# Role of ammonia in European air quality with changing land and ship emissions between 1990 and 2030

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10 **Abstract.** The focus of this modeling study is on the role of ammonia in European air quality in the past as well as in the future. Ammonia emissions have not decreased as much as the other secondary inorganic aerosol (SIA) precursors nitrogen oxides ($NO_x$) and sulfur dioxide ($SO_2$) since 1990s, and are still posing problems for air quality and the environment. In this study, air quality simulations were performed with a regional chemical transport model at decadal intervals between 1990 and 2030 to understand the changes in the chemical species associated

15 with SIA under varying land and ship emissions. We analyzed the changes in air concentrations of ammonia, nitric acid, ammonium, particulate nitrate and sulfate as well as changes in the dry and wet deposition of ammonia and ammonium. The results show that the approximately 40% decrease in SIA concentrations between 1990 and 2010 was mainly due to reductions of $NO_x$ and $SO_2$ emissions. The ammonia concentrations on the other hand decreased only near the high emission areas such as the Netherlands and northern Italy by about 30% while there

20 was a slight increase in other parts of Europe. Larger changes in concentrations occurred mostly during the first period (1990-2000). The model results indicate a transition period after 2000 for the composition of secondary inorganic aerosols due to a larger decrease in sulfate concentrations than nitrate. Changes between 2010 and 2030 - assuming the current legislation (CLE) scenario - are predicted to be smaller than those achieved earlier for all species analyzed in this study. The scenario simulations suggest that if ship emissions will be regulated more

25 strictly in the future, SIA formation will decrease especially around the Benelux area, North Sea, Baltic Sea, English Channel and the Mediterranean region, leaving more ammonia in the gas phase which would lead to an increase in dry deposition. In the North, the decrease in SIA would be mainly due to reduced formation of particulate nitrate while the change around the Mediterranean would be caused mainly by decreased sulfate aerosol concentrations. Sensitivity tests with reduced $NO_x$ and $NH_3$ emissions indicate a shift in the sensitivity of aerosol

30 formation from $NH_3$ towards $NO_x$ emissions between 1990 and 2030 in most of Europe except the eastern part of the model domain.

## 1 Introduction

Ammonia ($NH_3$) plays an important role in atmospheric chemistry. As an alkaline gas, it affects the acidity of clouds and precipitation and it is one of the main sources of reactive nitrogen (Simpson et al., 2011; Fowler et al.,

35 2015). Studies show that ammonia emissions not only are toxic for plants and lead to a loss of biodiversity (Jones et al., 2014; Roth et al., 2015), but they also contribute significantly to the formation of particulate matter (Maas and Grennfelt, 2016). Ammonia reacts with sulfuric acid ($H_2SO_4$) and nitric acid ($HNO_3$) in the atmosphere to form secondary inorganic aerosols (SIA) such as ammonium sulfate (($NH_4)_2SO_4$) and ammonium nitrate ($NH_4NO_3$) which contribute most to the fine particulate matter ($PM_{2.5}$) in Europe (Ciarelli et al., 2016; 2019;

40 Aksoyoglu et al, 2017). Although ammonia and ammonium are nutrients for plants and are used as fertilizers, they are the largest contributors to nitrogen pollution of ecosystems through eutrophication and acidification (Dentener et al., 2006). The main sources of ammonia emissions are agricultural, including volatilization of animal waste and synthetic fertilizers but a small fraction (< 10%) comes also from other sources such as industry, household and traffic (UNECE, 2019).

45 European anthropogenic emissions have decreased substantially since the 1990s as a result of large emission reductions following the Gothenburg Protocol (GP) (UNECE, 1999), revised GP (revised on 4 May 2012,

https://www.unece.org/env/lrtap/multi_h1.html) and EU Directives (https://www.eea.europa.eu/data-and-maps/indicators/main-anthropogenic-air-pollutant-emissions/assessment-6). The largest decrease was in $SO_2$ emissions by more than 90% in 2017 compared to 1990, followed by $NO_x$ and NMVOC (non-methane volatile

50 organic compounds) emission reductions by more than 50% while ammonia emissions decreased less, approximately 23% on average in the EU-28 countries. Ammonia emissions have been increasing again since 2014, however, posing problems for Europe (NEC, 2019). The large decrease in sulfur emissions over the last few decades has changed the aerosol composition: particulate nitrogen was dominated by ammonium sulfate in the 1990s while today ammonium nitrate predominates (Colette et al., 2016).

55 Recent studies showed that the decline in nitrogen deposition in the past was mainly due to the decreased deposition of oxidized nitrogen components as a consequence of large emission reductions in Europe. Deposition of reduced nitrogen (ammonia $NH_3$, particulate ammonium $PNH_4$) was predicted to increase further in the future (Aksoyoglu et al., 2014; Simpson et al., 2014; Colette et al., 2016).

While land emissions have been significantly reduced over the last few decades, emissions from the least regulated

60 sector, maritime transport, have been increasing (Jonson et al., 2015). The International Maritime Organization (IMO) controls ship emissions globally through the Marine Pollution Convention (MARPOL) Annex VI, which limits the main air pollutants contained in ship exhaust gas and prohibits deliberate emissions of ozone depleting substances (ODS) (http://www.imo.org/OurWork/Environment/PollutionPrevention/Pages/Default.aspx). The revised MARPOL Annex VI with the aim of significantly strengthening the emission limits entered into force on

65 1 July 2010. In addition, emission control areas (ECAs) were introduced to reduce emissions further in designated sea areas. For example, in Europe, the North Sea and Baltic Sea areas were defined as SECAs (sulfur emission control areas), where the limits are set at 0.1%. New sulfur emission regulations, which reduce limits from 3.5% to 0.5%) outside the SECA areas in Europe, are expected to start in 2020 (http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Air-

70 Pollution.aspx, last access on 19 August 2020). On the other hand, there has been an increase in the emissions of other species, especially $NO_x$, in all European sea areas (Colette et al., 2016). The nitrogen emission control area (NECA) around the North Sea, Baltic Sea and the English Channel will enter into force in 2021 but only for newly-built ships (EEA, 2013). $NO_x$ emissions from all existing and new ships outside the NECA areas will continue to be under-regulated.

75 Based on recent studies, ship emissions are considered to be a major source of air pollution especially around the coastal areas of Europe (Jonson et al., 2019; Pay et al., 2019; Toscana and Murena, 2019). According to the European Environment Agency, emissions of nitrogen oxides from international maritime transport in European waters are projected to increase and could be equal to land-based sources by 2020 (EEA, 2013). Viana et al. (2014) reviewed a series of studies dealing with the impact of shipping emissions on air quality in the European coastal

80 areas and reported that ship emissions contribute with 1-14% to $PM_{2.5}$ and 7-24% to $NO_2$. In a recent model-intercomparison study, Karl et al. (2019) evaluated the contribution of ship emissions to air quality in the Baltic Sea region to investigate the differences among model predictions and showed that variations in ship-related $PM_{2.5}$ were mainly due to differences in the models' schemes for inorganic aerosol formation. Another study reported a contribution of 45% to $PM_{2.5}$ concentrations by ship emissions in the Mediterranean and 10-15% around the Baltic

85 Sea and concluded that the evolution of $NO_x$ emissions from ships and land-based $NH_3$ emissions would play a significant role in future European air quality (Aksoyoglu et al., 2016).

In a previous study, we investigated the changes in ozone and $PM_{2.5}$ during the period of 1990-2030 (Jiang et al., 2020). In the present paper, we focus on ammonia as an important precursor of secondary inorganic aerosols to investigate 1) how it affected the air quality in Europe between 1990 and 2010 when land emissions were reduced

90 significantly; 2) how it will affect the air quality between 2010 and 2030 when there will also be more strict reductions in ship emissions in addition to further reductions in land emissions and 3) how the sensitivity of aerosol formation to $NO_x$ and $NH_3$ emissions would vary between 1990 and 2030.

## 2 Method

We performed simulations over the European domain using the regional air quality model CAMx (Comprehensive

95 Air quality Model with eXtensions) version 6.50 (Ramboll, 2018) for 1990, 2000, 2010, 2020 and 2030. The model domain covers an area from 17°W to 39.8°E and from 32°N to 70°N with a horizontal resolution of 0.25° and 0.4°. We selected from the meteorological layers 14 terrain-following vertical layers ranging from 50 to 8000 m asl to be used in CAMx. The gas-phase chemical mechanism was Carbon Bond 6 Revision 2 (CB6r2) (Hildebrandt Ruiz and Yarwood, 2013). The fine/coarse option for the particle size was selected to calculate the

100 aerosol concentrations in the $PM_{2.5}$ fraction. Organic aerosols were calculated using the secondary organic aerosol chemistry/partitioning (SOAP2.1) module (Ramboll, 2018) and the ISORROPIA thermodynamic model was used for the partitioning of inorganic aerosol components (Nenes et al., 1998). Dry and wet deposition of species were calculated using the Zhang scheme (Ramboll, 2018).

Some of the input data used in this study were obtained from the EURODELTA-Trends (EDT) project (Colette

105 et al., 2017). The meteorological data in the EDT project was produced by the Weather Research and Forecast Model (WRF version 3.3.1) in the EuroCordex domain with a resolution of 0.44º. We re-gridded the data to our model domain and prepared the meteorological input parameters for the CAMx model by means of the preprocessor WRFCAMx version 4.4 (http://www.camx.com/download/support-software.aspx). Another input dataset provided by the EDT project was the initial and boundary conditions which were based on monthly

110 climatological data (Colette et al., 2017). The ozone column densities were prepared using the Total Ozone Mapping Spectrometer (TOMS) data from NASA, and photolysis rates were calculated using the Tropospheric Ultraviolet and Visible (TUV) Radiation Model version 4.8.

Anthropogenic emissions for the three base cases (1990, 2000 and 2010) were obtained from the EDT database and adjusted to the CB6r2 chemical mechanism in CAMx as described in Jiang et al. (2020). The biogenic

115 emissions (isoprene, monoterpenes, sesquiterpenes and soil-NO) were generated using the Model of Emissions of Gases and Aerosol from Nature (MEGAN) version 2.1 (Guenther et al., 2012). The anthropogenic emissions for scenarios in 2020 and 2030 were prepared using the current legislations in Europe according to the revised Gothenburg Protocol (revised on 4 May 2012, https://www.unece.org /env/lrtap/multi_h1.html) and the National Emissions Ceilings (NEC) Directive (2016/2284/EU), respectively. The ship emissions in 2020 and 2030 are

120 projected based on current legislation (CLE) of the International Maritime Organization (IMO) and EU (2020_CLE, 2030_CLE). We also carried out two more scenarios for 2020 and 2030 in which ship emissions were kept the same as in 2010 to estimate the expected changes in their contributions to air pollution. Another scenario, 2030_MTFR (maximum technically feasible emission reductions) was used to investigate the effect of maximum reductions of ship emissions (Jiang et al., 2020). In all scenarios for 2020 and 2030, meteorology and

125 boundary conditions were kept the same as in 2010 and only anthropogenic emissions were changed.

Some additional tests were also performed to determine whether there has been any change in the sensitivity of aerosol formation to $NO_x$ and $NH_3$ emissions due to changes in the European emissions over the four decades. The simulations for 1990 and 2030_CLE were repeated with 30% reductions in $NO_x$ and $NH_3$ emissions: (1) 1990 with 70% $NO_x$, (2) 1990 with 70% $NH_3$, (3) 2030_CLE with 70% $NO_x$, (4) 2030_CLE 70% $NH_3$. The modelled

130    SIA concentrations in each case were compared with those in the corresponding base cases, *i.e.* 1990 and 2030_CLE, respectively. A description of all simulations performed in this study is shown in Table 1.

**3 Results and Discussion**

**3.1 Model evaluation**

The model results for 1990, 2000 and 2010 were compared with the measurements available at the EDT project

135    database based on EMEP datasets and model performance for $SO_2$, $NO_2$, $PM_{10}$, $PM_{2.5}$ and hourly $O_3$ was discussed in detail in Jiang et al. (2020). In the present study, we performed additional evaluations for ammonia, total ammonium, total nitrate and secondary inorganic aerosol components. Since measurements with large spatial and temporal coverage for such species are scarce, only the 2010 measurements from the EBAS database (http://ebas.nilu.no, last access: 10 July 2020) were compared with the model results.

140    The number of measurement sites for ammonia is relatively small (Table S1) and they are mostly located in the north around Scandinavian countries; only very few stations are in other parts of Europe (Fig. 1). Modelled ammonia concentrations are similar to the measured ones at the sites in the south while one site in eastern Europe shows an underestimation. On the other hand, ammonia is overestimated in high emission areas around the Netherlands and Denmark (Fig. 1). Recent studies show that a better agreement between model and measurements

145    can be achieved when ammonia emissions are modulated with local meteorological conditions (Backes et al., 2016; Hendriks et al., 2016). Overestimation might originate from either overestimated emissions or underestimated removal (deposition, particle formation). Comparison of measured and modeled total ammonium (sum of gaseous ammonia and particulate ammonium) is therefore an additional test for model evaluation. There are more stations with measurements of total ammonium and total nitrate (sum of nitric acid and particulate nitrate)

150    than for ammonia (Table S1).

Statistical parameters indicate an overestimation for total ammonium and nitrate, the variation of the mean bias among the measurement sites, however, suggests that overestimation mostly occurs around high emission areas in central Europe, while modelled and measured concentrations are similar at most of the sites, especially in the Iberian Peninsula and in Scandinavia (Fig. 1). These results suggest that ammonia emissions in the emission

155    inventory might be too high around the main emission sources in central Europe and/or deposition is underestimated by the model for which the resolution might also be critical factor.

Evaluation of total nitrogen deposition is challenging because of a lack of measurements, especially of dry deposition, estimates are therefore based on the concentrations and deposition velocities (Flechard et al., 2011). On the other hand, model performance for wet deposition depends strongly on the performance of the

160    meteorological model (Vivanco et al., 2018). Our model results for wet deposition of WNHx, WNOx and WSOx in 2010 are shown together with the available measurements in Fig. S1. The correlation between model results and measurements for wet deposition is high (between 0.61 for WSOx and 0.81 for WNHx), the wet deposition of

all three species, however, is underestimated with the mean fractional bias (MFB) ranging from -40% to -58% (Table S1).

165 Measurements of total nitrate and ammonia concentrations are mainly available for northern Europe and they have very little overlap with the wet-deposition sites in central and western Europe (see Figs. 1b,c and Fig. S1). Comparison of model performance for TNHx and WNHx with observations at 15 common sites (Fig. S2) suggests that at 9 stations overestimation of TNHx might partially be attributed to underestimation of WNHx (with the largest anti-correlation of bias at sites in the Czech Republic and Poland), as also found by other models used in

170 the EDT model intercomparison study (Theobald et al., 2019).

Among the SIA components, the best agreement between model and measurements is for sulfate (Table S1, Fig. 1). The modeled concentrations of the other SIA components - for which the spatial coverage in central and western Europe is rather poor - are higher than the measured ones, especially for nitrate (Fig. 1, Table S1). Overall, the performance of CAMx model in this study is similar to the other models participating in the EDT project

175 (Ciarelli et al., 2019; Theobald et al., 2019).

### 3.2 Changes in concentrations

### 3.2.1 Gaseous species: $NH_3$ and $HNO_3$

The highest ammonia concentrations are predicted around the Benelux area and northern Italy where emissions are the highest in Europe (Fig. 2a, left panel). The model results suggest that between 1990 and 2000, the annual

180 average concentrations of ammonia decreased only around the Netherlands and in the eastern part of the model domain by about 30-40%, while an increase was predicted in the rest of the domain (Fig. 2b, left panel, see Fig. S3a for relative changes). This is consistent with the observed trends in Europe during the same period (Colette et al., 2016). Ammonia concentrations decreased further around the Netherlands and started to decline also in northern Italy between 2000 and 2010, while they continued to increase in other parts of Europe (Fig. 2c, left

185 panel) The predictions based on the current legislation (CLE) scenario emissions suggest that the changes during the period between 2010 and 2030 will be much smaller (Fig. 2d, left panel).

Concentrations of nitric acid ($HNO_3$) are generally higher in areas with both high $NO_x$ emissions and photochemical activity, such as coastal areas and the shipping routes especially in the Mediterranean region (Fig. 2a, right panel). The large decline (50-60%) in nitric acid concentrations in those areas between 1990 and 2000 is

190 mainly due to a decrease in $NO_x$ emissions as a result of significant reductions in emission standards in Europe (Figs. 2b, right panel, S3b). The results show an increase in nitric acid concentrations later between 2000 and 2010 around the English Channel, North Sea, Baltic Sea and the Mediterranean Sea as well as northern Italy (Fig. 2c, right panel). These results suggest that decreased ammonia concentrations in the same period around the Netherlands, Denmark and northern Italy led to a lower ammonium nitrate formation and higher nitric acid

195 concentrations in the air. Our calculations suggest that with further reductions in $NO_x$ emissions, nitric acid levels would continue to decrease until 2030 due to reductions in both land and ship emissions (Fig. 2d, right panel). On the other hand, since simulations for 2030 were performed using the meteorological parameters of 2010, one should keep in mind that potentially higher temperatures in the future might increase the evaporation of ammonium nitrate to form its gaseous components $NH_3$ and $HNO_3$ (Fowler et al., 2015).

### 3.2.2 Secondary inorganic aerosols

Annual average concentrations of SIA components ($PNH_4$, $PNO_3$, $PSO_4$) in 1990, as well as changes in their concentrations between 1990-2000, 2000-2010 and 2010-2030_CLE are shown in Fig. 3. The highest ammonium concentrations are predicted for central Europe and the Po Valley (Fig. 3a, left panel). The concentrations in eastern Europe are relatively high as well. As seen in the left panel of Fig. 3b, the ammonium concentrations decreased significantly (by 40-50%) between 1990 and 2000 in central and eastern Europe (see Fig. S3c for relative changes) while there was some increase in the east after 2000 (Fig. 3c, left panel). Simulations using the CLE emission scenarios suggest that the ammonium concentrations will continue to decrease between 2010 and 2030 with no more increase anywhere in the whole model domain (Fig. 3d, left panel). A similar trend can be observed in the particulate nitrate concentrations, with a significant decrease over the continent between 1990 and 2010 (Fig. 3 a-c, middle panels). A slight increase, however, is noticeable over the Baltic Sea and Scandinavia between 1990 and 2000 as well as in the eastern part of the model domain between 2000 and 2010. On the other hand, particulate nitrate concentrations will decrease in all of Europe between 2010 and 2030 (Fig. 3d, middle panel).

The concentrations of particulate sulfate ($PSO_4$) in 1990 were higher in central and eastern Europe, the Balkans and along shipping routes than the rest of Europe (Fig. 3a, right panel). They decreased continuously (by 60-70%) over the period between 1990-2010 (Fig. 3b-c, right panels, Fig. S3e). Results of future scenario simulations suggest that sulfate concentrations will continue to decrease in central Europe as well as along shipping routes until 2030 assuming a current legislation (CLE) scenario (Fig. 3d, right panel).

The results obtained from these simulations indicate that a significant reduction (> 40%) in the secondary inorganic aerosol concentrations was achieved especially between 1990 and 2000 (Fig. S3f), consistent with the larger reductions in emissions during the first decade (Table 2). Continuous reductions of land and ship emissions until 2030 will lead to a further decrease in SIA concentrations in Europe. The model results suggest that the relative composition of secondary inorganic aerosols will be different in 2030 compared to 1990 due to a larger decrease in sulfate concentrations than nitrate in most of the model domain. An example for the change in the SIA composition is shown for Payerne, a rural site in Switzerland, in Fig. S4. The sulfate fraction decreases from 30% to 21% while the particulate nitrate fraction increases from 47% to 56% between 1990 and 2030, assuming the CLE scenario.

## 3.3 Changes in deposition

### 3.3.1 Dry deposition

A large fraction of the total nitrogen deposition in Europe is due to the deposition of reduced nitrogen compounds, with dry deposition of ammonia being the dominant one (Aksoyoglu et al., 2014; Aksoyoglu and Prévôt, 2018; Simpson et al., 2014). Removal of ammonia from the atmosphere through dry deposition is quite fast - *i.e.* deposition occurs in areas close to the emission sources (Fig. 4a, left panel). The model results show that dry deposition of ammonia decreased around the Netherlands, northern Germany and slightly also in the eastern part of the model domain between 1990 and 2000 while there was an increase in the rest of Europe (Fig. 4b, left panel). After 2000, however, dry deposition started increasing also in eastern Europe (Fig. 4c, left panel). Simulations for 2030 calculated by changing only the emissions according to the CLE scenario suggest that there will be a small decrease in dry deposition only in the north around the Netherlands and northern Germany between 2010 and

2030 (Fig. 4d, left panel). A slight increase along the coastal areas in western and northern Europe is probably
240     caused by the changes in ship emissions (see Section 3.4).

### 3.3.2 Wet deposition

The amount of precipitation which is generated by the meteorological models is crucial for simulating wet
deposition in air quality models. The wet deposition of ammonium in 1990 is shown in Fig. 4a, right panel. The
modelled wet deposition decreased in eastern Europe between 1990 and 2000 while it slightly increased around
245     the English Channel and North Sea (Fig. 4b, right panel). The decrease in the east is in line with the largest
emission reductions in that area. The increase in wet deposition in the north around the English Channel could be
due to increased precipitation between 1990-2000 (Theobald et al., 2019). Increased emissions from ships,
however, could also be the reason for the increased wet deposition of ammonium. After 2000, there was a decrease
in wet deposition in most of the domain (Fig. 4c, right panel). Other models which participated in the EDT project
250     and simulated the whole 21-year period between 1990 and 2010, found similar results with a decrease in wet
deposition in the east and an increase in north-west Europe during the first period between 1990-2000 (Theobald
et al., 2019). Assuming the CLE scenario (using the meteorology of 2010), wet deposition of ammonium is
predicted to decrease in Europe significantly (20-40%) between 2010 and 2030 (Fig. 4d, right panel). The change
in wet deposition between 1990 and 2010 might be due to a change in both the air concentrations and the amount
255     of precipitation, but it can only be due to a change in the concentrations between 2010 and 2030_CLE since the
same meteorological parameters were used for both years.

### 3.4 Effects of ship emissions

The two scenarios 2020_CLE and 2030_CLE take both land and ship emissions into account according to the
current legislation. In order to investigate the effect of ship emissions on the gaseous and particulate species, we
260     compared two ship emission scenarios for both 2020 and 2030 as described in Table 1. In scenarios 2020_CLEland
and 2030_CLEland, ship emissions were kept the same as in 2010 while they were projected to 2020 and 2030
using the CLE scenario in 2020_CLE and 2030_CLE, respectively. The difference in concentrations between CLE
and CLEland scenarios therefore shows the effect due to the changes in ship emissions in the corresponding years
(Fig. 5, Fig. S5).

265     In all three cases (2020, 2030_CLE and 2030_MTFR), gaseous ammonia concentrations are predicted to increase
due to changes in ship emissions especially around the Benelux area and along the Mediterranean coast (Fig. 5a).
On the other hand, nitric acid concentrations will decrease in the North Sea, Baltic Sea and the Mediterranean
Sea, and increase along the Atlantic coast under the assumptions of the CLE scenario in 2020 and 2030 (Fig. 5b).
The effect of emissions from international shipping activities along the Atlantic coast and Gibraltar Strait can also
270     be seen in the particulate nitrate concentrations (Fig. S5d). The importance of shipping activities due to their
relatively high NOx emissions was also reported for south-west Europe by Pay et al. (2019). Our scenario
calculations suggest that when ship emissions are reduced according to the MTFR scenario in 2030, nitric acid
and particulate nitrate would no longer increase along the Atlantic coast but decrease (see Fig. 5b, Fig. S5d).
The decrease in concentrations of secondary inorganic aerosols along the coastal areas -especially with the MTFR
275     scenario- (Figs. 5c, S5) is due to a significant decrease in nitrate and sulfate concentrations around the Benelux
region and the Mediterranean, respectively, as shown in Jiang et al. (2020). These results suggest that when ship

emissions are reduced further in the future, particulate ammonium formation will decrease especially around the Benelux area, North Sea, Baltic Sea, English Channel and the Mediterranean region, leaving more ammonia in the gas phase (Figs. 5a, S5a). This would then lead to an increase in the dry deposition of ammonia along the

280 coastal areas (Fig. 5d, S5g). On the other hand, wet deposition of ammonium will decrease along the Scandinavian and Adriatic coasts due to future reductions of ship emissions (Fig. S6).

### 3.5 Sensitivity of aerosol formation to $NO_x$ and $NH_3$ emissions

SIA formation depends strongly on its precursor emissions. Ammonia reacts rapidly with atmospheric sulfuric and nitric acids to form ammonium sulfate and ammonium nitrate (Behera et al., 2013). Reaction with sulfuric

285 acid (or with ammonium bisulfate) is favored over the reaction with nitric acid; ammonium nitrate is formed only after all sulfate is neutralized by $NH_3$. Ammonium nitrate formation, which is favored by low temperatures and high relative humidity is in a reversible equilibrium with ammonia and nitric acid.

An earlier model study covering Switzerland and northern Italy, using emissions from the year 2000, showed that aerosol formation was rather limited by $NO_x$ emissions in northern Switzerland while it was dependent on both

290 $NO_x$ and $NH_3$ emissions in northern Italy (Andreani-Aksoyoglu et al., 2008). Other sensitivity studies performed over the whole European domain for the period 2004-2006 suggested that SIA formation was more sensitive to $NH_3$ emissions in most of Europe, except the Netherlands, northern Switzerland and north-western France where ammonia emissions are high (Aksoyoglu et al., 2011; Pay et al., 2012). Since sulfate and nitrate aerosol formation depends on the availability of their precursors $SO_2$, $NO_x$ and $NH_3$, changes in emissions since the 1990s might

295 have affected the sensitivity of aerosol formation in Europe.

We tested the sensitivity of aerosol formation to emissions by reducing $NO_x$ and $NH_3$ emissions by 30% in two separate simulations for the past (1990) and the future (2030_CLE). The change in the annual average SIA concentrations for these simulations is shown in Fig. 6. Decreasing $NH_3$ emissions are predicted to be more effective in reducing SIA concentrations in 1990 for a large part of the model domain (Fig. 6, upper panels) as

300 also reported in Aksoyoglu et al. (2011). In 2030_CLE, however, the effectiveness of emission reductions looks different as a result of the larger reductions in $NO_x$ emissions compared to $NH_3$ emissions in Europe (Fig. 6, lower panels). The change in the colors from red in 1990 to blue in 2030_CLE in central and western Europe as well as the UK (right panels) suggests that aerosol formation will become more $NO_x$-sensitive in 2030 in those areas while in the eastern part of the model domain it will still be more sensitive to ammonia emissions (Fig. 6, lower

305 right panel). It should be noted however, that the sensitivity to emissions is weaker in 2030_CLE than in 1990 (see scales in Fig. 6).

### 4 Conclusions

In this study we investigated the role of ammonia in European air quality by means of CAMx model simulations for the period between 1990 and 2030 with 10-year intervals. We analyzed the modelled annual average

310 concentrations of ammonia ($NH_3$), nitric acid ($HNO_3$), secondary inorganic aerosol (SIA) and its components as well as dry ammonia and wet ammonium deposition. The model results suggest that the decrease in SIA concentrations by about 40% between 1990 and 2010 was mainly due to reductions of $NO_x$ and $SO_2$ emissions in Europe. Ammonia concentrations, on the other hand, decreased by about 30% only around high emission areas

(Benelux area, northern Italy) while there was a slight increase in the other parts of the model domain, leading
315 also to an increase in dry deposition of ammonia. The modeled changes in the annual concentrations were larger
for the first decade (1990-2000), especially for nitric acid, particulate ammonium and sulfate. On the other hand,
changes during the period between 2010 and 2030, assuming the current legislation (CLE) scenario, are predicted
to be smaller than those achieved earlier for all analyzed species in this study.

Simulations using the current legislation (CLE) and maximum technically feasible reduction (MTFR) scenarios
320 for 2020 and 2030 suggest that when ship emissions will be regulated more strictly in the future, particle formation
will decrease, especially around the Benelux area, North Sea, Baltic Sea, English Channel and the Mediterranean
region, leaving more ammonia in the gas phase, which will lead to an increase in dry deposition. In the north, the
decrease in aerosol concentrations will be mainly due to reduced particulate nitrate formation, while the change
in the Mediterranean area will be caused mainly by decreased sulfate aerosols.

325 In order to investigate whether the sensitivity of aerosol formation to $NO_x$ or $NH_3$ emissions changes during the
period between 1990 and 2030, we performed sensitivity tests by repeating the simulations for 1990 and
2030_CLE with $NO_x$ and $NH_3$ emissions reduced by 30%, separately. In 1990, SIA formation was more affected
by $NH_3$ emission reductions in central Europe because of relatively high $NO_x$ emissions at that time. In 2030,
however, $NO_x$ emission reductions reduce SIA concentrations more than $NH_3$ reductions. These results indicate a
330 shift in the sensitivity of aerosol formation from $NH_3$ towards $NO_x$ emissions in a large part of Europe between
1990 to 2030 due to a larger change in $NO_x$ levels during that period compared to changes in $NH_3$ concentrations.

*Data availability:* Data will be available online before publication on ACP

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**Table 1.** Description of simulations

| Name | Meteorology | Boundary conditions | Anthropogenic Emissions |
|---|---|---|---|
| **Base Cases** | | | |
| 1990 | 1990 | 1990 | 1990 |
| 2000 | 2000 | 2000 | 2000 |
| 2010 | 2010 | 2010 | 2010 |
| **Scenarios** | | | |
| 2020_CLE | 2010 | 2010 | 2020CLE |
| 2020_CLEland | 2010 | 2010 | 2020CLE land, 2010 ship emissions |
| 2030_CLE | 2010 | 2010 | 2030CLE |
| 2030_CLEland | 2010 | 2010 | 2030CLE land, 2010 ship emissions |
| 2030_MTFR | 2010 | 2010 | 2030CLE land, MTFR ship emissions |
| **Sensitivity Tests** | | | |
| 1990_70%$NO_x$ | 1990 | 1990 | 1990, $NO_x$ emissions reduced by 30% |
| 1990_70%$NH_3$ | 1990 | 1990 | 1990, $NH_3$ emissions reduced by 30% |
| 2030_70%$NO_x$ | 2010 | 2010 | 2030CLE, $NO_x$ emissions reduced by 30% |
| 2030_70%$NH_3$ | 2010 | 2010 | 2030CLE, $NH_3$ emissions reduced by 30% |

**Table 2.** Differences in emissions between various years and future scenarios

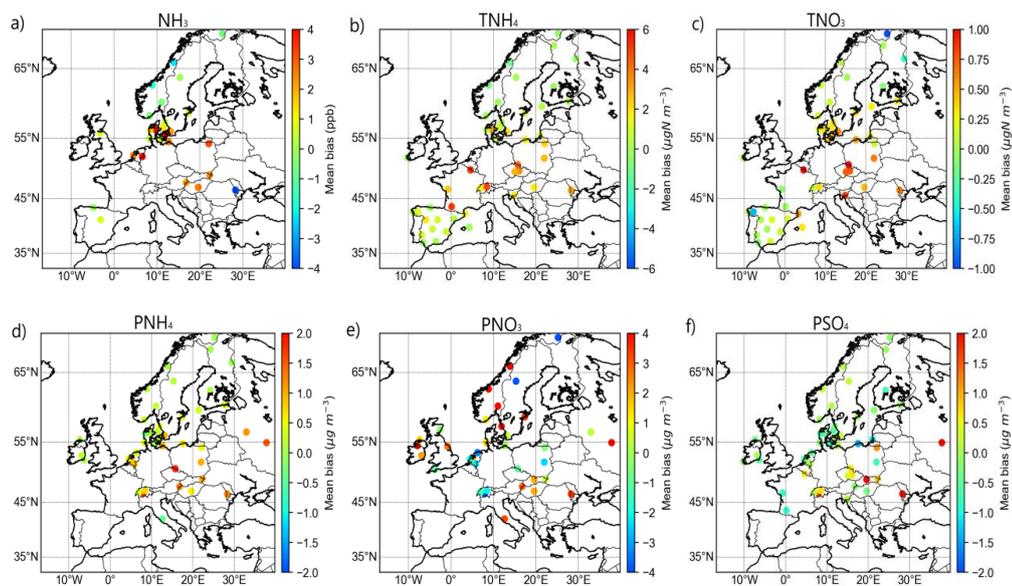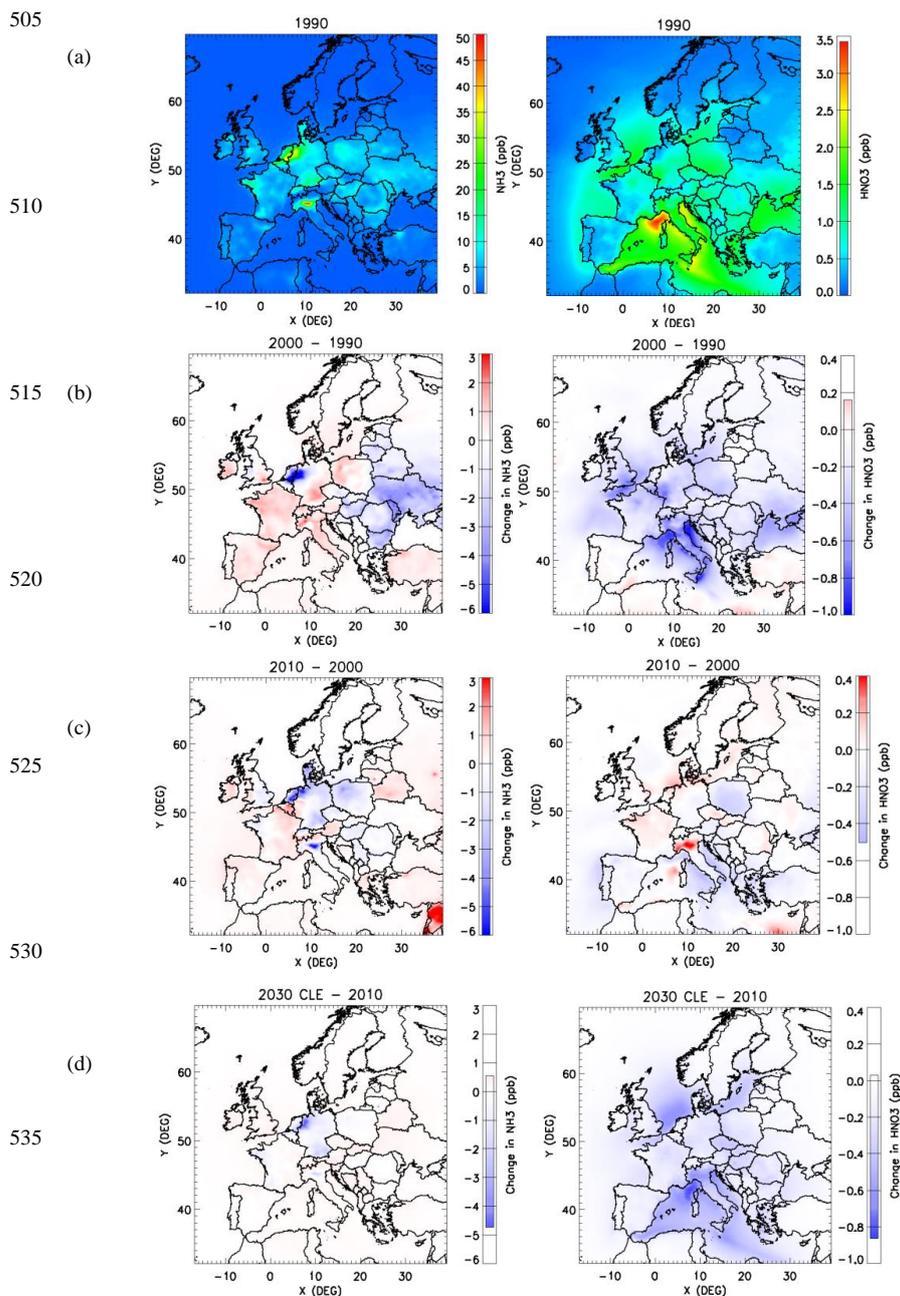| | Changes in emissions (%) | | |
|---|---|---|---|
| | $NO_x$ | $SO_x$ | $NH_3$ |
| 1990-2000 | -23.7 | -51.0 | -18.6 |
| 2000-2010 | -16.8 | -35.3 | -5.3 |
| 1990-2010 | -36.5 | -68.3 | -23.0 |
| 2010-2020_CLE | -28.1 | -41.5 | -4.1 |
| 2010-2030_CLE | -39.7 | -47.5 | -10.8 |
| 2010-2030_MTFR | -52.5 | -49.7 | -10.8 |

490

**Figure 1: Mean bias between measured and modelled concentrations of (a) ammonia (µg N m⁻³), (b) total ammonia (µg N m⁻³), (c) total nitrate (µg N m⁻³), (d) ammonium (µg m⁻³), (e) particulate nitrate (µg m⁻³) and (f) sulfate (µg m⁻³) at various sites in 2010.**

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**Figure 2: Ammonia (left) and nitric acid (right): (a) annual average concentrations (ppb) in 1990, changes (in ppb) between (b) 1990-2000, (c) 2000-2010 and (d) 2010-2030_CLE.**

545 (a)

550

(b)

555

(c)

560

565

(d)

570
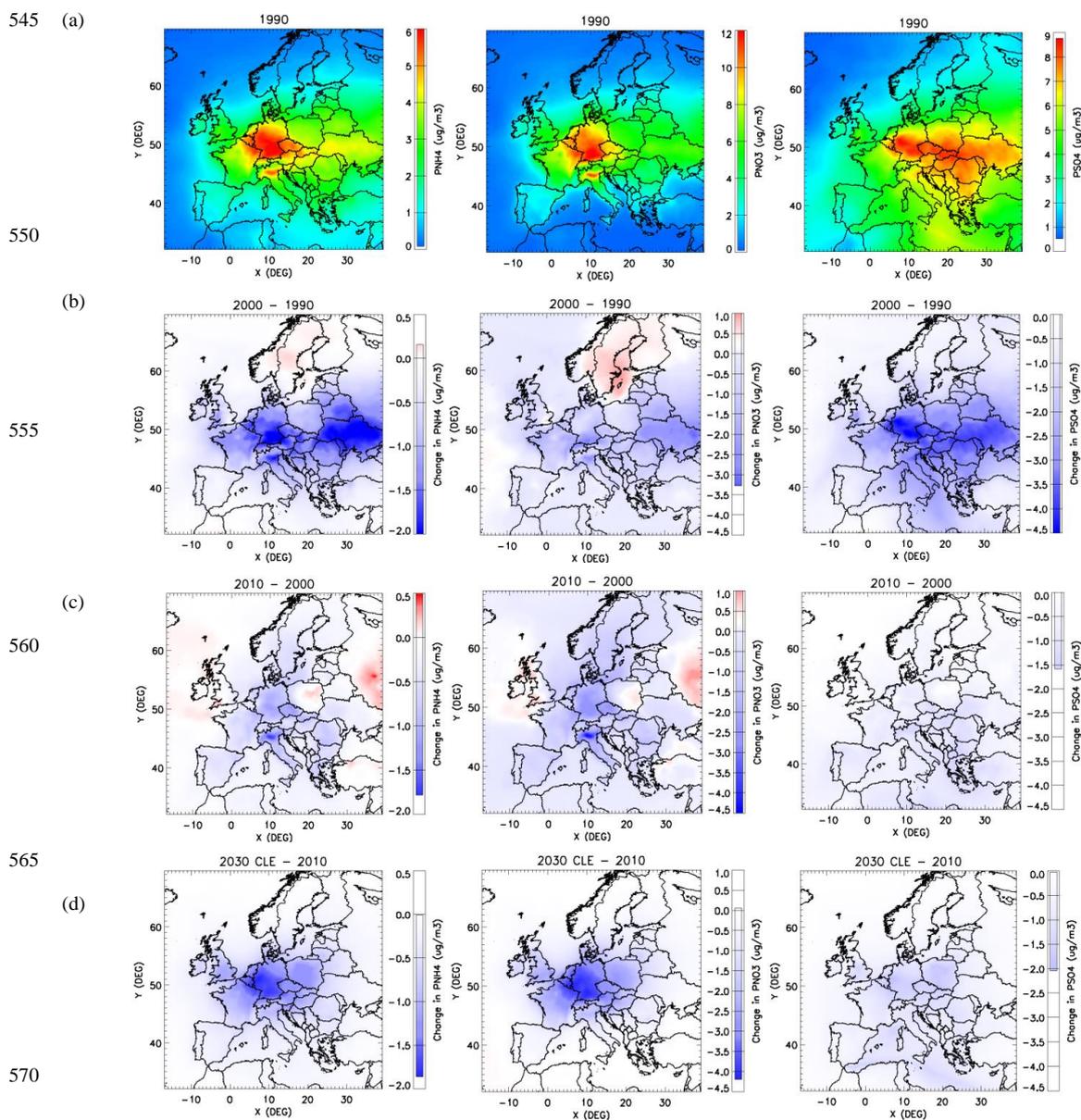


**Figure 3: Ammonium (left), particulate nitrate (middle) and sulfate (right): (a) annual average concentrations (μg m⁻³) in 1990, changes (μg m⁻³) between (b) 1990-2000, (c) 2000-2010 and (d) 2010-2030_CLE.**
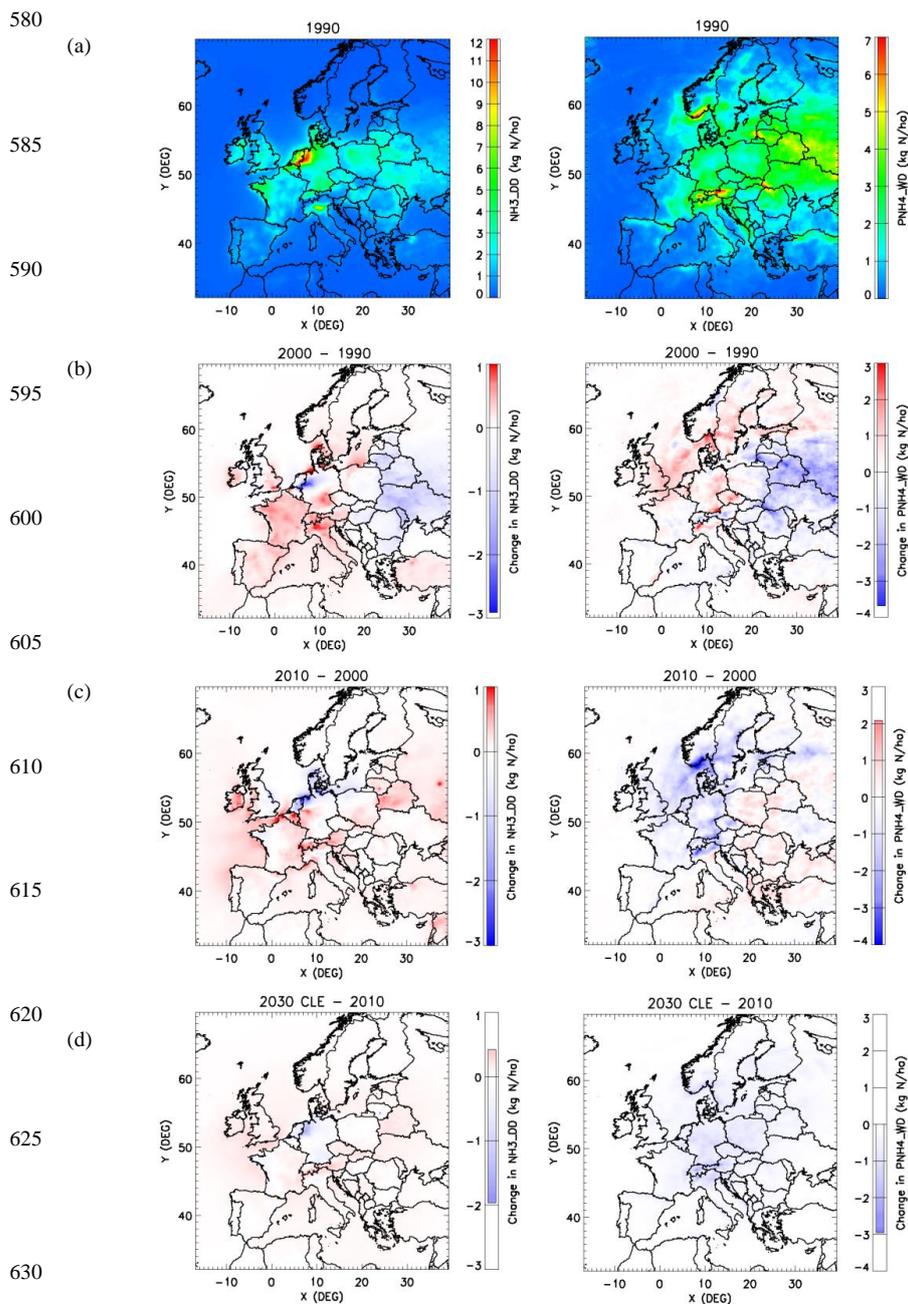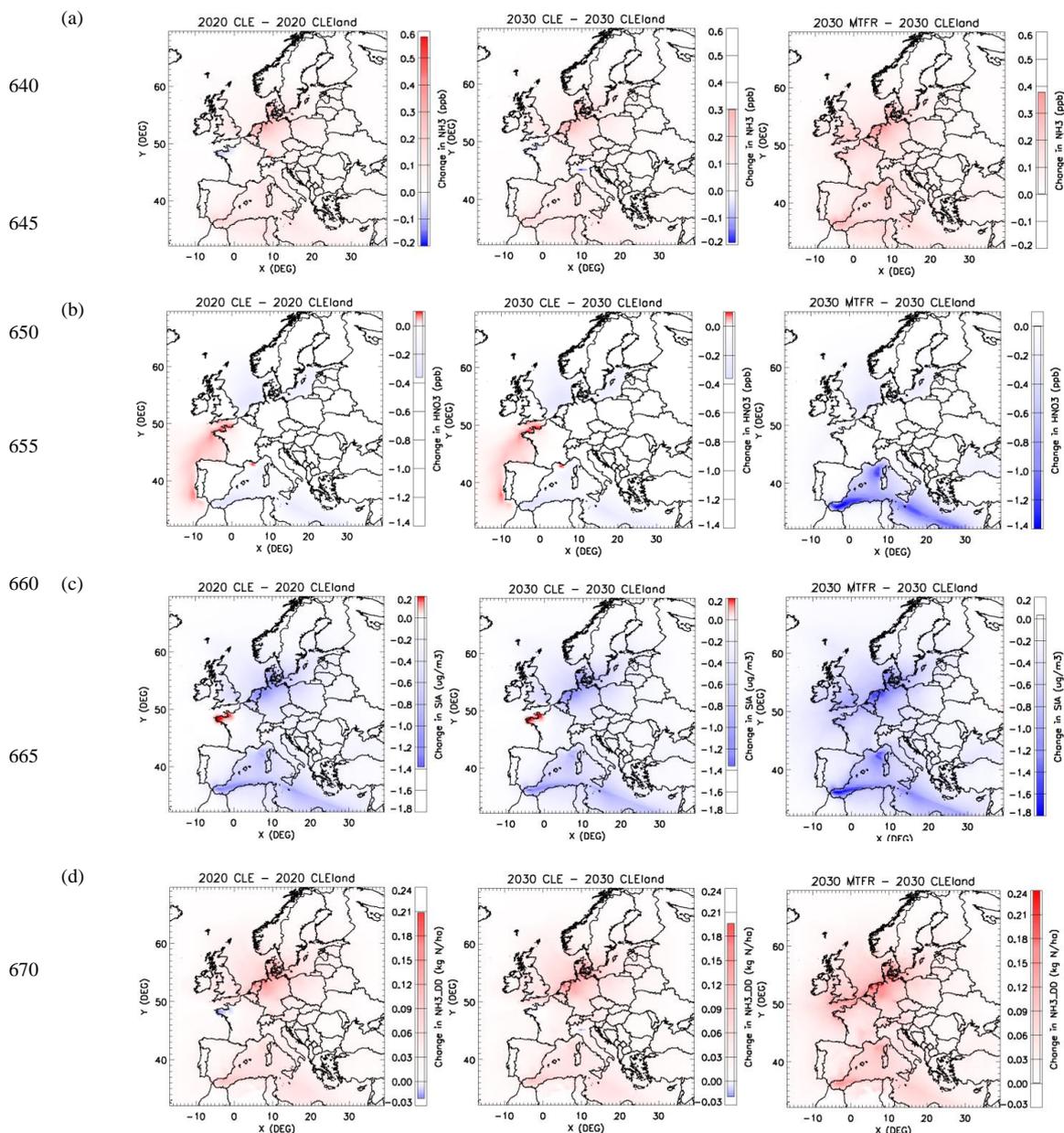
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**Figure 4: Dry deposition of ammonia (left panels) and wet deposition of ammonium (right panels): (a) accumulated deposition (kg N ha⁻¹) in 1990, changes (in kg N ha⁻¹) between (b) 1990 and 2000, (c) 2000 and 2010 and (d) 2010 and 2030_CLE.**

**Figure 5: Effect of ship emissions on annual average concentrations of (a) ammonia (ppb), (b) nitric acid (ppb), (c) secondary inorganic aerosols (µg m$^{-3}$) and (d) on dry deposition of ammonia (kg N ha$^{-1}$) in scenarios 2020 CLE (left), 2030 CLE (middle) and 2030 MTFR (right). CLEland: with ship emissions of 2010; CLE: with ship emissions under current legislation; MTFR: with ship emissions under maximum technically feasible reductions.**
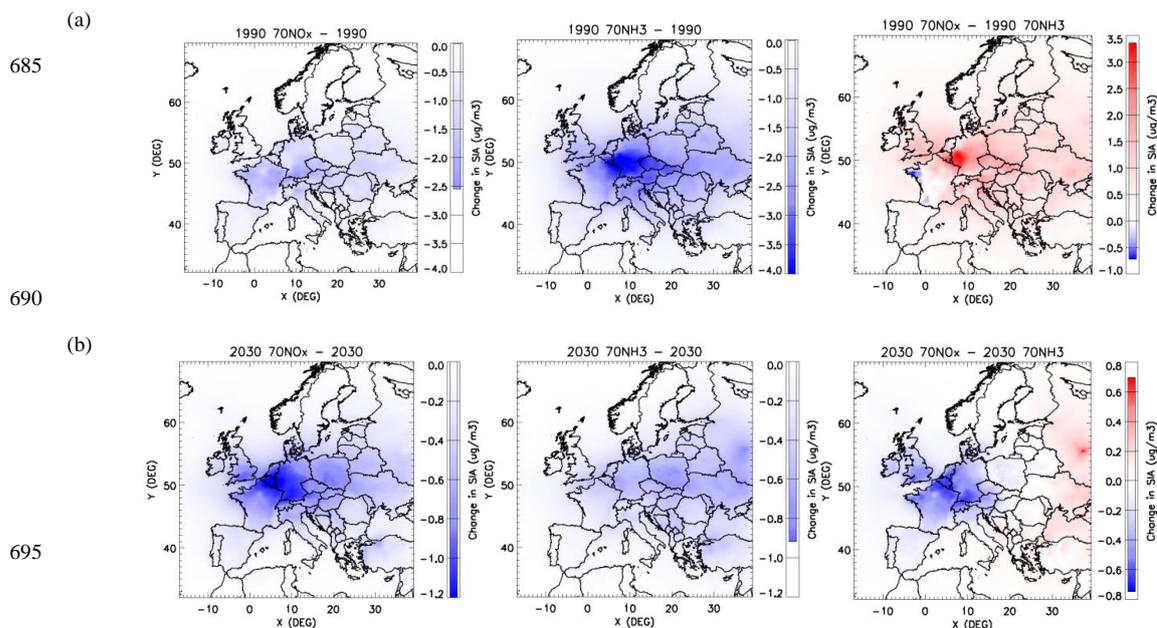
**Figure 6: Change in SIA concentrations (μg m⁻³) due to 30% reduction of NO$_x$ emissions (left), due to 30% reduction of NH$_3$ emissions (middle) and the difference between the two figures (right) (a) in 1990 and (b) in 2030_CLE. Red color in the right panels shows NH$_3$-sensitive areas; blue color shows NO$_x$-sensitive areas.**