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Ammonia emissions in Europe, part I: Development of a dynamical ammonia emission inventory

Anna Backes [a,*], Armin Aulinger [a], Johannes Bieser [a,b], Volker Matthias [a], Markus Quante [a]

[a] Helmholtz-Zentrum Geesthacht, Institute of Coastal Research, Max-Planck-Strasse 1, 21502 Geesthacht, Germany
[b] Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, 82234 Weßling, Germany

HIGHLIGHTS

- We present an evaluated dynamical ammonia emission inventory.
- The inventory is suitable to compare and assess ammonia abatement strategies.
- CMAQ model run of temporal parameterization influences on sec. aerosol formation.
- Correlation coefficient of NH_3 improved significantly for 12 out of 16 EMEP stations.

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ABSTRACT

Nitrogen input from agricultural ammonia emissions into the environment causes numerous environmental and health problems. The purpose of this study is to present and evaluate an improved ammonia emission inventory based on a dynamical temporal parameterization suitable to compare and assess ammonia abatement strategies. The setup of the dynamical time profile (DTP) consists of individual temporal profiles for ammonia emissions, calculated for each model grid cell, depending on temperature, crop type, fertilizer and manure application, as well as on local legislation. It is based on the method of Skjøth et al., 2004 and Gyldenkærne et al., 2005. The method has been modified to cover the study area and to improve the performance of the emission model. To compare the results of the dynamical approach with the results of the static time profile (STP) the ammonia emission parameterizations have been implemented in the SMOKE for Europe emission model. Furthermore, the influence on secondary aerosol formation in the North Sea region and possible changes triggered through the use of a modified temporal distribution of ammonia emissions were analysed with the CMAQ chemistry transport model. The results were evaluated with observations of the European Monitoring and Evaluation Programme (EMEP). The correlation coefficient of NH_3 improved significantly for 12 out of 16 EMEP measurement stations and an improvement in predicting the Normalized Mean Error can be seen for particulate NH_4^+ and NO_3^-. The prediction of the 95th percentile of the daily average concentrations has improved for NH_3, NH_4^+ and NO_3^-. The NH_3 concentration modelled with the STP is 157% higher in winter, and about 22% lower in early summer than the one modelled with the new DTP. Consequently, the influence of the DTP on the formation of secondary aerosols is particularly noticeable in winter, when the $PM_{2.5}$ concentration is 25% lower in comparison to the use of STP for temporal disaggregation. Besides, the formation of particulate SO_4^{2-} is not influenced by the use of the DTP.

1. Introduction

Emissions of reactive nitrogen into the atmosphere cause numerous problems of global significance, such as air pollution, eutrophication and soil acidification (Erisman et al., 2008; van Grinsven et al., 2013). The main components of reactive nitrogen are NO_x arising from transport or power generation and agricultural ammonia (NH_3) (Fowler et al., 2013). As there has been little progress in controlling agricultural ammonia emissions, their share in European air pollution is constantly increasing (Velthof et al., 2014; Sutton et al., 2011). The share of agriculture related NH_3

* Corresponding author.
E-mail address: anna.backes@hzg.de (A. Backes).

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emissions ranges between 85 and 99% in countries with high agricultural activity, such as e.g. Denmark (FAOSTAT, 2014; Schulze et al., 2010). Political restrictions as the National Emission Ceilings Directive (NECD) are often based on reported annual emissions of the member states, which make the future use of more accurate emission inventories, including agricultural procedures, likely (Hutchings et al., 2001).

> “This is likely to mean emission inventories become more complex in the future. We believe this extra complexity may be justified, particularly for countries with the highest animal densities” (Hutchings et al., 2001)

Ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3) make up a large fraction of fine particles (Anderson et al., 2003; Hristov, 2011; Werner et al., 2014), which pose a threat to human health (Aneja et al., 2009). As pointed out by Dentener et al. (2006) most model studies focus on the influence of emission reductions of sulphur- and nitrogen oxides on aerosol concentration. This study, however, has the aim of generating a better understanding of the ammonia emission distribution with regard to the geographical-, as well as the temporal aspect. The detailed, evaluated and applicable model was particularly designed to fit the needs of scenario studies (Backes et al., 2015) for investigating the influence of different ammonia abatement strategies on the formation of particles. Diverse studies suggested that a temporal component based on meteorological variables should be considered when applying ammonia emissions in a chemistry transport model (CTM) due to their high temporal variability (Hutchings et al., 2001; Skjøth et al., 2004; Gyldenkærne et al., 2005; Huang et al., 2012; Sutton et al., 2012; Reis et al., 2011). In this study, a parameterization was developed based on the dynamical ammonia emission parameterization by Skjøth et al., 2004 and presented in detail by Gyldenkærne et al., 2005. A profile named STP represents the time profile used to disaggregate ammonia emissions as presented in Schaap et al., 2005. The DTP was developed and applied to ammonia emissions in the here presented study. Recent studies (Skjøth et al., 2011; Megaritis et al., 2013) recommended the further analysis of the dynamical parameterization of ammonia on its transformation products with CTMs, even though this makes it more difficult to trace errors back to the time profile (Pinder et al., 2006). Therefore in this study the distribution of ammonia in the atmosphere and the contribution to the formation of secondary aerosols were modelled with the Community Multiscale Air Quality model (CMAQ). SMOKE for Europe (Bieser et al., 2011) served to prepare a model ready emission inventory for this CTM and EMEP observations were used to evaluate the results.

2. Methods and model description

The method and model description section includes a brief introduction to the emission model SMOKE for Europe, the applied time profiles and the emission inventories that have been used. Additionally the CTM CMAQ is described here with special focus on the implementation of the atmospheric transformation processes of gaseous NH_3 into particulate NH_4^+, SO_4^{2-} and NO_3^-.

2.1. Emission model SMOKE for Europe and the used emission inventories

The anthropogenic and biogenic emissions were processed by the emission model SMOKE for Europe (Bieser et al., 2011). SMOKE for Europe is the official emission model of the Community Modelling and Analysis System (CMAS) (Byun and Ching, 1999; Byun and Schere, 2006). Setting up and evaluating different ammonia emission scenarios, like done in a follow-up study (Backes et al., 2015), requires a sectorized emission inventory to ensure that different NH_3 emission sectors can be analysed separately. The Emissions Database for Global Atmospheric Research (EDGAR) consists of eleven emission sectors two of which are related to agriculture: *Emissions from Agricultural Soils* and *Manure Management* (EDGAR, 2009). This distinction of the inventory allows differentiated research on the influences of animal farming and crop farming, necessary for abatement strategy assessments. Furthermore, in contrast to the officially reported national emissions from EMEP, the EDGAR data set represents a bottom–up inventory based on expert estimates, activity data and emission factors (Hertel et al., 2011). The temporal profiles described in the next section have been applied to the sectors *Emissions from Agricultural Soils* and *Manure Management* based on the data analysis which revealed that 96% of the total European ammonia emission is caused by subsectors grouped in these two emission sectors. Within the study area the share of *Manure Management* accounts for 53% and the share of *Emissions from Agricultural Soils* for 43% of the ammonia emissions (see appendix table A3). Ammonia emissions from the sectors industry and transport, which made up 4% of the ammonia emissions used in this study, were taken from the according EMEP SNAP (Selected Nomenclature for sources of Air Pollution) sectors (European Environmental Agency, 2007). Due to their small share in total European emissions they were not replaced by the remaining nine EDGAR non-agricultural sectors.

It has been concluded in previous studies that atmospheric concentrations of NH_3 highly depend on the emission time profile and that therefore, the temporal component should be considered when implementing ammonia emissions for CTM modelling, preferably through meteorological variables (Denier van der Gon et al., 2011; Hutchings et al., 2001).

2.1.1. Static time profile (STP)

The STP is the commonly used temporal disaggregation scheme for ammonia emissions in current CTMs for Europe (compare AQMEII (Pouliot et al., 2012), Eurodelta III (EMEP, 2014) and COST 728 (World Meteorological Organization, 2008). The STP (Fig. 2) was adopted from the LOTOS-EUROS documentation (Schaap et al., 2005). It represents the SNAP sector level 1, category 10 (agriculture). A daily or weekly factor was not defined (Schaap et al., 2005); while a static hourly profile is included. The profile is static in the sense that the same profile was applied to every grid cell in the model domain. It lacks a dynamic, meteorology dependent component or the consideration of country specific differences in policies or intensity of animal husbandry. As the emission of ammonia is highly dependent on temperature, the missing variability is expected to be a considerable limitation. Denier van der Gon et al. (2011) tested the sensitivity of air quality models to temporal distribution of emissions and recommends an implementation of meteorology dependent functions.

2.1.2. Dynamical time profile (DTP)

Like the STP, the DTP was applied to the geographically distributed annual bulk emissions of the gridded EDGAR emission sectors *Emissions from Agricultural Soils* and *Manure Management.* In this approach, the annual emissions of the inventory have been temporally distributed across the year on the basis of the meteorological variables wind speed and surface temperature, resulting in individual ammonia emission data for every grid cell and every hour. Parts of the newly developed parameterization are a modified version of the dynamical ammonia emission parameterization by Skjøth et al. (2004) and Gyldenkærne et al. (2005), which is currently considered as a substantial improvement of the available ammonia emission estimates (Pinder et al., 2007). To improve the