



## RESEARCH LETTER

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## Key Points:

- PM<sub>2.5</sub> from agricultural sources dominate over nonagricultural and naturally occurring PM<sub>2.5</sub>
- By the end of the century, agriculture-driven aerosol formation will be reduced near to the Earth surface
- Source-receptor scenarios of emission reduction are testing ammonia versus nitrogen oxide reductions

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## Significant atmospheric aerosol pollution caused by world food cultivation

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**Abstract** Particulate matter is a major concern for public health, causing cancer and cardiopulmonary mortality. Therefore, governments in most industrialized countries monitor and set limits for particulate matter. To assist policy makers, it is important to connect the chemical composition and severity of particulate pollution to its sources. Here we show how agricultural practices, livestock production, and the use of nitrogen fertilizers impact near-surface air quality. In many densely populated areas, aerosols formed from gases that are released by fertilizer application and animal husbandry dominate over the combined contributions from all other anthropogenic pollution. Here we test reduction scenarios of combustion-based and agricultural emissions that could lower air pollution. For a future scenario, we find opposite trends, decreasing nitrate aerosol formation near the surface while total tropospheric loads increase. This suggests that food production could be increased to match the growing global population without sacrificing air quality if combustion emission is decreased.

### 1. Introduction

World food production is deeply connected to the Earth's reactive nitrogen cycle. The Nobel prize-winning Haber-Bosch process that converts atmospheric molecular nitrogen (N<sub>2</sub>) into ammonia (NH<sub>3</sub>) revolutionized the production of synthetic fertilizers that provides reactive nitrogen (Nr) to crops. It is estimated that 40% of the people alive today owe their life to the use of fertilizers produced by the Haber-Bosch process [Smil, 2001]. The addition of anthropogenic Nr into the atmosphere, the hydrosphere, and the biosphere has a wide variety of consequences, which are magnified as Nr moves along its biogeochemical pathway, known as the Nr cascade [Smil, 2001; Galloway et al., 2003]. Nr is now accumulating in the environment due to its human production, which has surpassed all natural sources, such as biological fixation and lightning, since the midtwentieth century [Erismann et al., 2008; Aneja et al., 2009]. In the past 70 years, global ammonia emissions have more than doubled, from 23 Tg/yr to 60 Tg/yr [Lamarque et al., 2010]. This increase is entirely attributed to NH<sub>3</sub> emissions from agriculture, with N fertilizer use contributing 33% and livestock production 66% [Bouwman et al., 1997; Beusen et al., 2008]. The emissions from livestock production come from animal houses and storage systems, animal manure, and grazing [Beusen et al., 2008]. Other sources of ammonia are the oceans (16%, in relation to total emission) and biomass burning (17%); nonagricultural anthropogenic sources contribute only 2% to global NH<sub>3</sub> emissions [Lamarque et al., 2011].

Anthropogenic nitrate aerosols are created by NH<sub>3</sub> and nitric oxide (NO<sub>x</sub>) emissions. The ammonia is largely supplied by agriculture, while NO<sub>x</sub> results from combustion. Gaseous ammonia in the atmosphere leads to secondary inorganic aerosol formation of ammonium compounds, including ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>), ammonium bisulfate (NH<sub>4</sub>HSO<sub>4</sub>), and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). The phase partitioning among these inorganic aerosols is driven by NH<sub>3</sub> concentrations, acids in the atmosphere, and environmental conditions. Inorganic aerosols are the main constituents of anthropogenic PM<sub>2.5</sub> pollution, the fine particulate matter with diameters below 2.5 μm. In the eastern United States, approximately half of the PM<sub>2.5</sub> has an inorganic chemical signature and is composed of ammonium (NH<sub>4</sub><sup>+</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and sulfate (SO<sub>4</sub><sup>2-</sup>) [Henze et al., 2009]. Fine particulate matter poses a significant health hazard, causing lung cancer and cardiopulmonary mortality [Pope et al., 2002], and significant amounts of outdoor air pollution are attributed to agriculture, enhancing premature mortality rates [Lelieveld et al., 2015]. The U.S. Environmental Protection Agency and the European Union have established ambient standards for PM<sub>2.5</sub> of 12 μg/m<sup>3</sup> and 25 μg/m<sup>3</sup>, respectively, on an annual average basis. However, the World Health Organization (WHO) advises concentrations below 10 μg/m<sup>3</sup> [World Health Organization Regional Office for Europe, 2006].

Studies with regional air quality models investigated source-receptor relationships between  $\text{SO}_x$ ,  $\text{NO}_x$ , and  $\text{NH}_3$  emissions and surface  $\text{PM}_{2.5}$  concentrations. For Europe, *Erismann and Schaap* [2004] concluded that secondary particulate matter (PM) comprises 50% or more of  $\text{PM}_{2.5}$ . Further, they concluded that since the 1980s,  $\text{SO}_2$  emissions have decreased by more than 60%, while particle concentrations have decreased less. In contrast,  $\text{NO}_x$  and  $\text{NH}_3$  emissions have decreased only slightly. *Erismann and Schaap* [2004] showed that secondary PM concentrations can only be effectively reduced if ammonia emissions are decreased comparably to those of  $\text{SO}_2$  and  $\text{NO}_x$ . For the eastern U.S., *Pinder et al.* [2007] performed scenarios of  $\text{NH}_3$ ,  $\text{SO}_2$ , and  $\text{NO}_x$  emission reductions, to find the least cost  $\text{SO}_2$  and  $\text{NO}_x$  control scenarios that achieve the same reduction in ambient inorganic  $\text{PM}_{2.5}$  concentration as a decrease in ammonia emissions. Their study found that  $\text{NH}_3$  emission reductions yield the largest decrease in  $\text{PM}_{2.5}$  pollution during the winter season, while  $\text{SO}_2$  reductions are more effective during the summer season. For northeast Asia, *Li et al.* [2014] studied  $\text{PM}_{10}$  pollution, segregated by country.

While prior studies have relied on regional models, here we use a global climate model that resolves aerosol microphysical as well as gas/aerosol partitioning, to investigate the effect of agricultural emission on surface air pollution under present-day and future conditions, as well as test reduction scenarios for Nr-containing emissions. The climate model allows us to study this problem globally, as well as investigate possible future atmospheric compositions. Future emission scenarios [*van Vuuren et al.*, 2011] anticipate globally increased agricultural emissions, while combustion sources are expected to regionally decrease. Thus, we test the impact of the agricultural sector upon surface air pollution (manifest in the  $\text{PM}_{2.5}$  concentration) for the present and future atmosphere. This understanding is important because effective regulatory measures to control  $\text{PM}_{2.5}$  pollution require a comprehensive understanding of its chemical composition, attribution to human activities, and segregation into natural and anthropogenic sources.

## 2. Method

This study is based upon the NASA Goddard Institute for Space Studies (GISS) Earth System ModelE2 [*Schmidt et al.*, 2014], with  $2^\circ$  latitude by  $2.5^\circ$  longitude resolution and 40 vertical layers. GISS ModelE2 is coupled to the aerosol microphysical scheme, MATRIX (Multiconfiguration Aerosol TRacker of mIXing state) [*Bauer et al.*, 2008], which in this study uses the aerosol thermodynamics model ISORROPIA II [*Fountoukis and Nenes*, 2007] for aerosol gas phase partitioning. MATRIX simulates aerosol microphysical processes such as particle formation, condensation, and coagulation, leading to evolving size distributions of internally mixed aerosol species. Results are discussed for 10 year mean conditions representing preindustrial (1850), present-day (2010), and future (2100) conditions following the representative emission pathway RCP8.5 scenario. Sea salt and dust emission are calculated interactively, and additional natural and anthropogenic emission fluxes are prescribed from the CMIP5 (Coupled Model Intercomparison Project Phase 5) inventory [*Lamarque et al.*, 2010; *van Vuuren et al.*, 2011]. The models initialization and boundary conditions, sea ice and sea surface temperatures, are taken from coupled transient climate simulations of the same model. Thus, the model simulations take into account emission as well as climate change information. In addition to the base run (E1) two sets of model runs were calculated: experiment (E2) where anthropogenic emissions are set to zero, and experiment (E3) where only agricultural emissions are set to zero. Those runs were used to calculate the natural (NAT, E2), the agricultural (AGR, E1 minus E3), and the anthropogenic without agriculture (ANT, E3 minus E2) effects, assuming a linear response from aerosol and aerosol precursor emissions to PM concentrations. In terms of reduced emitted mass by specie in Tg/yr, with the percent change reflecting the emission reduction compared to the total natural and anthropogenic sources, in (E2) this leads to a reduction of  $\text{SO}_2$  (102 (54%)), BC (5.4 (69%)), OC (13.2 (22%)),  $\text{NH}_3$  (38.8 (80%)), and  $\text{NO}_x$  (32.2 (65%)), and for (E3)  $\text{SO}_2$  (0.2 (0.1%)), BC (0.15 (2%)), OC (0.72 (1%)),  $\text{NH}_3$  (38 (78%)), and  $\text{NO}_x$  (0.19 (1%)) on top of the CO and nonmethane volatile organic compound emission changes.

Additional sensitivity experiments were carried out where emissions of anthropogenic  $\text{NO}_x$  and agricultural  $\text{NH}_3$  are reduced by 20, 40, and 60% separately and combined. These resulted in total emission of  $\text{NO}_x$  (compared to a base of 49.3 Tg/yr) corresponding to 42.3 (20%), 36.9 (40%), and 30.7 Tg/yr (60%) and  $\text{NH}_3$  (58.9 (base), 51.4 (20%), 44.0 (40%), and 36.6 Tg/yr (60%)) in the sensitivity runs. Note that these are the total emission fluxes, and reductions have been uniformly applied to the individual sectors.

Our definition of "agricultural air pollution" is based on the atmospheric composition change calculated when all agricultural emission sources are turned on and off (AGR, as defined above). Secondary changes

to the gas and aerosol phase chemistry that occur when agricultural sources are excluded obviously affect the other gases and aerosols in the atmosphere that are available, most importantly in the context of ammonium nitrate formation and nitrogen oxides. Thus, like any other sector study, we are looking at the “anthropogenically affected agricultural air pollution,” which is the net atmospheric response to the agricultural emissions sector. We prefer this definition over testing emission changed compared to preindustrial concentrations because it is more realistic, since it includes all interactions and feedbacks in the present atmosphere.

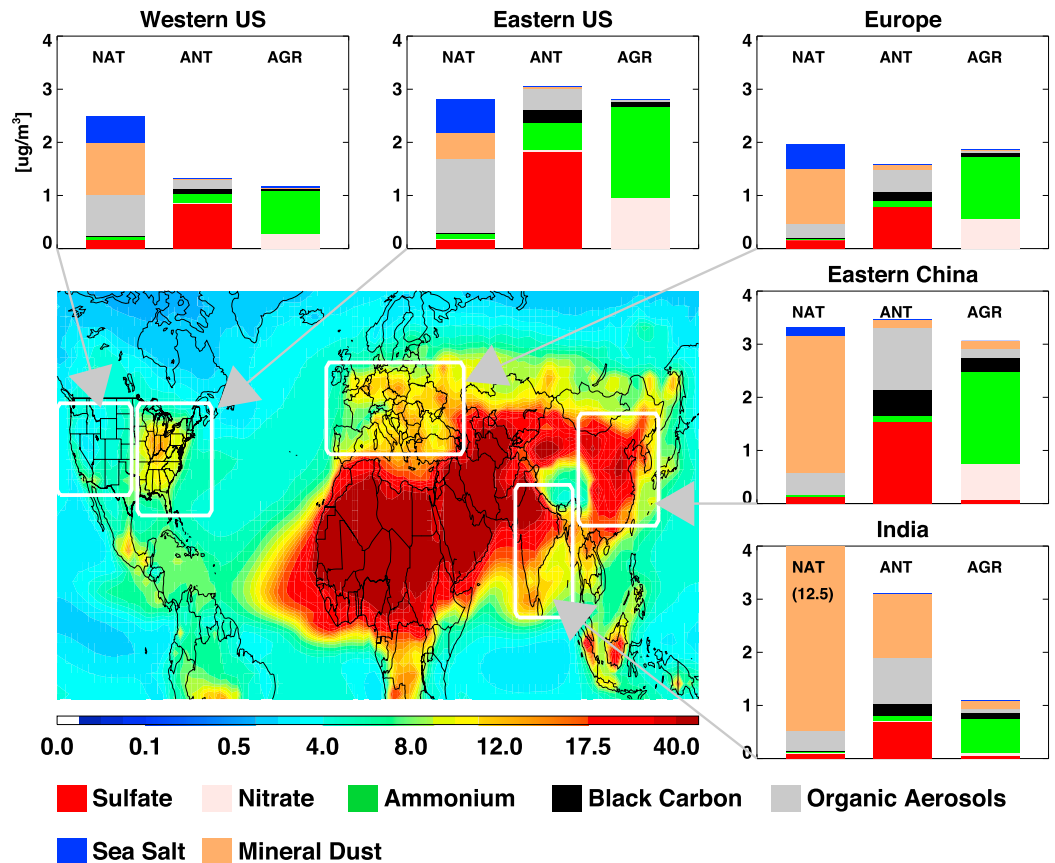
Model evaluation, including the configuration of MATRIX and ISORROPIA used here, against surface and aircraft data shows that sulfate and nitrate aerosols are equally well simulated, with high correlation coefficients (RMS around 0.9) but with the tendency to underestimate total mass concentrations [Mezuman *et al.*, 2016]. Hence, we expect that the impacts upon  $PM_{2.5}$  concentration presented here are biased low.

### 3. Results

For present-day conditions (Figure 1),  $PM_{2.5}$  is highest over Northern Africa, the Middle East, and large parts of Asia, where natural aerosols such as desert dust, sea spray, or aerosols formed from natural biomass burning, soil, and biosphere emissions are the largest source of  $PM_{2.5}$ . Anthropogenic pollution, resulting from activities such as energy and industrial production, manufacturing, transportation, and agriculture, is collocated with densely populated areas, underlining its relevance as a public health hazard. In order to get a better understanding of the impact of agricultural emissions, we grouped  $PM_{2.5}$  into natural (NAT), anthropogenic without agriculture (ANT), and agricultural categories (AGR), depending on their sources (see section 2). The contribution from each category in five highly populated areas is indicated in Figure 1.

$PM_{2.5}$  in the United States is dominated by anthropogenic sources (E2: ANT + AGR) that contribute 60% to the nationwide total. The more polluted eastern U.S. has regions with annual mean  $PM_{2.5}$  concentrations above  $14 \mu\text{g}/\text{m}^3$ , clearly exceeding the WHO and U.S. Environmental Protection Agency health standards. Note that seasonally and locally,  $PM_{2.5}$  can reach much higher levels. Agriculture is responsible for about half of the total anthropogenic pollution in both the eastern and western United States (Figure 1). Ammonium nitrate concentrations in the agricultural experiment originate solely from the use of nitrogen fertilizers and animal husbandry. This means that food production (without accounting for food transportation and processing) is responsible for about the same amount of anthropogenic  $PM_{2.5}$  as produced by all other human activities combined, such as motor vehicles, power plants, and other domestic or industrial processes. This strong impact of livestock and fertilizer on air quality is even more pronounced in Europe, where agricultural  $PM_{2.5}$  is responsible for 55% of air pollution linked to human activities. China shows the highest concentrations of agriculture-related air pollution and also the highest fraction of carbonaceous components:  $PM_{2.5}$  levels exceed  $3 \mu\text{g}/\text{m}^3$  (Figure 1) on a national level and  $20 \mu\text{g}/\text{m}^3$  on a regional basis. On the other hand, agricultural production in India is only responsible for 25% of the total anthropogenic pollution, and 80% of all  $PM_{2.5}$  is attributed to natural sources.

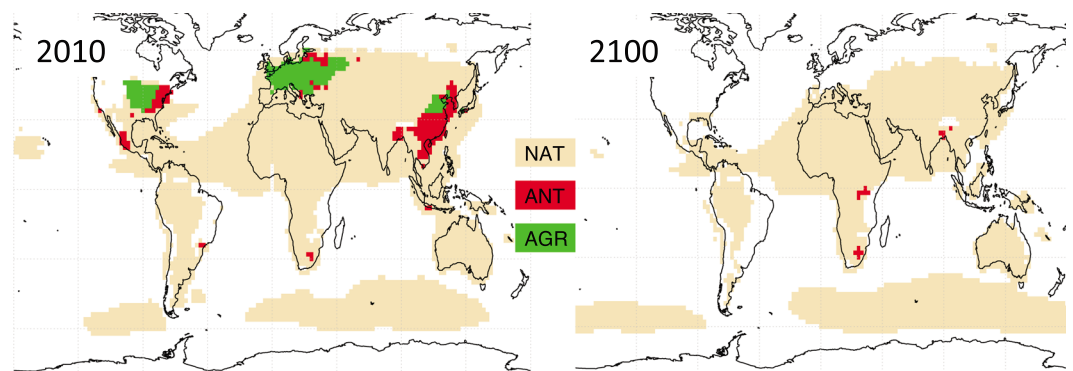
Figure 2 illustrates the category making the largest contribution to  $PM_{2.5}$ . At present, agricultural pollution dominates over wide areas in Europe, the central U.S., and in regions west of Beijing. We would expect this situation to worsen under future scenarios, due to the large anticipated increase in ammonia emissions (Figure 3, left). In this paper we are using the RCP8.5 scenario [van Vuuren *et al.*, 2011], but independent of which future RCP scenario is chosen, in all cases,  $\text{NH}_3$  emission increases in comparison to present-day conditions. This is rooted in the expected growing world population with increased food demands. Taking into account regional variations of population growth, fertilizer application, and increased efficiency in food production leads to an estimated doubling of ammonia emissions by the end of the century under the highest RCP scenario (Figure 3). The largest emission growth rates are expected in Africa, and the slowest growth rates are predicted for Europe. Nonetheless, our results show that  $PM_{2.5}$  attributed to anthropogenic and agricultural sources is widely reduced by the end of the century (Figure 2, right). The explanation for this behavior lies in the regional change of N emission (Figure 3, middle and right). While global ammonia emissions are expected to increase, at the same time global  $\text{NO}_x$  emissions are expected to decrease in the eastern U.S., Europe, and Eastern China, regions that are highly polluted at present. The assumption of reduced  $\text{NO}_x$  surface concentrations leads to reduced nitrate aerosol formation and thus  $PM_{2.5}$  near the



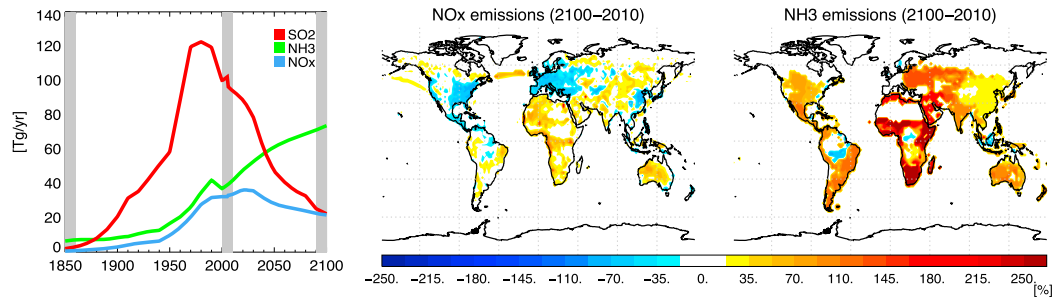
**Figure 1.** Annual mean total  $PM_{2.5}$  concentrations in  $\mu g/m^3$ . The individual bar charts show the chemical composition (colors), for the various sources (NAT = natural, ANT = anthropogenic without agricultural, AGR = agricultural sources) over the five populous regions marked in the map.

Earth surface, despite the global doubling of agricultural ammonia emission. Thus, under the RCP8.5 scenario, surface  $PM_{2.5}$  attributed to anthropogenic and agricultural sources is almost eliminated except for some industrial regions in Africa and India.

The future reduction of surface concentration is opposite to what happens in the free troposphere. Tropospheric nitrate aerosol loads increase in the RCP scenarios, as seen in our simulations and others [Bauer et al., 2007; Bellouin et al., 2013; Hauglustaine et al., 2014; Paulot et al., 2015]. In experiment E1 with



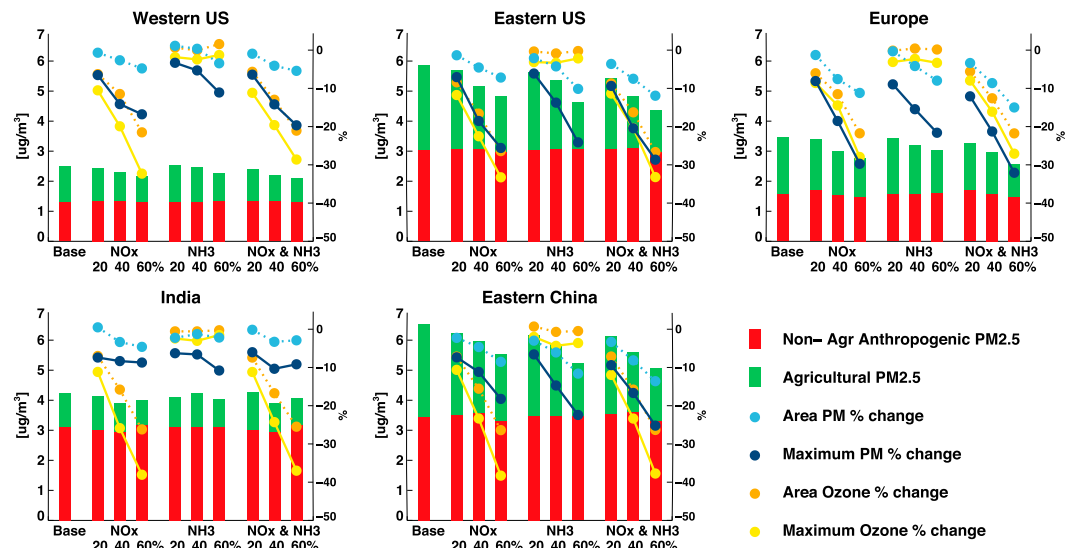
**Figure 2.** Dominant contributor to  $PM_{2.5}$  concentrations with respect to natural, anthropogenic (without agriculture), and agricultural sources. (left) Present-day conditions and (right) 2100 conditions for the RCP8.5 scenario. Areas with  $PM_{2.5}$  concentrations below  $3 \mu g/m^3$  are white.



**Figure 3.** (left) Time evolution of historical and future global mean SO<sub>2</sub>, NO<sub>x</sub>, and NH<sub>3</sub> emissions according to the RCP8.5 scenario (Tg/yr). Percentage differences in emission fluxes (%) between 2010 and 2100 for (middle) NO<sub>x</sub> and (right) NH<sub>3</sub>. Percentage change is masked for fluxes lower than 1 ng/s/m<sup>2</sup>.

all sources, the global nitrate aerosol load increases by 23% from 187 Gg under present day to 230 Gg by the end of this century. Future nitrate formation will increasingly rely on nitric acid, whose concentration is high throughout the atmosphere, due to its high stratospheric concentration, which supplies the troposphere via downward mixing.

We have interpreted the reduction of future PM<sub>2.5</sub> pollution, despite rising food production, as a result of reduced NO<sub>x</sub> emissions. We test this interpretation by considering present-day experiments where emissions of agricultural NH<sub>3</sub> and anthropogenic NO<sub>x</sub> are reduced individually (by 20, 40, and 60%), as well as simultaneously. The results of the nine sensitivity experiments (Figure 4) show that reducing NH<sub>3</sub> and NO<sub>x</sub> together lead to the largest reductions in PM<sub>2.5</sub> but only by a small margin compared to the individual reduction scenarios. Significant reductions in PM<sub>2.5</sub> can result from reducing either agricultural NH<sub>3</sub> or combustion NO<sub>x</sub> separately. In Europe, PM<sub>2.5</sub> pollution is more sensitive to NO<sub>x</sub> emission than to agricultural NH<sub>3</sub>. In the western U.S., mean surface PM<sub>2.5</sub> concentration is more sensitive to anthropogenic NO<sub>x</sub> emissions when the reduction is between 20% and 40%; for reduction from 40% to 60%, mean surface PM<sub>2.5</sub> concentration is more sensitive to agricultural NH<sub>3</sub> reduction. For the eastern U.S. and India, sensitivities to NH<sub>3</sub> and NO<sub>x</sub> are similar, whereas Eastern China shows a stronger sensitivity toward NH<sub>3</sub> reductions. In terms of total peak air pollution reduction, reducing emissions by 20, 40, and 60 % leads roughly to PM<sub>2.5</sub> reductions between 5 and



**Figure 4.** Regional means (same areas as in Figure 1) of PM<sub>2.5</sub> from the agricultural sector (green), and nonagricultural anthropogenic sector (red), for the base run and the experiments with reductions in anthropogenic NO<sub>x</sub> emissions, agricultural NH<sub>3</sub> emissions, and a combination of both by 20%, 40%, and 60%, respectively. Percentage changes for regional means (dashed lines) and maximum reductions per region (solid lines) are shown for PM<sub>2.5</sub> (blues) and surface ozone (yellow, orange) concentrations.

30%. Note that (nonagricultural) anthropogenic pollution (marked in red in Figure 4) is barely affected by these reductions, as here the drivers are  $SO_x$  and organic emissions. This study does not focus on ozone, but as the climate model also includes ozone chemistry, we briefly mention that surface ozone concentration would strongly be reduced by  $NO_x$  reductions (10–30 % in the US and Europe and up to 40% in China and India), but respond very weakly to  $NH_3$  emission changes (Figure 4). In terms of percentage change,  $NO_x$  emissions reductions would be even more effective at reducing ozone pollution than in reducing  $PM_{2.5}$ .

#### 4. Discussion

Our results point to three major regions where agriculture is a significant source of  $PM_{2.5}$ : Europe, the eastern U.S., and China. Nitrate aerosol precursors ( $NO_x$ ) are the link between agricultural emission and surface air pollution. The present-day sensitivity studies (Figure 4) show that a reduction in either  $NO_x$  or agricultural  $NH_3$  leads to reduced  $PM_{2.5}$  pollution. We have identified a stronger sensitivity of  $PM_{2.5}$  toward  $NO_x$  reductions in Europe, equal sensitivities toward  $NO_x$  and  $NH_3$  in the eastern U.S., and a stronger sensitivity toward  $NH_3$  in China. This is a welcome result, demonstrating that increased food production need not exacerbate air pollution if combustion sources of  $NO_x$  can be controlled. Under a future scenario, with doubled emission of agricultural ammonia but lower combustion emission, surface nitrate aerosol concentration will decrease to a level where “agricultural”  $PM_{2.5}$  would no longer be the dominating contribution. Globally, almost all near-surface  $PM_{2.5}$  in populated areas will originate from natural sources, such as desert dust and biomass burning, except for some industrial areas in Africa and northern India.

Besides air quality, increasing fertilization and meat production also threaten ecosystems via Nr deposition. We calculate that the globally averaged Nr deposition has increased from 49 TgN/yr during preindustrial times (1850) to 141 TgN/yr by 2010. This increase is largely caused by cereal production that increased from 1897 to 2270 Mt (20%) and meat production that increased from 207 to 260 Mt (26%). These rising agricultural demands were sustained by Nr creation by the Haber-Bosch process from 100 Tg Nyr<sup>-1</sup> to 121 Tg Nyr<sup>-1</sup> (20%) [Galloway *et al.*, 2008].

Reduced emission of agricultural ammonia would lead to further beneficial reductions of soil and water acidification [Vitousek *et al.*, 1997], aerosol radiative forcing [Bauer *et al.*, 2007; Hauglustaine *et al.*, 2014], and, very importantly, greenhouse gas (GHG) concentrations: nitrous oxide ( $N_2O$ ) is an important anthropogenic greenhouse gas, and agriculture represents its largest source [Reay *et al.*, 2012]. According to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) [IPCC AR5, 2013], agricultural practices emit at least one quarter of global anthropogenic GHG, and if food handling and processing activities are counted, the total share of emissions from the agriculture and food sector is at least one third of the total. In conclusion, significant atmospheric aerosol pollution caused by world food cultivation is at present a concern for health and climate change, whereas in the future the GHG contributions will be the main concern. Considering the high contribution of agriculture to anthropogenic GHG emissions, the choice of food production practices can be a problem or part of a solution in addressing climate change.

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