Atmospheric Environment 126 (2016) 153–161

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Ammonia emissions in Europe, part II: How ammonia emission abatement strategies affect secondary aerosols

Anna M. 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, Geesthacht 21502, Germany
[b] Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Weßling 82234, Germany

HIGHLIGHTS

- We studied the influence of ammonia emissions on the formation of secondary aerosols.
- We created 4 emission scenarios.
- Scenarios are based on NEC, technical feasibility and agricultural sectors.
- 50% emission reduction leads to up to 25% reduction of total $PM_{2.5}$ concentrations in winter.
- Ammonia reduction in the animal husbandry agricultural sector is highly efficient.

ARTICLE INFO

Article history:
Received 12 February 2015
Received in revised form
16 November 2015
Accepted 18 November 2015
Available online 22 November 2015

Keywords:
Ammonia
Emission modelling
SOA formations
Agricultural emissions
CTM
Ammonia abatement strategies
Scenario study
Animal husbandry

ABSTRACT

In central Europe, ammonium sulphate and ammonium nitrate make up a large fraction of fine particles which pose a threat to human health. Most studies on air pollution through particulate matter investigate the influence of emission reductions of sulphur- and nitrogen oxides on aerosol concentration. Here, we focus on the influence of ammonia (NH_3) emissions. Emission scenarios have been created on the basis of the improved ammonia emission parameterization implemented in the SMOKE for Europe and CMAQ model systems described in part I of this study. This includes emissions based on future European legislation (the National Emission Ceilings) as well as a dynamic evaluation of the influence of different agricultural sectors (e.g. animal husbandry) on particle formation. The study compares the concentrations of NH_3, NH_4^+, NO_3 -, sulphur compounds and the total concentration of particles in winter and summer for a political-, technical- and behavioural scenario. It was found that a reduction of ammonia emissions by 50% lead to a 24% reduction of the total $PM_{2.5}$ concentrations in northwest Europe. The observed reduction was mainly driven by reduced formation of ammonium nitrate. Moreover, emission reductions during winter had a larger impact than during the rest of the year. This leads to the conclusion that a reduction of the ammonia emissions from the agricultural sector related to animal husbandry could be more efficient than the reduction from other sectors due to its larger share in winter ammonia emissions.

1. Introduction

Ammonia emissions is considered as a major issue of political and scientific concern as it is a threat to health and the environment (Erisman et al., 2008; Grinsven et al., 2013). Next to NO_x, from transport or power generation, ammonia (NH_3) emitted from the agricultural sector has the highest contribution to the atmospheric nitrogen budget (Reis et al., 2009). Several approaches to manage the nitrogen cascade have been implemented by the EU or are under discussion (EC, 2001; EC, 2005; EC, 2008). But despite political intentions the European directives achieve relatively small reductions of NH_3 emissions from agriculture (Velthof et al., 2014) and the costs for additional measures are highest in this sector (Amann et al., 2011). Airborne particulate matter is one of Europe's most problematic pollutants in terms of harm to health (EEA, 2010). Aerosol formed with ammonia contributes to a high share to the

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

http://dx.doi.org/10.1016/j.atmosenv.2015.11.039

total mass of particulate matter, smaller 2.5 μm and 10 μm ($PM_{2.5}$ and $PM_{10)}$ (Anderson et al., 2003; Hristov, 2011; Werner et al., 2014), making them an important component in aerosol processes (Xu and Penner, 2012). The effect of the variations of ammonia emissions in time and amount on the formation and transport of secondary particulate matter will therefore be analysed in this study in form of a scenario analyses. To improve the knowledge about ammonia emissions, mitigation strategies and their impact on particle formation in human and ecological systems (Aneja et al., 2009; Shibata et al., 2015) the aerosol formation has been modelled with a chemistry transport model (CTM) for every scenario to evaluate effectiveness and efficiency of the different mitigation measures, as recent studies suggest (Moss et al., 2010; Vuuren et al., 2011). The abatement strategies compared are the European policy instrument of the anticipated National Emission Ceilings (NEC), the maximum technical feasible emission reduction (MTFR) and a change in consumer habits concerning the consumption of animal products. The reduced consumption of animal products (RCAP) assumed in the third scenario has been based on the diet recommendations of the Harvard medical school (Willett and Skerrett, 2005).

2. Methods and model description

The Model description includes a brief introduction to the Community Multiscale Air Quality model (CMAQ), the Sparse Matrix Operator Kernel Emissions Europe (SMOKE for Europe), the applied time profiles for NH_3 emissions, the emission inventories that have been used and an explanation of the atmospheric transformation processes of $NH_{3.}$ A detailed description of the underlying temporal parameterization and its contrasts to the former used time profile can be referred to in Part I of this study (Backes et al., 2015).

2.1. Emission model SMOKE for Europe

The surface emissions of anthropogenic and biogenic sources have been processed using the emission model SMOKE for Europe (Bieser et al., 2011). SMOKE is the official emission model of the Community Modelling and Analysis System (CMAS) and the emission data created is suitable for CMAQ (Byun and Ching, 1999; Byun and Schere, 2006). SMOKE for Europe is the adaptation of this emission model to Europe; it uses the BEIS version 3.14 to estimate VOC emission from soils and vegetation (Pouliot and Pierce (2009). The development and the implementation of the dynamical time profile (DTP) in the SMOKE for Europe model, which has been used in this study as a reference has been presented in detail in Part I of this study. A detailed temporal distribution of ammonia emissions is one cornerstone of this analysis to be able to study seasonal variations evolved by the different mitigation strategies.

Next to this validated dynamic ammonia emission parameterization a sectorized emission inventory has been a main column of this studies setup. The division of the Emissions Database for Global Atmospheric Research (EDGAR) ammonia emission inventory into the two main emission sectors *Emissions from Agricultural Soils* and *Manure Management* sector offers a major advantage concerning this analysis (EDGAR, 2009). The influences of animal farming and crop farming can be investigated independently from each other, even though the interaction through manure applied on fields had to be taken into account. The necessity of this split is further explained in Section 3.4. This sectorisation has been the main argument for the choice of the bottom up emission inventory EDGAR. An organization chart of the different sectors represented in the inventory is given in table A1 of the appendix.

2.2. Chemistry transport model CMAQ

The formation of secondary aerosols based on different scenarios has been modelled with the CTM CMAQ. CMAQ computes chemical and physical transformation, transportation and deposition of air pollutants contingent on the emission input. It includes gas phase, aerosol and aquatic chemistry, as well as primary and secondary particles (Byun and Ching, 1999; Byun and Schere, 2006; Matthias, 2008) and has been described in detail in Part I of this study. The modelling domain incorporates north-west Europe with a spatial resolution of 24 × 24 km^2 and 30 vertical layers. This model domain has been chosen, since all ammonia emission hotspots lying within this part of Europe due to its large share of utilized agricultural area. The results presented in this study refer to the lowest model layer which reaches to an altitude of approximately 40 m. In this study, the carbon bond V mechanism (CB-05) photochemical mechanism was used and boundary conditions were taken from monthly means of the TM5 global chemistry transport model system (Huijnen et al., 2010), provided by the Dutch Royal Meteorological Institute (KNMI). The boundary conditions for the nested 24 × 24 km^2 grid were calculated from the outer coarse 72 × 72 km^2 grid. The meteorological fields were derived from the regional, non-hydrostatic meteorology model COSMO-CLM 4.8 (Rockel and Geyer, 2008; Rockel et al., 2008). The meteorological data is based on the meteorological situation of the reference year 2008 as this year has not shown unusual meteorological conditions in Europe. Aerosols are represented in 3 different classes (Aitken, accumulation and coarse mode), whereat the Aitken and accumulation modes are summed up as $PM_{2.5}$ particles. The atmospheric transformation of ammonia is implemented in CMAQ as a condensation process onto existing aerosols or as a reaction with gas phase acids forming secondary aerosols. The state of the art ISORROPIA module version1.7 has been used in this study (Nenes et al., 1998). The importance of ammonia emissions for air pollution results from the transformation of gaseous emissions into a particulate species. The particle size is essential for discussing air quality and health topics, as it determines the respirability. Particles smaller than 2.5 μm ($PM_{2.5}$) are mainly formed through coagulation, coarse mode particles (all particles larger 2.5 μm) are mostly directly emitted and grow through a condensation process. This condensation onto an existing particle takes mainly place where large particles, as sea salt particles, occur. The contribution of particles in the coarse mode to the total particle mass is low. If present, gaseous NH_3 preferentially reacts with sulphuric acid (H_2SO_4), formed by the oxidation of SO_2 (Seinfeld and Pandis, 1998). If sulphuric acid is the limiting factor for particle formation, the reaction with other acid gaseous compounds as nitric acid (HNO_3) or hydrochloric acid (HCl) takes place. The aerosols ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4 Cl) are formed in balanced reactions in contrast to ammonium sulphate which is formed in an irreversible reaction (Hertel et al., 2011). With this setup, four CMAQ runs using different emission datasets (one for the Reference case and three representing the mitigation scenarios) have been performed.

3. Scenario development

3.1. Reference case

The Reference case is, as already mentioned, based on the modelled NH_3 emissions introduced as *Dynamical Time Profile* (DTP) in Part I. The time profile validated with the European Monitoring and Evaluation Programme's (EMEP) measurements forms the basis of this study. It serves as a reference for the comparison of mitigation approaches modelled in the form of the