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Effect of climate and climate change on ammonia emissions

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The effect of climate and climate change on ammonia emissions in Europe

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Abstract

We present here a dynamical method for modelling temporal and geographical variations in ammonia emissions in regional scale Chemistry Transport Models (CTMs) and Chemistry Climate Models (CCMs). The method is based on the meteorology in the models and gridded inventories. We use the dynamical method to investigating the spatio-temporal variability of the ammonia emissions across part of Europe and study how these emissions are related to geographical and year-to-year variations in atmospheric temperature alone. For simplicity we focus on the emission from a storage related to a Danish standard pig stable with 1000 animals and display how the emission from this source category vary geographically throughout central and northern Europe and from year to year. In view of future climate changes we also evaluate the potential future changes in the emission by including temperature projections from an ensemble of climate models. The results points towards four overall issues: (1) Emissions can easily vary with 20 % by changing geographical location within a country due to overall variations in climate. Largest uncertainties are seen for large countries like UK, Germany and France. (2) Annual variations in overall climate can at specific locations cause uncertainties in the range of 20 %. (3) Climate change will in general increase the emissions with 0–40 %, in central to northern Europe. (4) Gradients in existing emission inventories that are seen along country borders (e.g. between UK and France), can be reduced by using a dynamical methodology for calculating emissions.

Acting together these four issues can cause substantial uncertainties in emission. Emissions are generally considered among the largest uncertainties in the model calculations with CTM and CCM models. Efforts to reduce uncertainties are therefore highly relevant. It is therefore recommended that both CCMs and CTMs implement a dynamical methodology for simulating ammonia emissions in a similar way as for biogenic volatile organic compound (BVOCs) – a method that has been used for more than a decade in CTMs.

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1 Introduction

Ammonia plays an important role in many atmospheric processes. It is the main alkaline component in the atmosphere and is highly reactive in forming either aerosols (e.g. Seinfeld and Pandis, 2006) or by depositing fast to most surfaces including sensitive ecosystems (Sutton et al., 2007). Since its acknowledgement as an environmental pollutant, much effort has been put into understanding the fate of ammonia including the emission to the atmosphere and the following environmental effects (Sutton et al., 2008; Sutton et al., 2011b). Ammonia is emitted from wild animals (Sutton et al., 2000) ammonia-containing water areas (Barret, 1998; Sørensen et al., 2003), traffic (Kean et al., 2009), sewage systems (Reche et al., 2012), humans (Sutton et al., 2000) and agriculture. Here agriculture is considered the main source to emissions of ammonia globally (e.g. Bouwman et al., 1997) and regionally in e.g. Europe (Sutton et al., 2011b) or USA (Pinder et al., 2004). Emissions of ammonia cause considerable concentrations near strong agricultural sources (Fowler et al., 1998; Geels et al., 2012; Hallsworth et al., 2010; Kryza et al., 2011), but the overall ammonia concentrations are quickly reduced to a low background level (de Leeuw et al., 2003), as ammonia is dispersed and incorporated into aerosols. These aerosols typically contribute with 30 % to 50 % of the total aerosol mass of PM_{2.5} and PM₁₀ (Anderson et al., 2003). Ammonia containing aerosols are therefore a very important component in regional and global aerosols processes (Xu and Penner, 2012), as they may dominate the overall amount of aerosols in the PM_{2.5} and PM₁₀ fractions. It has also been suggested that ammonia containing aerosols are relevant for human health (Aneja et al., 2009), although the Nature commentary by Sutton et al. (2011b) highlights the high uncertainty on health effects that are related to ammonia emissions. Ammonia also contributes to acidification and eutrophication of natural ecosystems leading to e.g. biodiversity changes and reduced plant species richness (Krupa, 2003; Stevens et al., 2010). In Europe, more than 90 % of the emitted ammonia originates from the agricultural sector (Reis et al., 2009). Here the main sources are livestock, manure management and application of

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fertilizer (Reis et al., 2009; Skjøth et al., 2011). Nearly all ammonia emissions are due to volatilization of ammonia from wet surfaces (Elzing and Monteny, 1997; Gyldenkærne et al., 2005). Volatilization of ammonia is a physical process that is highly temperature dependent (Gyldenkærne et al., 2005). The temperature in and above the emission sources such as manure in the field (Sommer et al., 2003) or buildings dynamically follows the changes in air temperature and wind velocity from hour to hour and throughout the season (Gyldenkærne et al., 2005). Due to this temperature effect of local meteorology, hot years are likely to have higher ammonia emissions compared to cold years. Similarly, then hot days are likely to cause higher emissions than cold days. This temperature effect is according to a recent review by Menut and Bessagnet (2010) currently not taken into account by Chemistry Transport Models (CTMs). This is also the case for Chemistry Climate Models (CCM). CTMs and CCMs could therefore be improved by including a dynamic description of the processes that cause variations in ammonia emissions (Gyldenkærne et al., 2005; Hellsten et al., 2007; Pinder et al., 2006; Skjøth et al., 2004; Skjøth et al., 2011).

State-of-the-art Chemistry Transport Models such as DEHM (Brandt et al., 2012; Geels et al., 2012), EMEP (Simpson et al., 2012), TM5 (de Meij et al., 2006), CHIMERE (de Meij et al., 2009), MATCH (Langner et al., 2009; Langner et al., 2012) and LOTUS-EUROS (Barbu et al., 2009) rely on emissions from the EMEP system or similar sources with gridded emission inventories. These gridded emission inventories are often based on national emission factors. For ammonia emission inventories, then certain agricultural production methods or activities are related to specific emission rates that are obtained under specific climatic conditions within a country (Klimont and Brink, 2004). However, throughout even small countries like Denmark, certain areas have a warmer or colder climate compared to the mean across the country (Cappelen, 2011). Such spatial variations in climate will cause spatial variations in ammonia emissions from otherwise identical sources. This climate effect due to temperature differences will likely be much more pronounced for larger countries like Germany, France and the UK as these countries have larger national variations in climate compared to Denmark or

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Netherlands. To our knowledge, this spatial temperature effect has not previously been explored. It is therefore not known, if this effect is important for NH_3 emission models and the CTM models. But it is known that CTM models needs accurate emissions and accuracy of the NH_3 emissions are considered among the most important factors for improvement in relation to atmospheric chemistry and nitrogen compounds (Simpson et al., 2011). The regional variations of climate on ammonia emissions therefore need to be explored, and the governing processes that cause these variations must be included in CTM and CCM models.

The effect of ammonia on climate has recently been discussed (de Vries et al., 2011; Erisman et al., 2011). This also includes model calculations of ammonium containing aerosols (Xu and Penner, 2012). However, the impact of climate change on future ammonia emission was not studied, thus neglecting a potential feed-back mechanism between the nitrogen cycle and climate change. Such feed-back mechanisms in the climate-system that involve the nitrogen cycle have in a review in Nature Geosci. by Arneeth et al. (2010) been highlighted as a critical component for biosphere-atmosphere processes. According to Arneeth et al. (2010) then these effects remains to be studied by the climate models. Current climate models that are used in the IPCC report predict an increase in temperature in main agricultural sectors such as Europe and United States (IPCC, 2007). The predicted increases in temperature in these high emitting regions vary from location to location. Future ammonia emissions from these high emitting regions can therefore be expected to increase alone due to the temperature. This effect can also be expected to vary from region to region. How much these increases affect the feed-back mechanisms in the climate-system is currently not known.

In this study we will investigate how much ammonia emissions can change geographically and annually in central and Northern Europe due to variations in temperature alone. We will also investigate the potential effect of climate change on these ammonia emissions by using simulated surface temperatures from seven different climate models within the ENSEMBLES project. The tool for these investigations is an modified

version of the open-source dynamical NH₃ emission model Skjøth et al. (2011) with a spatial coverage from central Sweden to Northern Italy.

2 Methodology

Hourly NH₃ emissions are simulated using the dynamic model in an identical model domain as in Skjøth et al. (2011). Here we are studying the years 2006–2010, 2046–2050 and 2086–2090 using a fixed set of emissions for the year 2007 from Skjøth et al. (2011) to study how meteorology from year to year can change both amount and temporal variation of ammonia emissions. Similarly we superimpose the climatic signal from climate models on the meteorological data to also study the effect of climate change on ammonia emissions. Meteorological input data are gridded reanalysed fields for the years 2006–2010 and climate model simulations for the period 1961–2099. Additionally we isolate the climatic signal (geographically and temporally) by investigating the variations in emissions throughout Europe from a standard storage facility from a farm with 1000 pigs.

2.1 Meteorological input data

The emission model has in this study used an input of meteorological fields of 2-m air temperature and 10-m wind speed. The data are reanalysed meteorological data from the MM5 meteorological model (Grell et al., 1995) in an identical setup as in Skjøth et al. (2011) that provides 1 h temporal resolution and 16 km spatial resolution of the meteorological fields. Here we use meteorological data for the years 2006–2010. The reanalysed meteorological data with 1 h resolution have also been used in combination with long term trends in monthly mean temperatures obtained from the results in an ensemble of regional climate models. The input from the climate models is a gridded data set of monthly temperatures based on the ensemble mean from 11 scenarios run by eight different regional scale climate models obtained from the ENSEMBLES

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project (<http://ensemblesrt3.dmi.dk>). This combined data set from ENSEMBLES and the MM5 model have been used to construct an hourly temperature data set for the years 2046–2050 and 2086–2090 (Appendix Fig. A1) using the following procedure: First, the results from the climate models data have been re-projected to match the 16 km grid resolution in the meteorological data from the MM5 model. Then, a sliding mean has been calculated for the climate model results. This sliding mean has been calculated for each re-projected grid cell. The sliding means are calculated separately for each month (e.g. January 2011, January 2012, January 2013, etc.), to take into account seasonal trends in the results from the climate models. The result is a series of trends in two-meter temperatures, which vary between grid cells and vary throughout the season. The trends are then superimposed to the hourly gridded reanalysed meteorological data from the MM5 model. This procedure ensures that the long term temperature trend from the climate model scenarios are kept for the entire period and that the variations in these trends throughout the seasons also are kept. Likewise, the method ensures that the detailed hourly and spatial variation given by the MM5 model is maintained (Fig. A1 in the Appendix) and that the year to year variability seen over a five year period also is included. Finally the procedure corrects any bias in the surface temperatures that are present in the climate model results. It is well known that bias correction of climate model data is needed before they can be used as input to e.g. impact models or vegetation models (Dosio and Paruolo, 2011). By using trends from the climate model data, we have assumed that the overall bias in the results set from the climate models is the same throughout the simulation period.

2.2 Simulations with the Dynamical Ammonia Emission Model

The Dynamic Ammonia Emission Model use 15 different emission functions to describe the temporal variations of different agricultural activities (Gyldenkærne et al., 2005; Skjøth et al., 2004, 2011). The emission model also use an inventory of gridded NH₃ emissions (e.g. EMEP, EDGAR or national inventories), in combinations with information on agricultural activities. On European scale the inventory is based on a

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redistribution of the officially reported EMEP emissions and national numbers for a distribution of agricultural activities that contributes to the overall ammonia emission load. These numbers are obtained from Table 2 in Skjøth et al. (2011) to provide a gridded estimate for all 15 source categories. The Dynamic Ammonia Emission Model is then used to calculate a temporal variation of ammonia emissions for all 15 categories with one hour temporal resolution. Each of these 15 categories require a normalization factor (Skjøth et al, 2004) to ensure consistence with official annual emissions values. This normalisation factor will vary between grid cells and from year to year, depending on actual meteorology (Skjøth et al, 2004). Warm years will have higher normalisation factors than colder years. Here we have calculated the mean normalisation factor for the years 2006–2010 in each grid cell and used this as a reference in all subsequent calculations. So for the years 2007, 2047, 2087, etc. we use the actual meteorology for that particular year (e.g. 2047) and then use the mean normalization factor for the period 2006–2010. This allows us to assess annual variations in total emissions due to climate alone in each grid cell (e.g. warm years will have increased emissions etc.). The advantage of this method is that it allows us to isolate the climate signal (temporally and spatially). Secondly, the updated model procedure simplifies the model calculations significantly as a full pre-calculation run is not needed. The reason is that the annual normalization factor is removed in order to allow for annual fluctuations in the total emission. In this study we use the well-studied Tange area in Denmark as the reference site in order to investigate the spatio-temporal variability of the ammonia emissions. In Fig. 1 the ammonia emission from our standard storage located in the Tange area is displayed as a function of air temperature. We have here chosen the standard storage to be a storage related to a pig stable facility with 1000 animals with the same production methods as in Denmark. The applied temperature dependence is described in more detail in (Skjøth et al., 2004), but from Fig. 1 it can be seen that the emission stops at 0 °C (due to freezing) and thereafter increases strongly with increasing temperatures.

The emission model is then used for two scenarios:

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- Scenario one: A scenario to investigate how much ammonia emissions can change geographically over Central and Northern Europe for the years 2007 and 2010 from storage that is associated to a standard pig stable. For the storage at the Tange site in Denmark this gives a mean annual emission of 480 kg NH₃ (Klimont and Brink, 2004).
- Scenario two: A scenario on how much ammonia emissions may vary due to increasing temperatures under climate change by repeating the calculations above, but for the years 2047, 2050, 2087 and 2090, respectively.

3 Results

3.1 Scenario one

Figure 2 show maps of gridded annual emission of NH₃ from storage facilities for the years 2007 and 2010.

Figure 2a is the annual emission for the year 2007, which represents a relatively warm year in central Europe under current day conditions. Figure 2b is the annual emission for the year 2010, which represents a relatively cold year under current day conditions. In both years, substantial geographic differences are seen in the emissions from our standard storage facility. These geographical differences in emissions are in the current setup entirely driven by differences in air temperature. One result is an overall north-south gradient with lowest emissions in the northern part of the domain and highest emissions in the most southern part of the domain. These gradients are observed for both years but vary in absolute amount between the years. An area of low emissions is in both years seen in the mountainous regions of the Alps in the southern part of the domain following the lower temperatures with altitude. Largest geographical differences in emissions are seen in the warm year of 2007, where our standard storage facility would have an annual emission of ca. 300 kg NH₃-N yr⁻¹ in southern Norway and up to 540–620 kg NH₃-N yr⁻¹ in parts of UK, Benelux, Germany, northern France

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and northern Italy. These emission rates are exceeded in smaller areas with a possible emission of 620–700 kg NH₃-N yr⁻¹. Within a single country like Germany a difference in emission from about 460 to 700 kg NH₃-N yr⁻¹ is seen. In the colder year of 2010 the standard storage would generally emit less throughout the entire domain, with only a few places where the emission from the storage could reach 540–620 kg NH₃-N yr⁻¹.

3.2 Scenario two

Figure 3a, b, c, d is the annual emission for our standard storage facility for the year 2047, 2050, 2087, 2090, respectively. They represent relatively cold (2050 and 2090) and warm (2047 and 2087) years under future conditions according to the applied ensemble of climate models. In Table 1 the maximum and minimum emission rates within the countries are given for the warm years 2007, 2047 and 2087.

In all years substantial geographic differences are seen in the emissions. These differences are as in scenario one solely driven by geographical differences in air temperature. For all calculations a similar overall north-south gradient is seen in the emission pattern as for the years 2007 and 2010. Largest geographical differences are seen in the warm year of 2087, where our standard storage would have an emission rate of ca. 300–380 kg NH₃-N yr⁻¹ in southern Norway and up to 800–900 kg NH₃-N yr⁻¹ in parts of UK, Benelux, Germany, northern France and northern Italy (Table 1). Large areas with a possible emission of 620–700 kg NH₃-N yr⁻¹ are also seen throughout Germany, Denmark, Poland and England. Within a single country like Germany a difference in emission from a minimum of about 294 to a maximum of 775 kg NH₃-N yr⁻¹ is seen in the warm year of 2087.

In the colder year of 2050 the standard storage would generally emit less throughout the domain, with only a few places where the emission from the storage could reach 620–700 kg NH₃-N yr⁻¹. A similar pattern in the emission rates is seen for the year 2090, although the areas with emissions in the range from 620–700 covers most of North Italy, northern France and southern England.

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4 Discussion

4.1 Emission variations under existing climate

The model results show that if farmers use identical production methods, then meteorology and climate considerably affects the ammonia emission from this otherwise identical production. The model results (Fig. 2) for the historical years 2007 and 2010 are based on an input of reanalysed meteorological fields of air temperature to simulate emissions from this standard storage in a pig production. The results show that the mountainous regions (Norway and the Alp region) and the eastern part of the model domain (Poland, Lithuania, Russia and Belarus) had almost the same or slightly lower emission in 2010 compared to 2007. The central part: England, Netherlands, central and Northern Germany and Denmark had between 15 % and 25 % higher emissions in 2007 compared to 2010. This difference is the variation that is seen between cold and warm years at a specific location. Likewise, the emission from a storage facility can change considerably depending where it is located within a country, again due to gradients in the air temperature. This is of course more pronounced for large countries like Germany, where the emission from storage within the country can change by as much as 40 % alone due to differences in prevailing climate in different regions. In relation to official emission inventories such as the ones that are reported to EMEP or detailed national inventories (e.g. Velthof et al., 2012), then this is an important result. In EMEP as well as in the more detailed national inventories, the total ammonia emission is a product of the amount of animals and production methods by using emission factors for e.g. housing or storage (Velthof et al., 2012). As such even the more advanced emission models do not fully take into account climate and climate variability as the overall concept still rely on standard emission factors. These factors are applied in the model calculations and often there is one national emission factor per country (e.g. Klimont and Brink, 2004) for each activity. These emission factors are then used for a number of years before they are updated. This is clearly shown by the differences in the emissions factors for UK and France in Klimont and Brink (2004), where the

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emission factors is more than 50 % larger for storage in France compared to the UK. Graphically the methodology of relying on fixed emission factors and amounts of animals can result in large gradients in emissions between the regions that are covered by statistics as seen in Fig 5 by de Vries et al. (2011). Some of these gradients are due to variations in animal density, but the gradients are probably also due to model uncertainty. This uncertainty is both due to the fact that statistical information like animal numbers are available only on coarse scale, but also due to the fact, that the entire emission inventory system relies on fixed emission factors that does not directly take into account meteorological variables (Beusen et al., 2008; Pouliot et al., 2012). As such, the uncertainties we here present will affect the results from CTM models as well as results from CCM models through atmospheric chemistry and aerosol processes such as radiation. How large the uncertainty is on European scale or global scale and the associated effect on chemistry and climate is not known. The methods we have shown here are directly available for these types of models and will make it possible to assess the uncertainty.

4.2 Emission variations under future climate

The model results show that when climate and climate change projections are taken into account, then effect of temperature on ammonia emissions becomes even more pronounced. The results suggest that many areas can expect an increase in ammonia emissions from typical agricultural sources of up to 60 % from the year 2010. The increase varies from year to year and from location to location. The reason is that the results from the climate models show different temperature increases throughout central and Northern Europe. This result is also important as CTM models and CCM models that are used to study short-lived climate forcers require accurate emissions. In the latest years there has been increased scientific focus on emissions from biogenic volatile organic compounds (BVOCs) as they are a precursor for short lived climate forcers. Ammonia is the main alkalic component in the atmosphere and important component in the formation of aerosols (Xu and Penner, 2012). An increase in surface

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temperatures will enhance ammonia emissions. This will affect the amount of aerosols causing a feedback in the climate system. Additionally the increased amount of atmospheric ammonia is likely to end up in terrestrial or marine ecosystems, thus affecting the C-N cycle, which again is an important component of the climate system and a source to feed-back mechanisms. To our knowledge – the importance of the feedback between increased temperatures and increased ammonia emission in relation to the climate system – has not been studied. Again, the methods that are presented here can be implemented in CTM models as well as CCM models for climate change studies on atmospheric chemistry, radiation or the C-N cycle.

4.3 Overview of sensitivity of ammonia emissions in relation to increased temperatures and climate change

In Europe, more than 90 % of the total estimated ammonia emissions originate from Agriculture. In Table 2 we give an overview of the different agricultural sources to ammonia and their sensitivity to climate. Also the fraction that the individual sources contribute to the total agricultural emission in Europe is given. This is done in order to illustrate the overall sensitivity of the European emissions to changes in climate. Between 34 % and 43 % is estimated to originate directly from buildings and storage. The emission from these source categories are all sensitive to climate and climate change, which is not taken into account in current emission estimates (e.g. Velthof et al., 2012). The same is true for manure handling, mineral fertilizer and grazing animals that contribute from 6 % and up to 26 % of the total emission. In some emission estimates the overall climate in a region is used to modify emission factors e.g. Beusen et al. (2008). However, to our knowledge, then a dynamic response on emissions from meteorology and climate is not taken into account as this requires a direct coupling to weather variables. The emission model we have used here has been shown to be able to describe seasonal as well as daily pattern of emissions from a number of different agricultural activities (Geels et al., 2012; Gyldenkerne et al., 2005; Skjøth et al., 2004, 2011). Isolated studies on regional and local scale have shown that a main limitation of the

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precision is the accuracy of the total emissions as well as the definition of the agricultural activities (Sommer et al., 2009). The simple redefinition of the activities from overall mean pattern into one of the 15 activity functions given by Gyldenkærne et al. (2005) has significantly improved the models results (Skjøth et al., 2004). It is well known that agricultural production methods vary considerably throughout Europe, such as the use of application methods of fertilizer during spring. The activities that are least affected by production methods and machinery are stables without heating and physical ventilation (typically cattle), storage facilities (pigs and cattle) and grazing animals (cattle, horses and sheep). However, grazing animals will typically be outside during summer and inside the stable systems during winter, which therefore again affect the emissions pattern. These uncertainties only affects pigs production to a small degree, as pigs are typically kept inside the buildings during the entire year in all the studied countries (Klimont and Brink, 2004). We have therefore chosen to study storage from a standard pig stable as this facility will have the most uniform emission pattern throughout Europe due to variations in agricultural production methods. A similar emission pattern can be found from cattle barns and storage from cattle barns. But the emission from cattle barn is slightly more complicated to describe, as cattle is typically kept outside the building during a fraction of the year, which varies from country to country (Skjøth et al., 2011). Emissions from application of manure and mineral fertilizer are just as sensitive as emissions from buildings and storage. However, there is a feed-back mechanism in the climate system from that typical source. Farmers can change their application time due to overall change in climate. This can cause an increased growth period (e.g. due to warmer spring), a change in prevailing crops (e.g. from barley to sunflower in Central and Northern Europe) or due to a change in number of annual crop cycles (e.g. from one to two harvesting periods). Experiments with this kind of change have all been carried out by Danish farmers the last 10 yr and they can all have a large impact on the emissions from manure and mineral fertilizer that have been applied to the fields. This as well as the previous studies by Skjøth et al. (2004) suggest that the key to accurate description of ammonia emissions is to connect the emission model with

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accurate mappings of agricultural activities such as types of production, use of fertilizer and amount of animals.

4.4 Relevance of including dynamical modelling of NH₃ emissions in CCM and CTM models and current possibilities?

5 The next generation of Earth system models such as EC-Earth (<http://eearth.knmi.nl/>) are now under development and used for studies that includes feed-back effects in the climate system (e.g. Bintanja et al., 2012). In EC-Earth this includes radiation, a chemistry model like TM5, an ecosystem model like LPJ-Guess, which also ensures emissions of BVOCs for aerosol production. Our results suggest that Earth system models
10 should also include a model that dynamically calculates ammonia emissions from humans, industry, traffic and agriculture as ammonia emissions are affected by climate. This will induce feed-back effects on atmospheric chemistry and aerosols and hence back-scattering as well as nitrogen deposition, which both may affect ecosystem behaviour (Sheppard et al., 2011). To our knowledge dynamical models for ammonia are not under development in global models, although several Nature papers have stated that the nitrogen issue is probably one of the biggest challenges humans will face in the future under an increased population load (Gruber and Galloway, 2008). Additionally, the effect of climate and ammonia emissions and potential feed-back mechanisms has not been taken into account by the IPCC reports. Recent opinions by Erismann et al. (2011) as well as the European Nitrogen Assessment, chapter 9 (Hertel et al., 2011)
15 does not take this effect into account, probably because the scientific understanding has not been available. A scientific meeting in the Royal Society, 5–6 December 2011 on the global nitrogen cycle had an entire session dedicated to the ammonia issue. Here one of the recommendations was that the scientific community should put efforts into a common vision on ammonia emission modelling for use by CTM and climate models (Sutton et al., 2011a). Our results here suggest that the recommendation by Sutton et al. (2011a) is highly relevant and we propose that such model efforts should
25 use an open source methodology to contribute towards this vision. Another important

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aspect is access to the relevant information from agricultural activities. In Europe, this information can potentially be obtained from modelling approaches that combines data bases such as the Corine Land Cover2000 (European Commission, 2005) with the EU-ROFARM database (Neumann et al., 2009), while global scale studies need more work, e.g. by combing existing inventories with data from FAO and detailed global land cover data. Despite of these complicating factors the emission pattern is relatively stable with respect to two climatic factors: (1) Increased temperatures due to climate change will increase emissions from almost all typical agricultural sources. (2) Regional climatic variations will change emissions from otherwise identical agricultural production facilities alone due to local variations in the prevailing climate.

5 Conclusions

Our studies suggest, that annual variations in meteorology, variations in overall climate between regions and climate change all affects the emission of ammonia substantially. The main reason is that volatilization of ammonia is very sensitive to air temperature. This effect is currently not taken into account in CTM models (Menut and Bessagnet, 2010) as well as in climate model simulations. In fact, Menut and Bessagnet (2010) dedicate an entire subchapter to the ammonia issue in their review of CTM models that are used in forecasting of air quality. Similarly results are obtained from a new model inter-comparison study by Pouliot et al. (2012). Menut and Bessagnet state that the temporal profile is not accurate enough in any of the European models and Pouliot et al. (2012) state the temporal profile in ammonia is not correct in CTM models, when they rely on fixed profiles. We therefore suggest that next generation of CTM models and especially CCM models take this feedback effect between emissions and climate into account by dynamically calculating ammonia emissions within the models.

In relation to ozone and future air quality the term “climate penalty” is often used, which means that stronger emission controls will be needed to meet a given future air quality standard (e.g. Jacob and Winner, 2009). Our results indicate that the same

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term can be used for ammonia as the projected change in climate alone will lead to an increased emission of ammonia. This increased amount of ammonia will affect the known cascade of effects (Galloway et al., 2003) that are related to the nitrogen cycle. This includes effects on aerosols and radiation (e.g. Xu and Penner, 2012), effects on atmospheric chemistry (Seinfeld and Pandis, 2006), effects on ecosystems (e.g. Sheppard et al., 2011) including possible feed-back mechanisms. According to a recent article in Nature by Gruber and Galloway (2008), the feedback mechanisms in the climate system that are related to the nitrogen cycle are poorly understood. In fact, a review in Nature Geosci. by Arneeth et al. (2010) claims, that despite nitrogen is a critical component for vegetation-atmosphere processes, then nearly all possible feed-back processes remains to be studied. This also includes increased ammonia emissions as in our study. This lack of consideration on feedback processes leads to substantial uncertainties (Gruber and Galloway, 2008). Therefore evaluations of future abatement strategies for ammonia need to take the possible effect of a general temperature changes into account as well as the associated cascade of effects in the nitrogen cycle, which is initiated with the emission.

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Table 1. National maximum and minimum annual ammonia emission from the standard pig storage from the gridded calculations.

	2007		2047		2087	
	Min [kg NH ₃ y]	Max [kg NH ₃ y]	Min [kg NH ₃ y]	Max [kg NH ₃ y]	Min [kg NH ₃ y]	Max [kg NH ₃ y]
Germany	208.6	630.5	262.9	716.6	294.4	775.0
Netherlands	536.3	679.4	615.8	766.9	664.6	827.1
Belgium	454.9	633.3	538.5	718.0	586.7	773.3
France	223.9	749.8	283.3	840.1	316.9	900.2
UnitedKingdom	423.8	686.1	509.3	773.5	562.5	831.9
Denmark	484.9	609.6	563.7	695.5	613.1	753.4
Sweden	292.9	540.2	348.8	624.1	379.3	681.1
Poland	370.7	557.2	444.8	639.8	485.6	696.4
Italy	138.3	729.9	181.2	802.8	205.7	852.6
Austria	138.7	529.6	179.3	605.2	205.7	647.0
Switzerland	133.9	516.1	176.6	584.4	204.3	627.5
Czech Republic	367.5	521.5	444.4	598.0	485.3	641.5

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Table 2. The main ammonia emission categories in Europe, an indication of their sensitivity to climatic variables as well as an overview of the contribution to the total emission from agricultural sources in Europe.

Description	Climate sensitivity	Temporal emission pattern	Fraction of European Agricultural Emissions*
Animal houses with forced ventilation and heating	Sensitive in warm regions, less sensitive in cold regions	Follows outside temperatures to some degree	34 %–43 %
Open animal houses with natural ventilation	Sensitive	Follows outside temperatures	
Storages	Sensitive	Follows outside temperatures	
Manure handling	Sensitive	Follows outside temperatures during application	22 %–26 %
Mineral fertilizer	Sensitive	Follows outside temperatures during application	17 %–26 %
Grazing animals	Sensitive	Follows outside temperatures	6 %–10 %
Other sources: Emission from plants	Unknown	Tightly connected to compensation point and ambient NH ₃ concentrations	?
Other sources: waste treatment	Not sensitive		2 %
Other sources: traffic	Not sensitive		2 %
Remaining other sources: e.g. industry	Not sensitive		3 %
Other sources: Water areas	Unknown	Depends on NH ₄ ⁺ concentration in water and ambient NH ₃ concentrations	?

* Numbers are based on calculations from the European Nitrogen Assessment, chapter 15 by DeVries et al. (2011) as well as sector based emissions from EMEP extracted the 10 May 2012.

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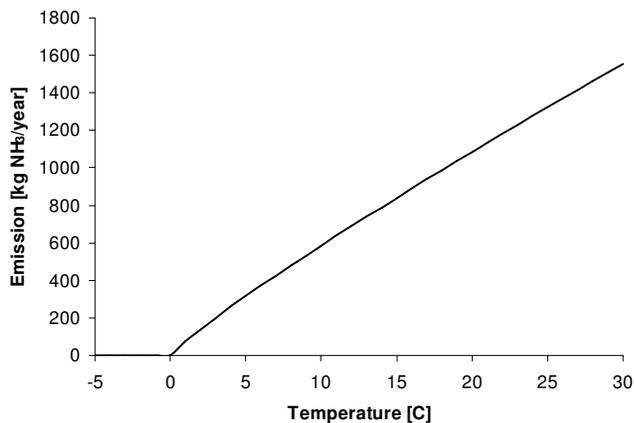


Fig. 1. Modelled annual ammonia emission from storage as function of temperature from a pig production facility with 1000 animals and production methods as in Denmark.

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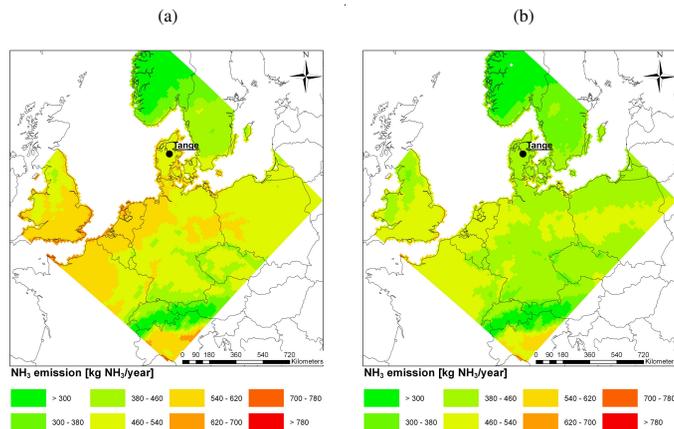


Fig. 2. Annual emission from storage in a reference pig production facility (located at Tange) with 1000 animals, when the facility is placed throughout central Europe and exposed to different European climatic conditions in the years **(a)** 2007 and **(b)** 2010.

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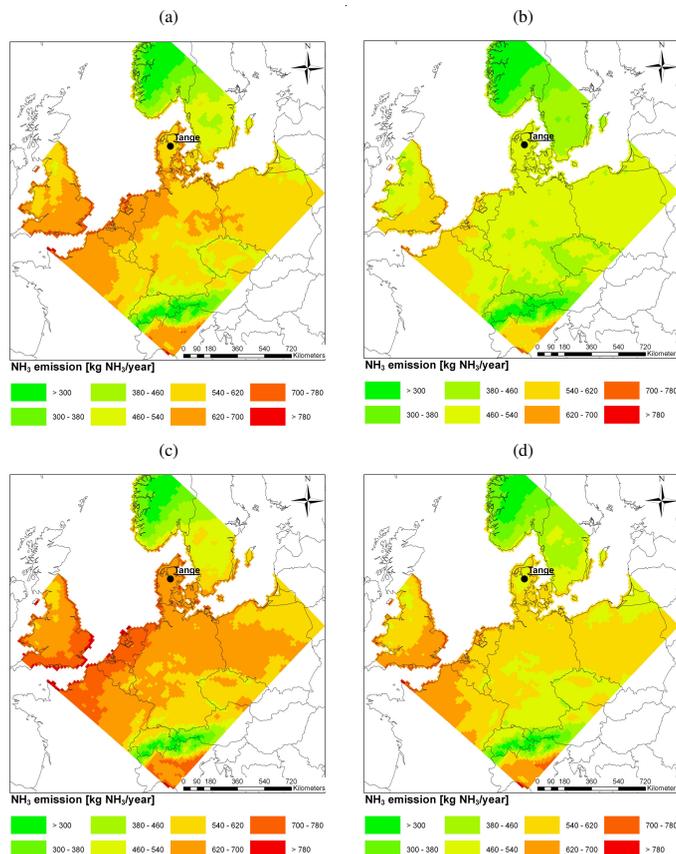


Fig. 3. Annual emission from storage in a reference pig production facility (located at Tange) with 1000 animals, when the facility is placed throughout central Europe and exposed to different European climatic conditions in the years (a) 2047, (b) 2050, (c) 2087, (d) 2090, respectively.

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Effect of climate and climate change on ammonia emissions

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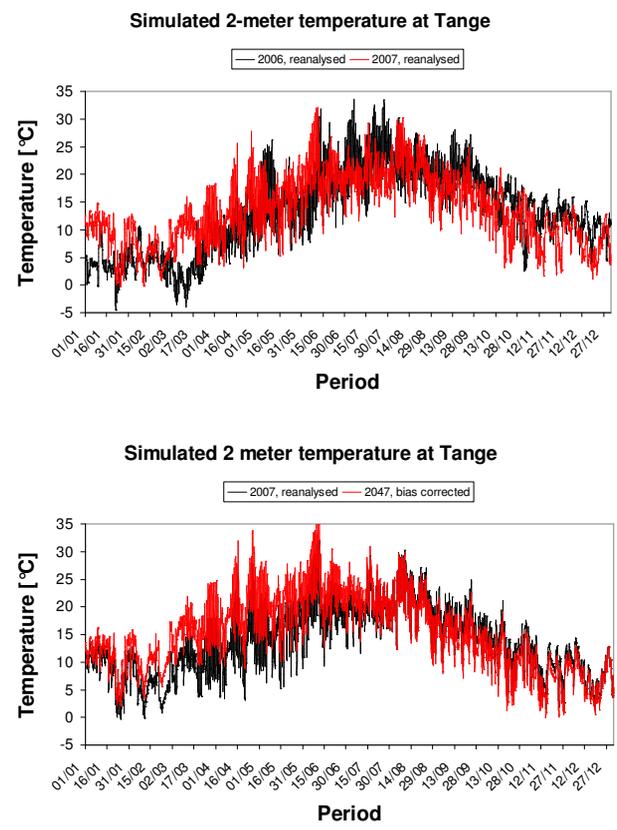


Fig. A1. (a) The hourly variation in simulated 2 m temperature for the Tange area that were used in the paper by Skjøth et al. (2011) and correspondingly hourly variations for the year 2006 using an identical model setup. **(b)** The hourly variation in simulated 2 m temperature for the Tange area that were used in the paper by Skjøth et al. (2011) and the simulated future variation on 2 m temperature using climate model output from the ENSEMBLES data set and bias correction.

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