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

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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.

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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

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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_4Cl) 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 scenarios NEC2020, MTFR and RCAP. While the political scenario is based on European legislations and the technical scenario is adopted from a study where it has been used already to model ammonia development, the behavioural scenario has been particularly developed for this study.

3.2. Political scenario NEC 2020 – national emission ceilings coming into force 2020/ 30

The first scenario focuses on future political restrictions concerning the NH_3 emissions. In the Thematic Strategy on Air Pollution (TSAP) the European Commission has set itself objectives for the protection of human health and the environment (EC, 2005). Relative to the year 2000 the years of life loss (YOLLs) as a result of exposure to particulate matter should be reduced by −47% (EC, 2005). The establishment of the National Emission Ceilings attainable since 2010 has been part of this strategic approach. The revision of the National Emission Ceilings Directive NECD (EC, 2001), has been considered as one of the main policy instruments to achieve the environmental objectives for 2020 (EC, 2005). As the proposal of the NECD is still under preparation the statutory NEC 2020 cannot serve as the basis of this scenario. Rather an elementary part of the revision has been used to develop the scenario: the integrated modelling services provided by the International Institute of Applied Systems Analysis (IIASA). The additional emission reductions are presented in the NEC Scenario Analysis Report Nr.8 and have built the foundation of this model approach (Amann et al., 2011). On the basis of these calculations the NEC 2020 is discussed by politicians, thus it is expected that the revised policy instrument, expected to enter into force 2020/30, will give similar limits as the report. IIASA indicates that the reductions would be necessary to meet the environment and health objectives of the TSAP. Another requirement has been that non-European countries emit at the previous level, not implementing the emission ceilings on a voluntary basis. The scenario reflects the national energy and climate policies of the year 2010 and is referenced in the report as the Price-driven equilibrium Model of the Energy System and markets for Europe (PRIMES) Baseline scenario (Amann et al., 2011). Table A3 offers an overview on the maximum emissions per country in kt/a for the scenarios NEC2020 and MTFR.

3.3. Technical scenario MTFR – maximum technical feasible reduction

The maximum technical feasible emission reduction shows opportunities and limitations of a NH_3 abatement strategy mainly based on technical possibilities. The NEC Scenario Analyses Report Nr. 8 (Amann et al., 2011) presents a scenario where the NH_3 emissions were modelled with the Greenhouse Gas and Air Pollution Interactions and Synergies model (GAINS), assuming the use of latest technologies. Table A4 summarizes the technologies used to model this scenario. Underlying activities, e.g. a switch towards renewable fuel that would reduce emissions of particles below a level that could be achieved with filter technologies have been considered in this approach. Various control technologies which have been used in the GAINS model of IIASA are listed in Table A3. The scenario is, as scenario NEC2020, based on the PRIMES Baseline scenario (Amann et al., 2011).

3.4. Behavioural scenario RCAP – reduced consumption of animal products

The European Nitrogen Assessment has identified “Lowering the human consumption of animal protein” as one of seven key actions to manage the nitrogen cascade (Sutton and van Grinsven, 2011). The approach is addressed to policy makers accompanied by the recommendation of a stronger effort in this sector due to the limited success in reducing agricultural nitrogen emissions so far (Sutton and van Grinsven, 2011) and the huge reduction potential of animal husbandry (Galloway et al., 2008; Erisman et al., 2008). Under these circumstances the RCAP scenario assumes a European society whose diet is based on the recommendation of the Harvard medical school (Willett and Skerrett, 2005). These recommendations aim at a choice of a healthier source of proteins for health reasons. Furthermore the guideline has been used in other studies to model the influence of this diet on climate change (Stehfest et al., 2009). The recommended diet includes a daily intake of 10 g beef, 10 g pork, 46.6 g chicken and eggs, 23.5 g fish per capita and no change in the consumption of milk (Stehfest et al., 2009). To calculate the decrease in NH_3 arising from the consumption change, the actual consumption of ruminant, pork and poultry meat (including eggs) per country has been compared with the consumption in case of a healthy diet. The data of meat consumption per capita has been adopted from the statistic division of the food and agriculture organization (FAOSTAT, 2014) for all countries of the European Union. The per capita consumption of ruminant meat consists of food commodities of cattle, sheep and goat meat in 2008. To be able to calculate the difference in consumption in the case of a reduced consumption of animal products, the aggregation of all ruminants has been necessary as the recommendation of the Harvard medical school is given in g/ruminant meat in this field. For the same reason poultry meat consumptions and egg consumption per capita have been aggregated as poultry products. An overview of the classification of the food commodities has been given in table A2. The difference of the actual consumed amount and the recommended amount has been calculated for the three categories, so that the percentage of reduction per species and country could be applied on the gridded livestock inventories of the Food and Agriculture Organization (FAO) (FAO, 2007). This decrease of the amount of animals per species due to the consumption change is given in percent in Table A3. As the recommendation assumes no change in the intake of milk products, only cattle and no change in the amount of dairy cows has been considered in the calculations. A change in the export of animal products has not been considered either, assuming that other regions in the world with an increasing consumption will be able to meet their own market demands in the future. Furthermore this scenario has not drastically affected the emissions from the sector *Emissions from Agricultural Soils*. Mere its subsector *Application of Manure on Fields to empty Storage Facilities* has been omitted. It has been assumed that fields will be tended and fertilized, also under the assumption of a lower consumption of animal products. The reduction used in the RCAP scenario has been presented in Table A3 in percent per species, resulting in an emission reduction based only on the animal husbandry related sector.

4. Results

In this section the impact of emission changes on air pollutants concentrations will be presented. The results show seasonal emission and concentration values of NH_3 as well as concentration of acid gases and particles which play a major role in the formation process of particulate matter in the atmosphere. PM_{10} and $PM_{2.5}$ concentrations are both presented as totals for all substances as well as particle sizes for the most important substances involved in the formation processes. The time periods used to visualize the data were defined as meteorological seasons of three month each (whereby December of the year 2008 has been grouped with January and February of the same year for the winter season). The emission values in Table 1 show the seasonal sum of NH_3 emissions (t/season), while the concentration values are the average

Table 1
Annual ammonia emissions in the model domain for the Reference case and the three scenarios.

Scenarios	Reference case	NEC2020 Political scenario	MTFR Technical scenario	RCAP Behavioural scenario
Total NH_3 emissions	4182 kt/a	3735 kt/a	3224 kt/a	2716 kt/a

concentration of the season in μg/m^3. Table 1 gives a first impression of the reductions' dimension showing the annual emissions in kt/a of the Reference case and the three scenarios.

4.1. NH_3 emissions

Emission sums calculated with the SMOKE for Europe model are presented as annual totals in Table 1. The analysis of the emission distribution patterns (Fig. 1) demonstrate, regardless of the scenario modelled, that the highest ammonia emissions occur in Denmark, Brittany, Belgium, the Netherlands, Northern Ireland and the Po-valley. It is expected that the emission hotspots can be identified in NH_3 concentration maps as well, due to the low transportability of ammonia. Fig. 1 shows that all scenarios show lowest emissions for the remaining Scandinavian countries, the northern UK and Eastern Europe.

A constant decrease of ammonia emissions can be recognized in every scenario as well as in the Reference case from spring to winter, intermitted by an autumn peak of emissions, see Fig. 2. The reduction of ammonia shows different seasonal patterns for the three scenarios. The NEC2020 scenario and the MTFR scenario show a seasonal reduction proportional to the seasonal emissions in the Reference case. The differences among the seasons are low except for a stronger reduction in spring due to higher emissions in the Reference case. Fig. 2 demonstrates that the reduction in the RCAP case is overall higher as the reduction achieved with the political or technical scenario. In addition, the reduction pattern shows a different seasonal character. Ammonia emissions decrease drastically in winter while the difference to the MTFR or NEC2020 reductions in spring and summer are marginal. This gap arises from the larger share of the animal husbandry related *Manure Management* sector in the cold season (Hristov, 2011), as no fertilizing takes place (see the organization chart, table A1).

4.2. Concentrations

As the reduction scenarios NEC2020 and MTFR are very similar in their spatial and temporal distribution, the following comparisons focus on the scenarios NEC2020 as an effect-oriented scenario and RCAP as a driver-oriented scenario. While the effect oriented scenario defines environmental targets (as described in Section 3.2.) and identifies emission control values to reach the target level, a driver oriented scenario uses a driver (as e.g. the population and their consumption) to predict the environmental impacts. To show the maximum differences between the seasons, summer and winter seasons are chosen for the comparison. The results of the modelled concentrations are presented in order of their relevance for the chemical transformation process. The aerosols chemistry is presented as fine particles of $PM_{2.5}$.

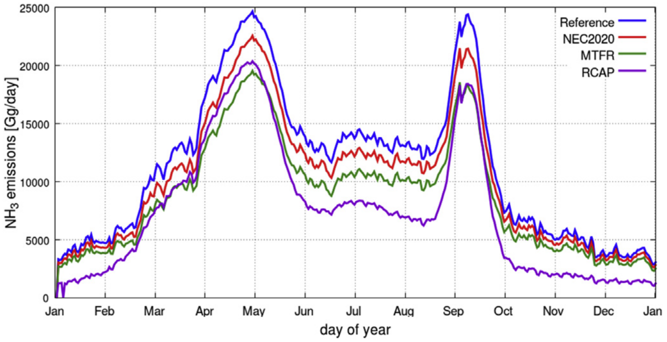

Fig. 2. Time series of the Reference case, the NEC2020, MTFR and RCAP scenario. The mean value of all grid cells enclosing land is presented.

4.2.1. Ammonia

The concentration patterns of NH_3 are in line with the NH_3 emission pattern. A decrease of ammonia from spring to winter in all scenarios and the Reference case occurs. The concentration has decreased by 20% in the NEC2020 scenario compared to the Reference case in winter. For the other seasons the reduction in this scenario varies from −12% in spring and summer to −14% in autumn. The reductions of the RCAP scenario in summer (−46%), autumn (−46%) and winter (−59%) are strong, whereas the weaker spring concentrations decline (−23%) is still a stronger decrease than any achieved in the political scenario (compare overview Table 2). The results need to be interpreted relatively to the concentrations per season in the Reference case. In the Reference case the hotspots of animal farming in Europe (the Netherlands and the Weser-Ems region in West Germany are clearly visible in Fig. 3. In line with this high emission areas the concentration pattern in the RCAP scenario shows, that the difference is highest in the area with extensive animal farming.

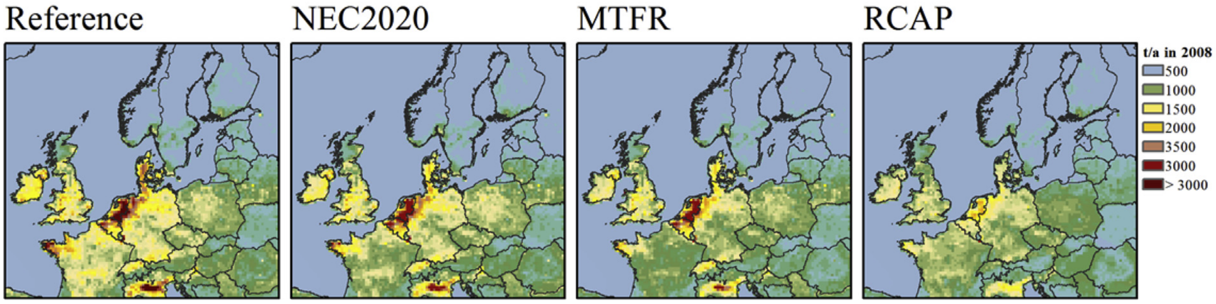

Fig. 1. Emission patterns of the annual total NH_3 emissions in t/a. Reference case and the scenarios NEC2020, MTFR and RCAP.

Table 2
Seasonal reductions in emission, concentrations and deposition of the individual substances achieved through the scenarios NEC2020 and RCAP. The difference of the concentrations of the scenarios NEC2020 and RCAP to the reference case is given as mean difference over all land cells in percentage. The table does not include concentrations of MTFR due to the similarity of this scenario with the NEC2020 scenario concerning spatial and temporal distribution.

Substance	NEC2020 spring	NEC2020 summer	NEC2020 autumn	NEC2020 Winter	RCAP spring	RACP summer	RCAP autumn	RCAP winter
NH3 EMISSIONS	−11%	−11%	−11%	−12%	−22%	−42%	−46%	−56%
NO_3^-	−4%	−7%	−6%	−6%	−16%	−31%	−50%	−48%
HNO_3	6%	3%	6%	6%	30%	13%	55%	51%
NH3	−12%	−12%	−14%	−20%	−23%	−46%	−46%	−59%
NH_4^+	−2%	−2%	−4%	−4%	−10%	−10%	−33%	−39%
N	−5%	−6%	−4%	−3%	−12%	−23%	−22%	−19%
SO_4^{2-}	0%	0%	0%	0%	−1%	0%	−2%	−3%
H_2SO_4	1%	0%	2%	2%	6%	2%	14%	24%
$PM_{2.5}$	−2%	−1%	−3%	−2%	−8%	−6%	−22%	−24%
PM_{10}	−2%	−1%	−2%	−1%	−7%	−5%	−19%	−21%
N -DEPOSITION	−8%	−8%	−6%	−4%	−15%	−31%	−20%	−13%

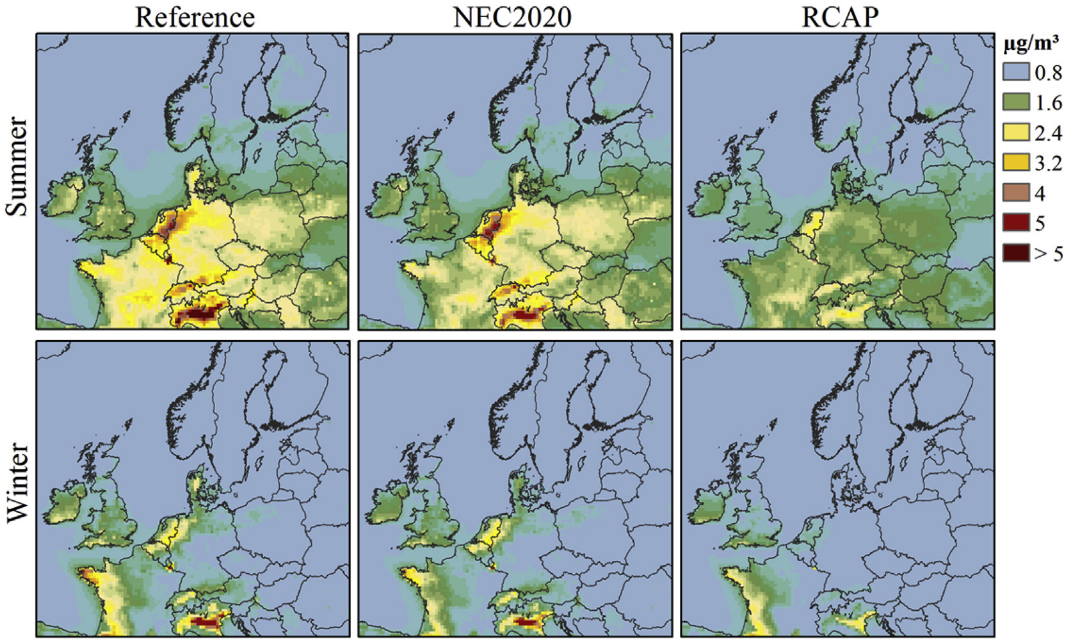

Fig. 3. NH_3 concentration for the Reference case, the NEC2020 and the RCAP scenario. The reduction is presented in µg/m^3 for winter and summer seasons.

4.2.2. *Ammonium*

Although the formation of particulate NH_4^+ primarily depends on the occurrence of ammonia in the atmosphere (Section 2) the seasonal NH_4^+ concentrations differ from the seasonal distribution of NH_3 concentrations in the Reference case. While spring (1.54 µg/m^3) and summer (1.34 µg/m^3) concentrations of NH_3 have been the higher than in autumn (0.84 µg/m^3) and winter (0.31 µg/m^3) the seasonal patterns of NH_4^+ have their annual peaks in winter (1.36 µg/m^3). Spring values of NH_4^+ (1.15 µg/m^3) are comparable to those of NH_3 while summer values differ strongly (0.66 µg/m^3) (Table 2). This is because ammonium nitrate particles are not stable in summer and decompose back into the gas phase at high temperatures. Despite the seasonal differences between NH_3 and NH_4^+ the reduction scenarios for NH_4^+ have been described below. All reduction scenarios have additionally been set out in Table 2. The reduction in the NEC2020 is much less than the reduction in the RCAP as Fig. 4 indicates. The maximum reduction of NH_4^+ achieved through NEC2020 is −4%. For the RCAP the reduction in autumn (−33%) and winter (−39%) is highest compared to the Reference case, while the concentration reduction is lowest in summer (−10%). The relatively high NH_4^+ concentrations in spring in the Reference case do not result in an equally high reduction (−10%) in the RCAP scenario.

4.2.3. *Sulphur compounds*

The SO_4^{2-} concentration of any scenario has changed only marginally compared to the Reference scenario (from 0% in summer to −3% in winter for the RCAP, Table 2). The concentrations are constant concerning the seasons within the scenarios and the Reference case. The maximum variation ranges from 2.1 µg/m^3 in winter to 1.63 µg/m^3 in summer in the Reference case. This indicates that the formation of SO_4^{2-} is limited by H_2SO_4 in every scenario. As expected the concentration of H_2SO_4 has neither changed in the NEC2020 scenario nor in the RCAP scenario compared to the Reference case. The results presented show the same pattern as the SO_4^{2-} concentrations concerning seasonal and scenario-based reductions.

4.2.4. *Nitrate*

In contrast to the nearly constant SO_4^{2-} concentrations, the NO_3^- concentrations are neither constant between the seasons nor in comparison of the scenarios to the Reference case as shown in

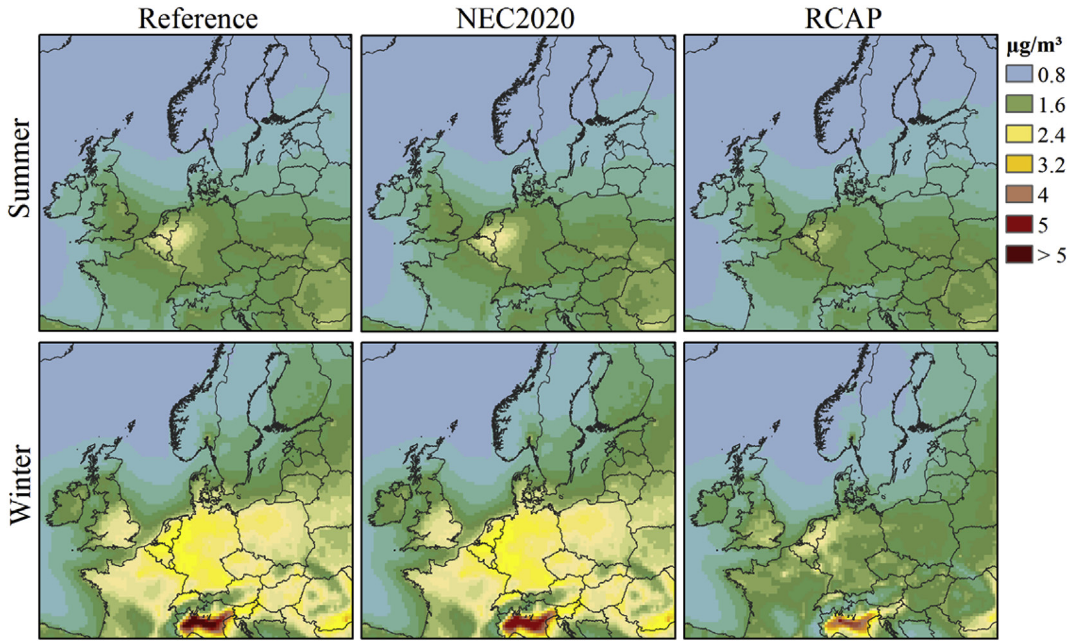

Fig. 4. NH_4^+ $_{(PM2.5)}$ concentration for the Reference case, the NEC2020 and the RCAP scenario. The reduction is presented in μg/m^3 for winter and summer seasons.

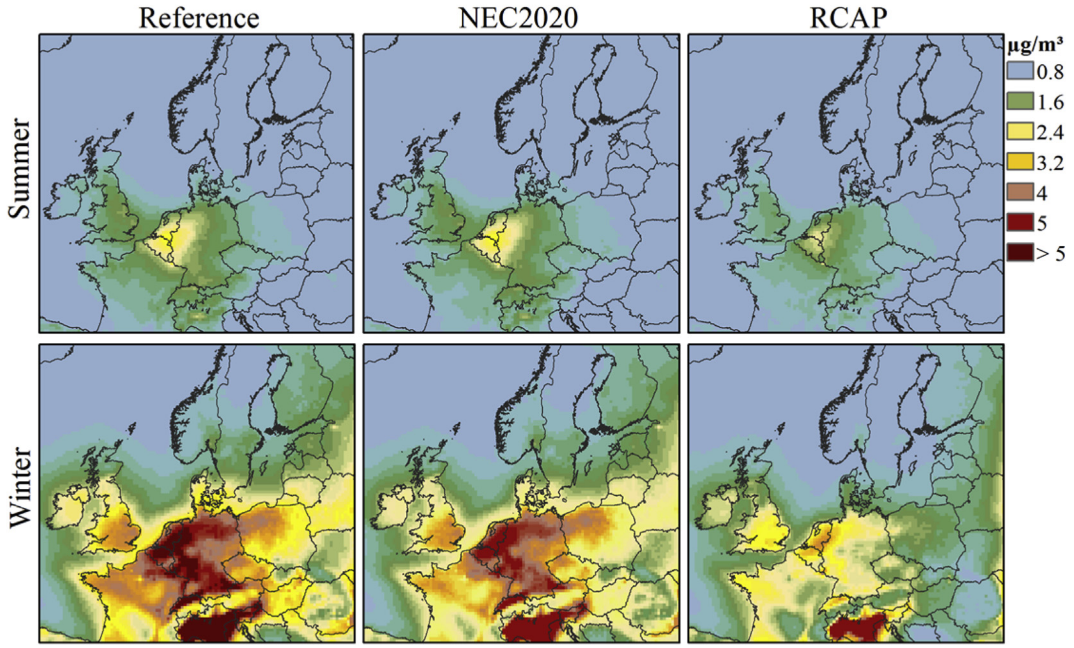

Fig. 5. NO_3^- $_{(PM2.5)}$ concentration for the Reference case, the NEC2020 and the RCAP scenario. The reduction is presented in μg/m^3 for winter and summer seasons.

Fig. 5. The Reference case presents a seasonal variability of 0.48 μg/m^3 in summer to 2.67 μg/m^3 in winter. The political scenario shows a moderate decrease of nitrate ranging from 4% in spring to 7% in summer. The RCAP scenario concentrations of NO_3^- in comparison to the Reference case is highest in autumn (−50%), winter (−48%) and summer (−31%) and lowest in spring (−16%) (Table 2).

4.2.5. Nitric acid

The HNO_3 concentrations exhibit weakened seasonal patterns compared to NO_3^-. While the summer concentration is similar to that of NO_3^- (0.48 μg/m^3), in winter (0.85 μg/^{3}), spring (0.36 μg/m^3) and autumn (0.66 μg/m^3) the concentration is far lower than the equivalents of NO_3^-. The NEC2020 scenario shows a moderate increase in gaseous HNO_3 on the same level as NO_3^- has decreased in this scenario (3% in summer to 6% in all other seasons). The RCAP scenario shows a major increase in all seasons proportional to the decrease in nitrate presented above. The increase is highest in autumn (55%), winter (51%) and spring (30%) and lowest in summer (13%). The maximum difference between the increase in the nitric acid and the decrease in nitrate is −18% in summer. As indicated by a comparison of nitrate and nitric acid in Table 2, the summer reduction of nitrate is higher than the summer increase of nitric acid. In comparison to all other species investigated in this study, the hotspots are neither in the Po-valley region nor over the Benelux states. Noticeable hotspots of HNO_3 can be seen over the seas and in various parts of Eastern Europe.

4.2.6. *Total $PM_{2.5}$ and PM_{10} concentrations*

The total $PM_{2.5}$ concentration has decreased in comparison to the Reference case for the political scenario NEC2020 as well as for the reduced consumption scenario RCAP (Table 2). Since SO_4^-, NO_3^- and NH_4^+ aerosols in the accumulation mode make up for a large share of total $PM_{2.5}$ particles, it is not surprising that the results are in line with the ones presented above for the individual substances. Other particles included in the $PM_{2.5}$ totals are organic and elemental carbon. The decrease achieved through the scenarios differs in total amount and seasonal variability. The $PM_{2.5}$ pattern for NEC2020 represents a very slight reduction of $PM_{2.5}$ concentrations in all seasons. The results for the RCAP scenario show a severe decrease in the $PM_{2.5}$ concentrations, mainly noticeable in winter (−24%) and autumn (−22%). A moderate decrease in spring (−8%) and summer (−6%) is presented in Table 2.

The results of NH_4^+ in the coarse mode show that the particle concentrations are decreasing in the same way over time and in the reduction scenarios as the NH_4^+ particles have. The SO_4^{2-} coarse mode particles show as little reduction in the scenarios and the same weak seasonal variability as the $PM_{2.5}$ concentrations. Unlike these concentrations, the NO_3^- coarse mode particles show an interesting pattern over sea areas. As expected the increasing HNO_3 concentration, as shown in Section 4.2.5, leads to an increased condensation onto existing particles. These particles are mainly NaCl from sea spray emissions, which explains the geographical pattern of NO_3^- coarse mode particles presented in Fig. 6.

Fig. 6 indicates that the concentration of NO_3^- coarse mode particles modelled with the RCAP scenario is increasing in winter and in summer. As expected, the concentration pattern shows a formation of coarse mode NO_3^- over sea areas as the North and Baltic Seas and the Adriatic Sea. The total PM_{10} concentration is decreasing though, in all scenarios. The total PM_{10} concentration has, as well as the total $PM_{2.5}$ concentration decreased in all scenarios, but the most substantial reduction can be seen in autumn (−19%) and winter (−21%) for the RCAP scenario.

The seasonal differences of the substance in the scenario in comparison to this substance in the Reference case are summarized in Table 2. The deviations are given in Table 2 as average concentration over all land cells in percentage. The mean over all land cells is given to be able to describe the influence of air pollution changes on population.

5. Discussion

To evaluate the impact of ammonia emission abatement strategies on air quality we run the CTM CMAQ using a dynamical temporal emission parametrization. The emission model results show good agreement with the temporally distributed NH_3 emissions of Skjøth et al. (2011). CMAQ results showed that the temporal disaggregation of NH_3 emissions has a strong impact on concentrations of NH_3 leading to a higher correlation between model and observations. Furthermore, the emissions influenced the formation of ammonium nitrate particles. Related modelling studies found similar results for Europe (Sutton et al., 2012; Damme et al., 2014), the US and China (Paulot et al., 2014). A detailed discussion of the CMAQ results and the impact of the dynamic temporal profiles can be found in Backes et al. (2015.).

The impact of the reduction scenarios is varying due to the annual total reduction as well as the reduction per season. With 17% (2% NEC2020, 6% MTFR) annual average reduction of $PM_{2.5}$ the behavioural scenario RCAP shows the highest influence on the reduction of particle concentrations. A higher efficiency of $PM_{2.5}$ concentration reduction based on a more ambitious abatement scenario has been found by Bessagnet et al. (2014) in a study comparing the three models CHIMERE, EMEP and LOTOS-EUROS. In contrary to the NEC2020 and the MTFR scenarios, where the reduction per season is proportional to the total emissions in the

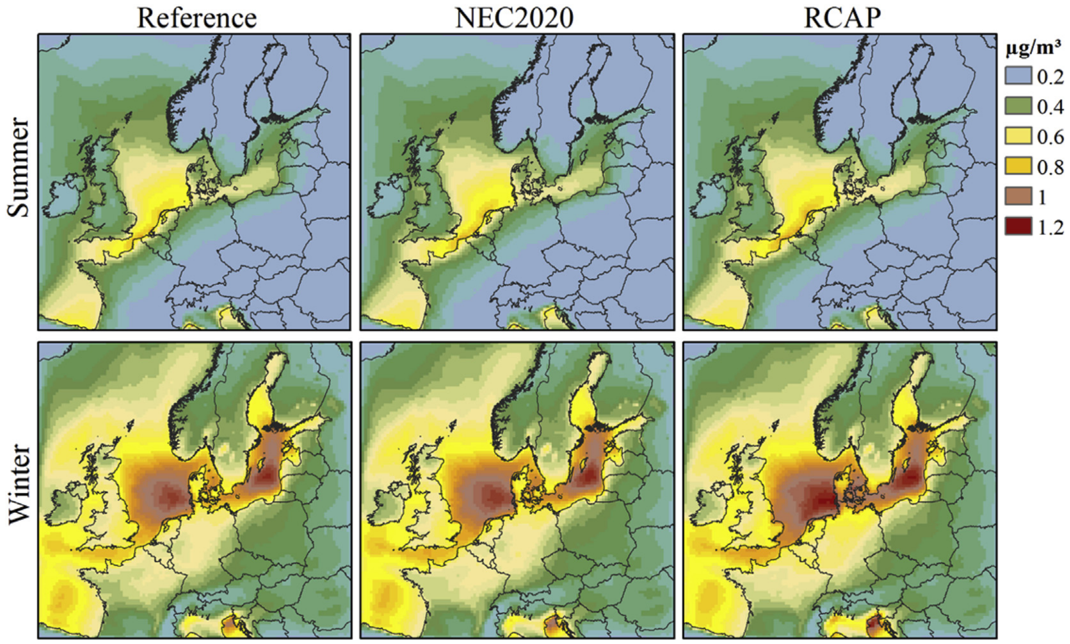

Fig. 6. NO_3^- (coarse mode) concentration for the Reference case, the NEC2020 and the RCAP scenario. The reduction of the coarse mode particles is presented in µg/m^3 for winter and summer seasons.

reference case, the influence of the RCAP scenario on the ammonia reduction is higher during winter. The ammonia emissions from the sector *Emissions from Agricultural Soil*, which are not reduced in the RCAP scenario, are mainly occurring in spring and have therefore a stronger impact in this season. This spring peak leads to an ammonia concentration high enough to cover the decrease in NH_3 of the animal related *Manure Management* sector. Meanwhile the animal farming related emissions of the *Manure Management* sector have a higher share (>80%) in the emission totals in autumn and winter, as explained in Section 4.1. This finding is confirmed by the results of Hristov (2011). Therefore the decrease of ammonia in the RCAP scenario, which modifies only emissions related to animal product consumption rather than manure application, is strongest during these seasons where agricultural activity other than animal farming is low.

While the emission patterns show a noticeable peak in spring, the $PM_{2.5}$ and PM_{10} concentration patterns show higher concentration peaks during winter in all scenarios as well as in the Reference case. The condensation process and the formation of aerosols is favoured by cold temperatures, humidity and higher concentrations of potential reactants, as the intensified heating during this period is a further source of NO_x and SO_2 (Roedel and Wagner, 2011). Furthermore the boundary layer is typically shallower during winter, resulting in a higher aerosol concentration in the colder seasons confirmed by the results of Ma et al. (2014). Skjøth and Geels (2013) imply a dependency of ammonia emissions on the meteorological conditions which finally should have a strong influence on the resulting concentrations. This is obvious looking at the large differences between winter and summer shown here. The results presented in this paper are related to one year only (2008) and to its meteorological conditions, because the focus is on the effects of emission changes. However, because atmospheric chemistry and transport depend on meteorological conditions, the results of our study could vary slightly depending on the underlying meteorological year.

The strong NH_3 emission reduction in the RCAP scenario especially during winter is passed on to the reduction of NH_3 concentration in this season. Despite this reduction in ammonia concentration neither the SO_4^{2-} particles, nor the gaseous sulphuric acid have been reduced in the RCAP scenario. This suggests that even assuming a drastic reduction of ammonia, the ammonia concentration in the atmosphere is high enough to saturate the reaction forming SO_4^{2-} particles. In contrary to the reaction with H_2SO_4, the NH_3 concentration in the atmosphere is not high enough to saturate the reaction with HNO_3 to form ammonium nitrate (NH_4 NO_3) particles. This reduced formation of ammonium nitrate particles leads to a shift towards gas phase nitric acid, which is intensified in winter, where the $PM_{2.5}$ particle formation is reduced strongest. Also other studies argue that the aerosol formation is winter-sensitive to ammonia emissions (Aksoyoglu et al., 2011) and thus their reduction during this season. The balance has not only shifted towards gaseous nitric acid in the atmosphere, but also to a stronger formation of coarse mode particles over sea areas as presented in Fig. 6. This increased formation is linked to sea spray particles. The elevated concentration of gas phase HNO_3 leads to an increased condensation onto existing particles such as sodium chloride (NaCl). The replacement of Cl^- with NO_3^- results in an increasing gas phase concentration of HCl in the atmosphere.

Ammonia emission scenarios reported in other studies (Vuuren et al., 2011; Amann et al., 2014) often cover a smaller range of the possible developments of ammonia emissions as they assume progressive politic restrictions and technical solutions but focus very rarely on behavioural changes. This is why in most studies an increase in future ammonia emissions due to expanding livestock or climate warming is assumed (Amann et al., 2013; Skjøth and Geels, 2013). Presenting an independent emission scenario as proposed by Vuuren et al. (2011) this work goes beyond previous studies by investigating a larger range of possible NH_3 emissions reductions.

6. Conclusions

The effect of different mitigation strategies of NH_3 emissions with respect to the formation of particles has been discussed in this paper. The model results presented give an impression of a possible impact of the different approaches on air quality improvements. As the particle formation in the Aitken and accumulation modes due to the limitation of NH_3 is reduced, the ratio of NO_3^- and HNO_3 has shifted towards gaseous HNO_3. The reaction of H_2SO_4 and SO_4^{2-} has, not even in the extreme reduction scenario RCAP, been affected. The shift towards gas phase HNO_3 leads to a heightened condensation of HNO_3 onto existing sea spray particles as the NaCl, which results in an increasing gas phase concentration of HCL in the atmosphere. A slight shift from $PM_{2.5}$ aerosols to PM_{10} aerosols and gases has taken place. In the perspective of air pollution and human health a shift towards particles which are less respirable and whose formation takes mainly place over sea could be a possible development.

Furthermore, as the seasonal variability has been considered in this study, it can be concluded that the time of the year when NH_3 emissions are reduced is an important factor for the formation of particles. A shift of NH_3 emissions from winter to spring results in a strong decline of winter $PM_{2.5}$ and PM_{10} concentrations. This effect is heightened by different meteorological conditions favouring the condensation process (temperature and humidity) and the presence of reactants from intensified heating as explained in Section 5. Therefore it can be concluded that even though the ammonia emission is highest in spring and summer due to the application of manure on the fields, a reduction of the emissions in winter time has a stronger impact on the formation of secondary aerosols. The potential of reducing ammonia emissions in winter is highest through the reduction of animal farming. The RCAP scenario shows an annual reduction of total N deposited of up to 50% for grid cells located in areas with intensive livestock farming. This reduction could be achieved by a technical or a sufficiency strategy, devoted to the reduction of ammonia emissions from animal husbandry, as e.g. a choice of a diet low in meat. The main conclusion of this study, however, is the remarkable potential for air quality that a reduction of NH_3 emission in the cold season bears.

The comparison has shown that only minor improvements of air quality can be expected through a purely political approach (NEC2020) or an approach purely based on technical feasibility (MTFR). Even though an extreme reduction scenario as the RCAP involves strong behavioural changes and thus may presumably operate on a longer timescale, it underlines the effectiveness and potential of a sufficiency approach.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.11.039.

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