al., 2014)

and improvements in the representation of ammonia emission from different agricultural activities, e.g. livestock emission (Zhu et al., 2015) and mineral fertilizers (Hamaoui-Laguel, 2014). Focus on the processes that generate ammonia emissions and the initial dispersion has however had limited attention in the development of existing CTM models. Addressing this knowledge gap is one of the objectives in the FP7 project ECLAIRE (Sutton et al., 2013).

In our study we aim at improving the basic understanding of ammonia in the atmosphere. We will also identify current limitations in relation to the governing processes that cause ammonia emissions and initial dispersion due to meteorological parameters. For this purpose we have implemented the emission from a dynamical ammonia emission model into WRF-Chem and evaluated the model results against a commonly used static approach for describing the emissions. To reveal the limitations in descriptions of the processes, we analyse and compare the model results against hourly observations from one station and daily observations from six stations during the year 2012. The sites have been chosen because they are all located in areas under the influence of agricultural activities. For selected episodes in February, April, July and October, we have analysed both the vertical and temporal development of ammonia concentrations in order to highlight the governing processes that are responsible for initial distribution of ammonia concentrations using meteograms. This analysis of ammonia concentrations both vertically and temporally has not been undertaken before with regional scale atmospheric models. This therefore represents a substantial step forward in understanding the behaviour of ammonia in the atmosphere.

2 Data and methods

2.1 WRF-Chem model

The Weather Research and Forecasting model coupled online with chemistry (WRF-Chem) was used to simulate the meteorological conditions and ammonia concentra-

tions over Europe for the entire year 2012. A complete description of the model is given by Grell et al. (2005) and Fast et al. (2006). The GFS FNL global analysis, created and maintained by the National Centre for Environmental Prediction (NCEP), with a spatial resolution of 1° × 1° (longitude–latitude) and a vertical resolution of 27 pressure levels, were used to define the initial and boundary meteorological conditions. The main setup and the physical and chemical schemes used in this study are listed in Table 1. The last five days of the year 2011 were used as a spin up for the chemistry as in Forkel et al. (2014). We used the RADM2 gas phase chemistry (Stockwell et al., 1990) and the MADE/SORGAM aerosol module (Ackermann et al., 1998; Schell et al., 2001) with the aerosol direct and indirect radiative effect included (Forkel et al., 2012; Werner et al., 2015). Chemistry transport modelling in general benefits from a high number of layers within the Planetary Boundary Layer (PBL) (Zhang et al., 2010), especially near the surface, when calculations concern gases with a fast deposition velocity, like e.g. ammonia (Ellis et al., 2011). Thus, we adjusted the vertical resolution in WRF-Chem by decreasing the thickness of the lowest layer from 53 to 20 m and doubling the number of layers within the first 1015 m, which gives 48 layers in total.

The WRF-Chem model has been extensively used and evaluated for both meteorological and air quality studies in Europe. The model performance for meteorology affects both the air quality results and the calculated emissions. Several studies, focused on the entire Europe, report biases for both air temperature and precipitation, e.g. Miglietta et al. (2012), Katragkou et al. (2015), Wałaszek et al. (2014), Kim et al. (2013), Warrach-Sagi et al. (2013). Recent findings provided by Skjøth et al. (2015) show that the bias in air temperature at 2 m varies spatially and seasonally. These biases are significant and might affect e.g. online calculated emissions and the processes in vegetation models. Similar findings are reported by Kryza et al. (2015) for the area of Poland, where the air temperature bias is low in winter, but summer temperatures are significantly overestimated. A bias in WRF calculated air temperatures were also reported by Mooney et al. (2013) and Miglietta et al. (2012). The model performs well at simulating wind speed (Jiménez and Dudhia, 2013; Miglietta et al., 2012; Santos-Alamillos et al.,

2013; Vieno et al., 2010) which is the second variable affecting ammonia emission in this study.

The WRF-Chem model was run twice in our study. In the first simulation we ran WRF-Chem using the TNO MACC II emission data set with $1/8° \times 1/16°$ spatial resolution (Kuenen et al., 2014) and a temporal emission profile commonly used for the temporal disaggregation in ammonia emissions in current CTMs (Schaap et al., 2005). It includes a seasonal variation that changes each month, but the seasonal variation is the same throughout the entire model domain. This simulation is subsequently referred to as "BASE". In the second simulation we ran the WRF-Chem model with hourly variations in ammonia emissions for the entire year 2012 by using the same approach as for the Danish Eulerian Hemispheric Model (Geels et al., 2012), where the ammonia emission model uses gridded hourly meteorology (calculated with WRF-Chem) to simulate the hourly emission variations (a description of the dynamic model is given in the following section). This simulation is in the following referred to as "DYNAMIC".

2.2 Dynamic emission model

The fundamentals of the dynamic emission model are provided by Gyldenkærne et al. (2005), Skjøth et al. (2004, 2011). The model code is freely available and flexible for use with respect to geographical area and underlying assumptions (Skjøth et al., 2011). The general idea behind the emission model is to use the gridded annual total NH_3 emissions, in this study the TNO MACC II (Kuenen et al., 2014), and to use available activity data to make a disaggregation of the gridded annual totals into specific agricultural sectors. The emission from each sector is then simulated with individual parametrizations. The TNO emission was re-gridded to the WRF-Chem grid using a mass conservation approach. The emission from each agricultural sector uses a parameterisation that depends on both the volatilisation as a function of meteorology and the temporal pattern of activity. The meteorological parameters used in the dynamic model, 2 m temperature and 10 m wind speed, were calculated with WRF-Chem with 1 h temporal resolution and with $36\,km \times 36\,km$ spatial resolution. The emission parame-

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terization consists of 16 additive continuous functions, describing emission from animal houses and storage (3 functions), application of manure and mineral fertilizer (7 functions), emission from crops (4 functions), grazing animals, and ammonia treatment of straw, respectively. 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, b), are based on Europe-wide studies and are considered appropriate for large geographical regions (Skjøth et al., 2011).

2.3   Model evaluation

The focus in this study is on improved understanding of short term variations of ammonia and the impact from agricultural emission sources. Only one site in the EMEP network – Harwell (UK) provided hourly ammonia data from an agricultural region for the year 2012. Additionally, we used all available daily ammonia concentration measurements for 2012 from sites located near the agricultural sources. These sites were: Jarczew (Poland) – data were provided by the Chief Inspectorate of Environmental Protection in Poland and gathered within the State Environmental Monitoring programme and four Danish stations (Risø, Tange, Ulborg and Anholt) that are part of the Danish Background Air Quality Monitoring Program. Thus, we were able to evaluate the model results for remote geographical areas that differ in terms of climate conditions and agriculture practice.

The NH$_3$ and NH$_4^+$ ambient air concentrations were measured using the filter pack method for all the Danish sites and the Polish site Jarczew. The filter pack method does not give a complete separation of NH$_3$ and NH$_4$ (Skjøth et al., 2004). However, comparisons between filter pack and denuder sampling have demonstrated that for Danish monitoring stations a satisfactory separation can be obtained (Andersen and Hovmand, 1994). The NH$_3$ air concentrations at Harwell are measured with the online IC method.

In the model evaluation process, firstly we compared ammonia concentrations from the WRF-Chem model with hourly measurements from Harwell for four seasons (Ta-

ble 2). The seasons were calculated as follows: winter (January, February and December), spring (March, April and May), summer (June, July and August) and autumn (September, October and November). We also explored the temporal variation of ammonia concentrations with height using meteograms for four selected episodes and
compared the surface concentrations with the PBL height. Secondly, we calculated statistics for the entire study period and the six measuring sites using the daily mean NH$_3$ concentrations, both for the BASE and DYNAMIC simulations (Table 3). The following measures were used to summarize the WRF-Chem model performance for both runs: factor of two (FAC2), mean bias (MB), normalized mean bias (NMB), root mean
squared error (RMSE) and correlation coefficient (R). We then calculated mean statistics from all stations according to the seasons (Table 4). The number of observations available for each season is listed in Tables 2, 3, and 4 as "n". Finally, we evaluated the spatial pattern in ammonia emission by calculating the day of the year (Julian Day) for which the model estimated the highest hourly ammonia concentrations, with calculation
performed independently for each grid cell.

3   Results

The results are presented in the following order: (1) temporal pattern of ammonia emission and concentrations including vertical distribution of concentrations, (2) spatial distribution of NH$_3$ concentrations for the BASE and DYNAMIC simulations, (3) comparisons of WRF-Chem ammonia concentrations with observations for the both BASE and
DYNAMIC runs.

3.1   Temporal and vertical distribution of NH$_3$ emissions and concentrations

3.1.1   Hourly ammonia emission and concentration – temporal pattern

The hourly profiles of ammonia emissions (DYNAMIC approach only) and modelled
and measured concentrations (both BASE and DYNAMIC) have been compared for

the Harwell station. These profiles are calculated for each of the four seasons and then normalised for this comparison (Fig. 1). The dynamic approach shows that the emissions typically peak during the afternoon in each case, starting from 1.00 p.m. in the autumn to 3.00 p.m. in the winter and that the minimum is around 6 in the morning,

5　where the difference is up to a factor of two between minimum and maximum. According to the observed values, the highest NH_3 concentrations are during the day time, between 9.00 a.m. and 3.00 p.m. Modelled concentration peaks are shifted towards afternoon hours, when compared with measured NH_3 concentrations. The lowest concentrations are modelled at midday and highest at night. This pattern is similar for all

10　the seasons considered. For each season, DYNAMIC pattern is slightly closer to measurements than BASE. The closest agreement between the measurements and the DYNAMIC run is for winter. For this season, there is also the largest improvement if the BASE and DYNAMIC runs are compared.

　　The effect of meteorology on ammonia emissions and concentrations is also high-

15　lighted with Fig. 2, that shows that the DYNAMIC time series agree better with observations e.g. for the beginning of February and the second part of April. Despite the improvement with the dynamic approach, there is still a significant disagreement on overall peak time between observations and simulations. The plots for July and October reveal that for both BASE and DYNAMIC runs the model is in relatively poor agree-

20　ment with the hourly NH_3 measurements. An example e.g. for the last week of July shows that both the BASE and DYNAMIC simulations generally follow the increased NH_3 measured at Harwell, but are not capable of resolving observed diurnal variations.

3.1.2　Hourly ammonia concentrations – vertical pattern

The vertical distribution of NH_3 concentration has been studied at one location for the

25　selected periods of 1–7 February, April, July and October 2012 using WRF-Chem. We have chosen Harwell as it is the only site with available hourly observations. The vertical distribution of ammonia for the BASE and DYNAMIC simulation is similar for all seasons (Figs. 3 and S1). The general pattern indicates that the highest concentra-

tions are at the surface layer with a linear decrease towards the upper layers. The WRF-Chem model calculates the highest ammonia concentrations at night time, with the time of the maximum varying according to the month. During the periods analysed, this usually occurs before or at midnight in February and July, and after or at midnight in April. In October, the maximum values appear both before and after midnight. In July and October, there are individual days with increased concentrations in the upper layers (4 July and 5 October), which are accompanied by high surface concentrations. The daily pattern of the ammonia concentration is seen to be less regular for some short periods – e.g. on the 5 April, which is related to a precipitation event during that day and washout of ammonia from the atmosphere. For all episodes, ammonia concentration peaks are negatively correlated with the PBL height (Fig. 4). In fact, the periods with the strongest diurnal pattern of ammonia concentrations are also on the days with large differences in PBLH between day and night. For April, we have illustrated the vertical distribution of NO_3^-, NH_4^+ and SO_4^{2-} concentrations in the Supplement (Fig. S2). It can be seen that high concentrations of airborne aerosols (Fig. S2), are slightly shifted with a later peak compared to the peak values of ammonia concentrations (aerosol peaks are about 1–2 h later). These figures also show that the maximum aerosol concentrations appear above the surface layer – for NO_3^- and NH_4^+ this is usually about 200 m above ground level.

3.1.3 Daily ammonia concentrations – temporal pattern

The time series with observed and modelled (BASE and DYNAMIC) ammonia concentrations are presented for all stations in Fig. 5. The modelled peak of ammonia concentrations starts at the beginning of February for the BASE simulation and is moved towards March and April for the DYNAMIC simulation. This results in a better agreement with measurements for each station (Fig. 5). However, the magnitude of this spring time peak is overestimated for all the stations, both for BASE and DYNAMIC. The spring peak is much more extended in time in the BASE simulation, if compared to both the observations and the DYNAMIC simulation. The DYNAMIC simulation pro-

vides a second peak in autumn, which is not present in the observed data for Danish sites and is less pronounced for the BASE simulation.

3.2 Spatial distribution of NH_3 concentrations

The BASE simulation shows that the highest ammonia concentrations in February are in western France, northern Italy and several regions of Eastern Europe (Fig. 6). The DYNAMIC simulation shows that the maximum values in February are for the same regions, but the NH_3 air concentrations are 10 times lower if compared to the BASE simulation. In April, high concentrations with value above $10 \, \mu g \, m^{-3}$ are found in Germany, France, Denmark, and northern Italy for the BASE simulation. For the DYNAMIC simulation high concentrations cover the entire central Europe. In October, both model runs show similar spatial distribution of NH_3 concentrations.

Figure 7 shows the Julian day number (for calendar year 2012), for which the model calculated the highest hourly ammonia concentrations. This was calculated to check whether there is a clear south–north pattern, with increasing number of day towards north for the DYNAMIC simulation and compare this with the results for the BASE simulation. In Central, Eastern and Western Europe, this annual peak day appears later for the DYNAMIC simulation than for the BASE simulation. In the areas with low or no ammonia emissions (e.g. northern Scandinavia) the difference is small between the two simulations. For BASE it is at around day number 60–70 in Central and Eastern Europe, day number 60–90 in Western Europe and above day number 190 in Scandinavian Peninsula. The DYNAMIC simulation shows day number 100–120, day number 70–90, and above day number 190, respectively. Spring application of manure and fertilizers (day number 90–120) described in the DYNAMIC simulation is responsible for the maximum ammonia concentrations in most of the European countries with high ammonia emissions, like e.g. the UK, Denmark, France or Poland.

3.3 Model evaluation for the BASE and DYNAMIC simulation

Comparison of modelled and measured hourly NH_3 concentrations for Harwell for the BASE simulation indicates the lowest NMGE and the highest R for spring (Table 2). The lowest performance according to NMGE and R is for autumn and summer, respectively. For the DYNAMIC simulation all statistics are improved, in comparison to BASE, for the winter and autumn season. The only exception here is R for autumn, which slightly decreases for the DYNAMIC approach.

Mean statistics, based on daily values from 6 stations (Harwell, Jarczew, Risø, Tange, Ulborg and Anholt) for 2012, indicate generally better performance (higher FAC2 and R and lower bias) for the DYNAMIC simulation than for BASE (Table 3). FAC2 and R increase moving from BASE to DYNAMIC for winter, spring and summer and bias and error measures are improved for winter and spring (Table 4). The most significant improvement is for the winter season, whereas the smallest changes are for summer. The DYNAMIC simulation has a larger positive bias than the BASE simulation during spring.

A general pattern of the differences between BASE and DYNAMIC, described above, is present at all the individual sites (Fig. 8). The BASE simulation significantly overestimates the measured ammonia concentration for winter, which is improved for the DYNAMIC simulation (RMSE decreases for all stations, Fig. 8). An improvement in correlation coefficient between BASE and DYNAMIC for Harwell, Jarczew and Tange is accompanied by an increase of the RMSE. The highest decrease in the model performance between BASE and DYNAMIC is for autumn for Danish sites.

4 Discussion and conclusion

We have observed an improvement for the DYNAMIC approach for the winter and autumn seasons but for the entire year the modelled hourly ammonia peaks are shifted toward the afternoon hours if compared with measurements. This occurs both for the

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BASE and DYNAMIC simulation, despite a strong peak of emission in mid-day for the DYNAMIC approach. Analysis of the vertical distribution of modelled ammonia concentrations indicates that the main source of ammonia in the air is from the surface, as the highest concentrations are close to the ground and decrease linearly with height. Moreover, the maximum aerosol concentration appears after the peak of NH_3 concentration, which infers that the ammonia peaks are not related to release of ammonia from aerosols. A similar modelled ammonia concentration pattern to that described above was reported by Wen et al. (2014) for STILT-Chem simulations over southern Ontario in the US, with the limitation that the results were not evaluated against observations. Studies presented by Pinder et al. (2006) for the US emphasise that diurnal profile of ammonia emission is especially important for accurately predicting the concentrations at night, since the decreased atmospheric mixing makes the night especially sensitive to emission changes. Without diurnal variation in the emission, the model overestimates the concentrations at night, which leads to a bias in the average ammonia and ammonium concentration. Pinder et al. (2006) compared the model against mean daily observations and found that in January and July, the seasonally varied inventories significantly improve the predictions of NH_3 and NH_4^+ concentrations. Our study confirms this improvement for the winter season, for which there is an increase e.g. in correlation coefficient or decrease of bias for all four stations representing different geographical areas and climatological conditions. In our investigation however we go a step further by analysing the hourly and vertical ammonia pattern, and found that there is still an unresolved problem with a discrepancy in time of peaks of the ammonia concentration between modelled and measured concentrations.

We have observed that for each analysed episode (March, April, July and October), the surface ammonia concentrations are anti-correlated with the planetary boundary layer height – midday peaks of PBLH are accompanied by a local minimum of NH_3 concentrations. It has been previously shown that PBLH is an important variable for air quality modelling, which is often difficult to simulate accurately in numerical models (Dabberdt et al., 2004; Hu et al., 2010b; Xie et al., 2012). Determining the PBLH is

important in atmospheric numerical models, because it is used in other physical param-eterisations and because it is a governing parameter for the distribution of trace gases (Geels et al., 2007). Meteorological conditions are known to exert a direct impact on the air quality simulation. Han et al. (2008) showed that the difference in modelled vertical turbulent mixing is one of the main reasons for the discrepancy in pollutant concentra-tion among the chemical transport models. The purpose of a PBL parametrization is to redistribute energy and humidity in the PBL. Both humidity and temperature have an impact on ammonia concentrations. Previous studies suggest that the YSU scheme in WRF (Hong et al., 2006), which was also applied in our study, tends to overestimate the PBL height (Hu et al., 2010a; Xie et al., 2012). The highest variability between mea-surements and modelled data are in the midday and are rather constant at night (Ács et al., 2014). Kim et al. (2013) have found that the YSU and MYJ schemes in WRF overestimate, while the ACM2 and MYNN underestimate the PBL height. Their study over Greater Paris indicates that the modelled mean PBLH differs significantly among the schemes and by more than 300 % between the MYNN and YSU. Overestimation of PBLH in chemical transport models, like e.g. WRF-Chem might cause an overes-timation of mixing layer depth and result in an underestimation of modelled pollution concentrations. The latter might be especially relevant for emissions that are released from the surface. Based on the meteograms plotted here, we suggest that the verti-cal extent of the PBL will directly impact the overall concentration of ammonia in the PBL layer. The PBL physics – and therefore also the choice of parametrisation – must therefore affect the ammonia concentrations both at the surface as well as throughout the PBL layer.

The potential higher bias in T2 from the WRF model, reported for Central Europe for summer season by e.g. Skjøth et al. (2015) and Kryza et al. (2015) will impact the mod-elled ammonia concentrations. The Jarczew station, located in this region reveals an increased bias in ammonia concentration for the DYNAMIC simulation in comparison to the BASE simulation during the summer season. However, this increased bias for the DYNAMIC simulation is also present for the spring season. This suggests that the

bias in ammonia concentrations is not related to a bias in temperatures that is mainly seen during summer. Most likely the reason is a combination of a too coarse grid in WRF-Chem and how the ammonia emission is distributed between different emission sectors implemented into the dynamic emission model, which are then affected by the meteorological factors. Several studies showed that significant differences may occur between measured and modelled ammonia concentrations due to the grid size resolution. Especially, the highly localized nature of NH_3 emissions is causing this difference (Dore et al., 2007; Van Pul et al., 2009).

Overall, the results suggest the disagreements between the model and the observations in the hourly profiles are mainly related to emission of ammonia and dilution processes (e.g. increased PBL height and chemical conversion). The results with the aerosols suggest that the high night-time concentrations are not due to release of ammonia from nitrogen containing aerosols. The most likely cause of these high concentrations is that the flux of ammonia from the surface is too high during the night time. The flux of ammonia from agricultural sources away from the surface is mainly dependent on two processes – direct emission due to volatilization (e.g. higher temperatures give higher emission) and the effect of turbulence. The parametrisation we have used does not provide an increase of 100 % in emission from manure that is applied to the field as it is directly linked to previous studies with the ALFAM model (Gyldenkærne et al., 2005; Skjøth et al., 2004) and the parameterisation does not take into account turbulence at the surface but instead uses wind speed data from the ALFAM model (Skjøth et al., 2004). The combination of the temperature and wind speed effect has the consequence that there will be a continuous release of ammonia during night-time even during low temperatures and low wind speeds. Therefore, it must be expected that this limitation causes a redistribution of emission from day to night-time thereby reducing the diurnal emission profile causing a lower daytime peak. If this hypothesis is correct, then new field experiments as a replacement of older experimental data from the ALFAM would be appropriate. Such observations could be used to update the ef-

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fect of environmental parameters on the volatilization of ammonia from fields within the emission model.

Application of the dynamic ammonia emission into the WRF-Chem model results in the improvement of modelled daily ammonia concentrations in comparison to the static approach, which is currently widely used in CTMs. All mean annual statistics based on daily values taken from all the stations have improved when moving from the BASE to the DYNAMIC approach. The smallest differences between both simulations are seen during the summer period, which is likely due to the fact that e.g. the application of manure or fertilizers is limited during this period. For other periods the changes are significant. Due to the implementation of the meteorological conditions in the DYNAMIC approach, a significant proportion of the emission was moved from the winter period to spring, giving much better model performance for winter and a more reliable pattern of daily ammonia concentrations for spring. However, the spring modelled concentrations are overestimated, which most likely can be improved by replacing the Europe-wide default setting, used here after Skjøth et al. (2011) with national practice and regulations for individual countries, as suggested by Werner et al. (2015). Studies of the negative effects related to high ammonia have shown that the plants sensitivity to ammonia might depend on the evolution of the plants and hence can vary throughout the seasons (Sheppard et al., 2009). If CTMs are going to be used for environmental assessments that focus on the seasonal and also the short term variations of ammonia, then it is recommended to adjust the Europe-scale settings in the model with nation-scale settings over the target areas.

The spatial distribution of ammonia emission varies substantially between the BASE and DYNAMIC simulations. This difference is especially noticed for countries with high ammonia emissions. High ammonia concentrations (above $10\,\mu g\,m^{-3}$) occur during the winter period obtained with the BASE simulation for northern Italy, northern France, Germany and Poland. These high concentrations are caused by both emissions (higher than in the DYNAMIC simulation) and meteorological conditions, such as low PBL height that decrease dilution and low temperatures that decrease chemical conver-

sion. In April, for the same areas, much higher concentrations are modelled with the DYNAMIC simulation than with the BASE simulation. This is related to high ammonia emissions during that period caused by application of manure and fertilizers in growing crops. Simultaneously for April, the DYNAMIC simulation calculated the highest ammonia concentrations for many European regions, presented in this study as the number of day with the highest hourly concentrations. Similar findings on spatial variability were also documented by Hamaoui-Laguel et al. (2014), for a study conducted over France. Their study focused on the effect of emission from mineral fertilizers and these results showed that the spatial pattern was highly dependent on actual meteorological conditions. These results using the CHIMERE model, the results by Wen et al. (2014) with STILT-Chem, our results with WRF-Chem as well as previous studies with this emission model all suggest that it is important to have a direct connection between hourly meteorological variables and the level of ammonia emission.

Our study does not include bi-directional exchange, which can further influence the modelled ammonia concentrations. Recent study provided by Zhu et al. (2015) suggests that although the implementation of bi-directional exchange leads to a better fundamental description of NH$_3$ emissions from fertilizers, it does not uniformly improve estimation of NH$_3$ concentrations, NH$_4^+$ wet deposition and nitrate aerosol concentrations. However, Bash et al. (2013) reported that an implementation of bi-directional exchange of NH$_3$ improved the simulations of NH$_x$ wet deposition and improved the simulation of ambient nitrate aerosol concentrations for the US. Wichink Kruit et al. (2012) showed that with the new description in the LOTOS-EUROS model, which includes bi-directional surface–atmosphere exchange, the modelled ammonia concentrations increase almost everywhere, in particular in agricultural source areas. The reason for this is that by using a compensation point the ammonia lifetime and transport distance is increased. A comparison with measurements shows that the model results better represent measured ammonia concentrations; however the concentrations in nature areas are slightly overestimated, while the concentrations in agricultural sources are underestimated.

Our study indicates that the current description of the diurnal cycle of the ammonia concentration from fields is not sufficiently accurate and more research is needed in order to improve the processes that describe the emission from fields. The results suggest that the governing processes in relation to the diurnal cycle are the atmospheric mixing and the stronger daily pattern of ammonia emission, e.g. through increased evaporation or increased fluxes from the surface. The latter is still quite difficult to observe as only very few sites measure ammonia concentrations with a sufficiently high temporal resolution to that required for a proper evaluation and developments of better emissions models. New field experiments as a replacement of older experimental data would be appropriate in order to update an observational assessment of the emission process parameterised in the model. This is one of the objectives of the ECLAIRE project.

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Table 1. Model components and configuration.

Category	Model setup
Simulation period	Jan–Dec 2012
Domains	Europe, 161×131 grids
Horizontal resolutions	$36 \, \text{km} \times 36 \, \text{km}$
Vertical resolution	48 layers
Shortwave and Longwave radiation	RRTMG
Land-surface model	Noah LSM
Boundary layer scheme	YSU
Cumulus parameterization	Grell and Denvenyi (2002)
Microphysics	Lin et al. (1983)
Chemistry	RADM2 and MADE/SORGAM with aqueous reactions

Please refer to the WRF and the WRF-Chem user's guides for a complete description of the options.

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Table 2. Mean statistics based on hourly data (N) for Harwell for 2012: FAC2 (factor of two), MB (mean bias), NMB (normalised mean bias), RMSE (root mean squared error), R (correlation coefficient).

Season	N	FAC2	MB $\mu g\,m^{-3}$	NMB $\mu g\,m^{-3}$	NMGE $\mu g\,m^{-3}$	RMSE $\mu g\,m^{-3}$	R
			BASE				
winter	1491	0.60	0.44	0.60	0.98	1.31	0.36
spring	1949	0.47	0.91	0.35	0.82	2.94	0.44
summer	1156	0.36	0.35	0.33	0.97	1.33	0.22
autumn	1523	0.50	0.56	0.69	1.02	1.15	0.26
			DYNAMIC				
winter	1491	0.64	−0.12	−0.16	0.52	0.63	0.45
spring	1949	0.47	2.26	0.87	1.23	5.60	0.57
summer	1156	0.35	0.52	0.49	1.07	1.50	0.28
autumn	1523	0.51	0.46	0.56	0.94	1.06	0.24

Table 3. Mean statistics for NH$_3$ concentrations based on daily in (N) observations from 6 sites (Harwell, Jarczew, Risø, Tange, Ulborg and Anholt) for 2012.

	N	FAC2	MB $\mu g\,m^{-3}$	NMB $\mu g\,m^{-3}$	NMGE $\mu g\,m^{-3}$	RMSE $\mu g\,m^{-3}$	R
BASE	2020	0.38	0.93	1.10	1.28	1.76	0.55
DYNAMIC	2020	0.42	0.85	1.00	1.23	1.94	0.66

Table 4. Mean statistics split into seasons for NH$_3$ concentrations based on daily observations (N) from 6 sites (Harwell, Jarczew, Risø, Tange, Ulborg and Anholt) for 2012.

Season	N	FAC2	MB $\mu g\,m^{-3}$	MGE $\mu g\,m^{-3}$	NMB $\mu g\,m^{-3}$	NMGE $\mu g\,m^{-3}$	RMSE $\mu g\,m^{-3}$	R
BASE								
winter	502	0.22	1.18	1.21	3.65	3.75	2.10	0.34
spring	539	0.46	1.34	1.60	0.87	1.04	2.34	0.57
summer	491	0.46	0.57	0.72	0.69	0.86	0.98	0.60
autumn	488	0.37	0.58	0.76	0.92	1.19	1.09	0.46
DYNAMIC								
winter	502	0.38	0.20	0.34	0.61	1.06	0.49	0.42
spring	539	0.46	1.71	1.97	1.11	1.28	3.22	0.66
summer	491	0.49	0.56	0.71	0.67	0.85	0.97	0.63
autumn	488	0.33	0.84	1.06	1.32	1.65	1.72	0.27
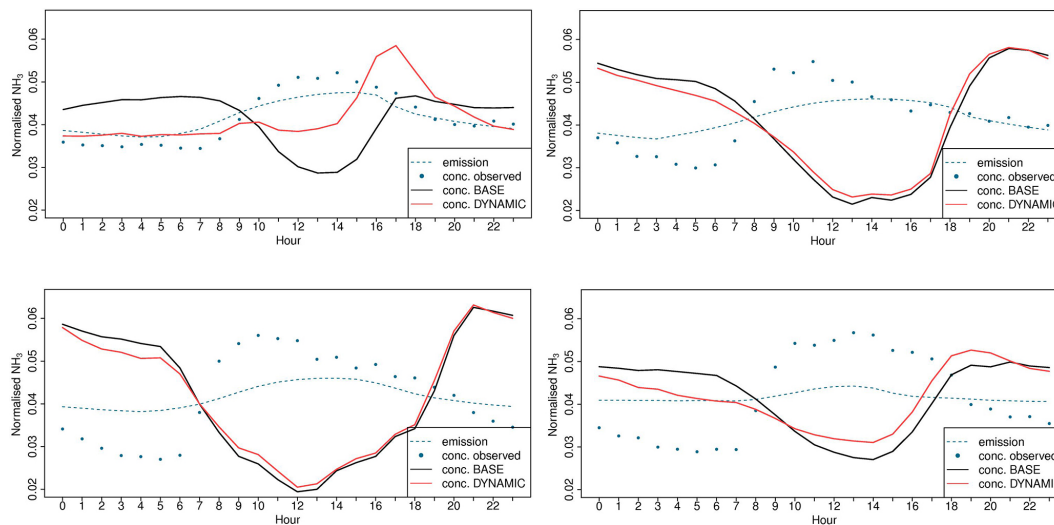
Figure 1. Normalised NH$_3$ concentrations and emission according to hours for four seasons for the grid corresponds to the Harwell station. Normalisation procedure: the sum of emission/concentration for each individual hour (0–23) was divided by the total sum of emission/concentration in the season.
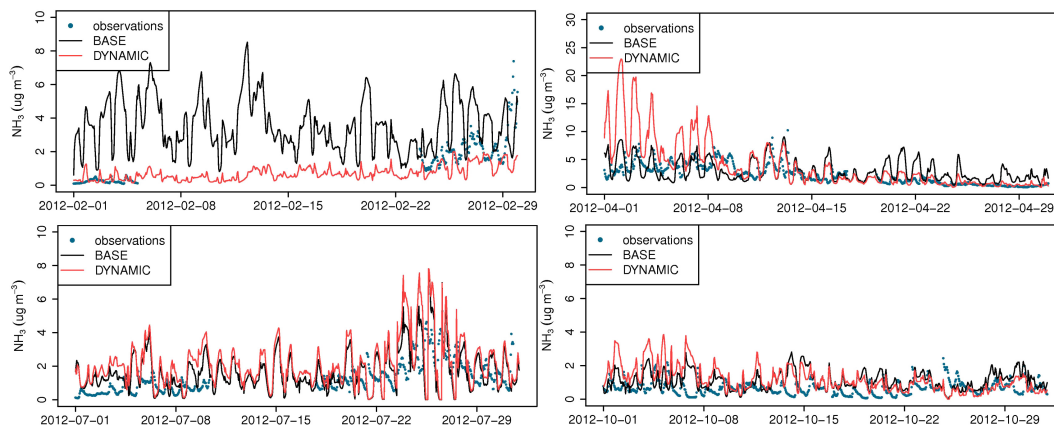
Figure 2. Modelled and observed hourly NH$_3$ concentrations for Harwell for February, April, July and October 2012. Please notice there is a different y scale for April.
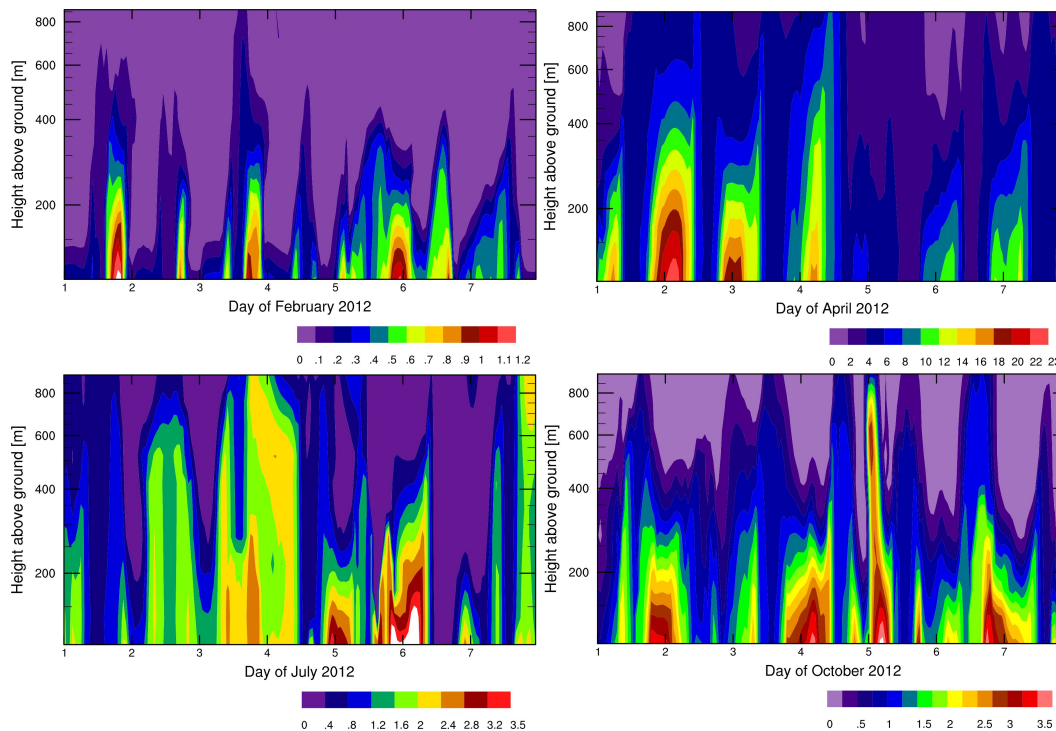
Figure 3. Temporal and vertical distribution of NH₃ concentrations [µg m⁻³] for the DYNAMIC scenario.
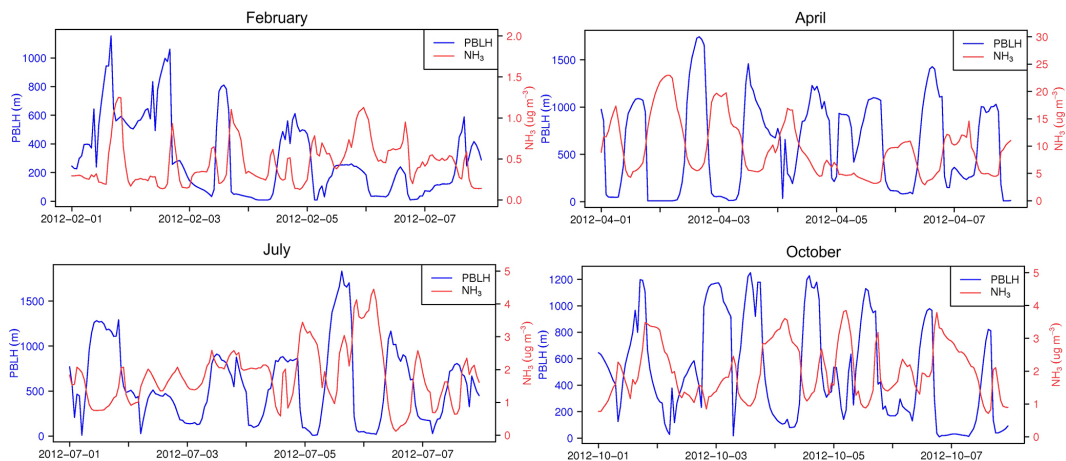
Figure 4. Time series of modelled (DYNAMIC) PBLH and NH₃ concentrations for the episode of February, April, July and October 2012.
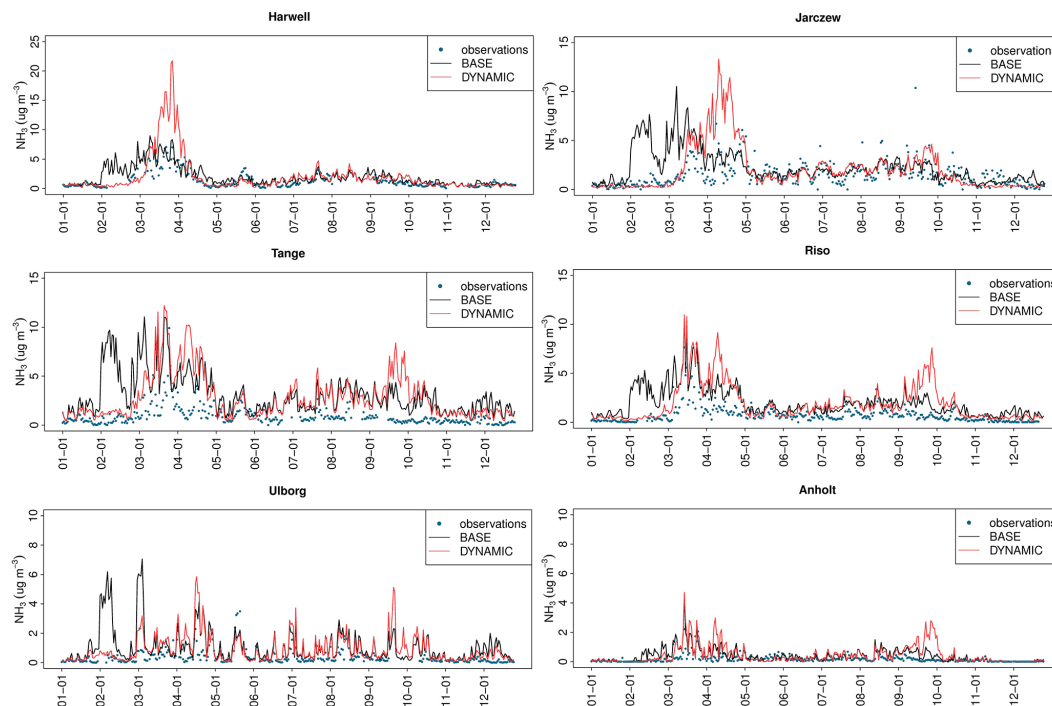
Figure 5. Modelled and observed daily NH$_3$ concentrations for 2012. Different *y* axis scales are used for the different stations.
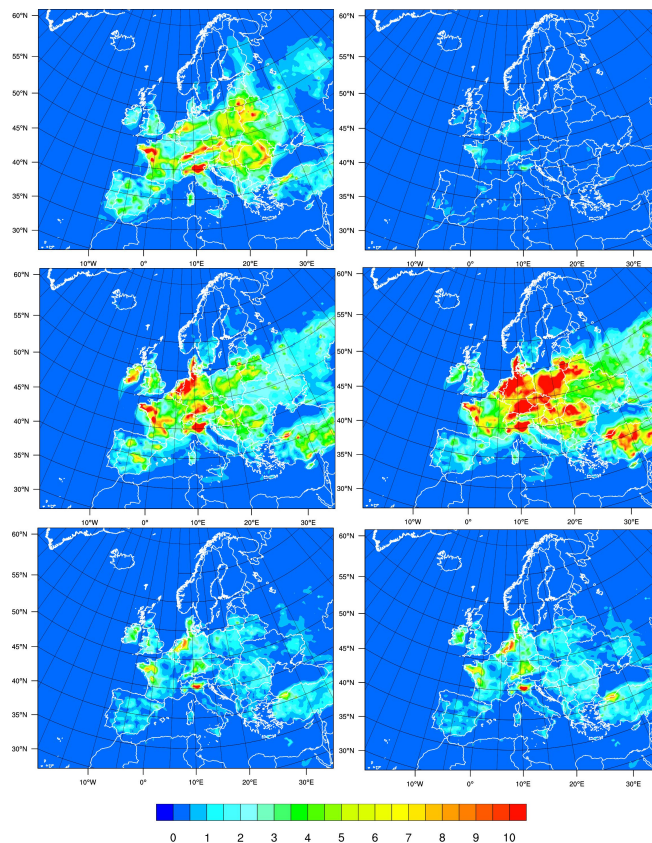
Figure 6. Spatial distribution of NH$_3$ concentrations on 15 February (upper), 15 April (middle), and 15 October (lower) 2012 at 12.00 a.m. BASE scenation – left column, DYNAMIC scenario – right column. Units: µg m^{-3}.

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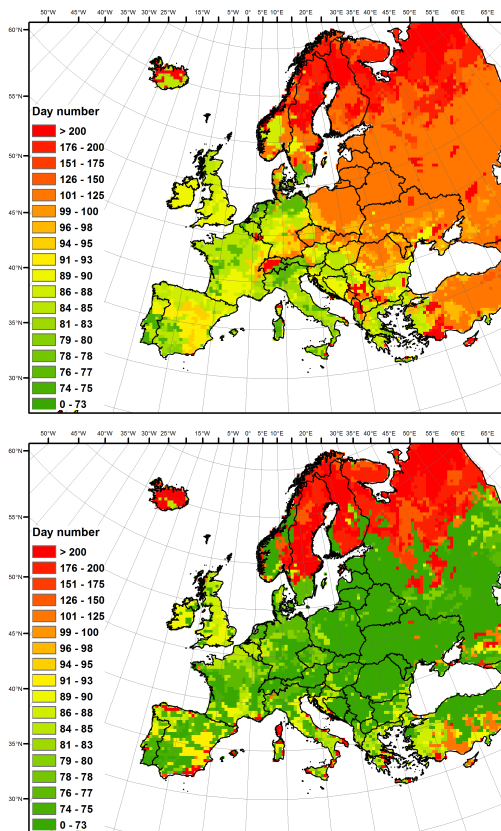
Figure 7. Julian day number in the year 2012, for which the model calculated the highest hourly ammonia concentrations – upper figure for DYNAMIC and lower for BASE.
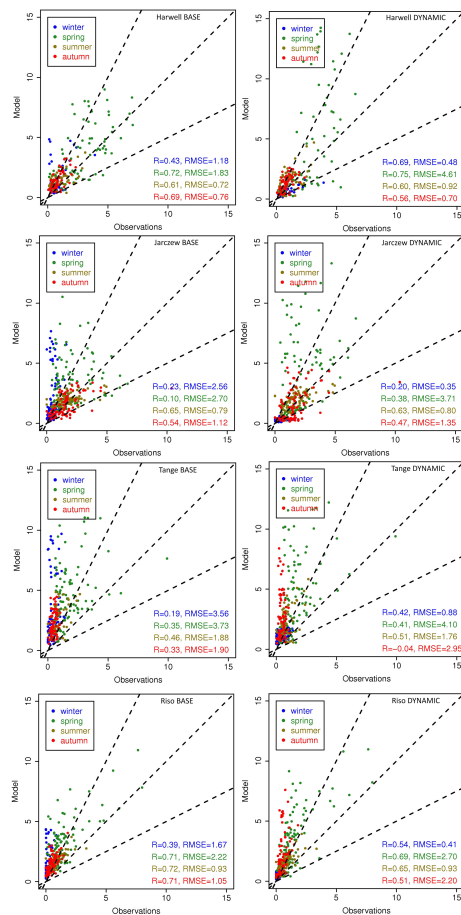
Figure 8. Scatter plots of NH₃ concentrations based on daily values for individual sites for the BASE (left) and DYNAMIC (right) simulations and according to seasons.