Dear Dr. Qiang Zhang,

here we have listed the changes applied to the manuscript, following the referees' suggestions. To facilitate the comparison with the ACPD published version, the text modifications are highlighted in the manuscript appended to this letter (see last pages). In this letter, we summarized the changes keeping the same order of the reply, although the page/line number do not correspond to the revised version.

Following the revision of the manuscript, we noticed an error in the naming and description of the simulation, as the 66% reduction simulation was actually wrong. Instead a reduction of 75% was applied in the model, and this has been corrected through the entire manuscript. This does not affect the conclusions.

Correction made following comments of referee # 1

1. **Presenting the global perspective is interesting but I personally doubt that such work has any implications on regional policy as it entirely misses discussion of regionally specific aspect of mitigation opportunities analyzing rather unrealistic scenarios of agricultural emissions; additionally referring to 2010 levels while emissions from livestock and arable land production (fertilizer use) are likely to increase further in several regions, especially in Asia.**
   We extended end of Sect.2 to include the feasibility of the ammonia reduction by agricultural emissions.
2. **Another aspect of this work that needs more clarity is the issue of temporal distribution of agricultural emissions used in the simulations.**
   We extended the discussion, moving the related text in Sect.2, where the model is evaluated, explaining the issue in USA emissions.
3. **another issue is the spatial resolution of the modelled PM2.5 concentrations and its use for calculation of population exposure.**
   We added the discussion on model resolution in Sect.3.3

Other comments:

ABSTRACT: I am not sure if the last sentence about the impact of 100% reduction is of any significance; such reductions are not even theoretically possible.
Following the discussion we decided to keep simulation REF_100 as important from the scientific point of view, although not realistically achievable.

Page 1, Line 23: One could add there a reference to the EU policy which includes now targets for NH_3 emissions within the revised air quality legislation. The authors include a reference to that later in the paper.
The text has been expanded.

Page 2, Line 4: not clear what is meant by manure processing , suggest replacing with manure storage and on field application
We reformulated the sentence.

Page 2, line 4: suggest add N or nitrogen before fertilizer
The change has been implemented.

Page 2, line 12: maybe leads should be replaced with would lead or could lead as this is a modelling study rather than impact observed anywhere.
The text has been changed.

Page 2, line 18: by agriculture should be replaced with from agriculture and resulted can be possibly modified to would or could result
Changed following referee's suggestion.

Page 2, last paragraph: As before, suggest adding a reference to the recent European air quality policy and possibly underlying analysis.
Additional text and reference to European air quality policy has been added here.

Page 3, from line 21: The emissions are for the year 2010 but the references are for data sets until 2005. Few words of explanation?
The text has been extended clarifying that the original references were for the year 2005.

Page 4, Figure 2: A bit small, hard to read the axis
The axis label were increased.

Page 5, last paragraph: I believe it would be beneficial to put these assumptions in perspective of what has been discussed as feasible since the reductions given here, even the lowest level, are in most regions perceived as either infeasible or close to maximum reduction potential unless dietary changes are considered reducing meat demand.
We have extended the text at the end of the section.

Page 6; first paragraph: Presumably the first sentence refers to agricultural burning and so it could be moved to the end of this paragraph where combustion emissions are mentioned.

In general this paragraph should be clear as to which sources are meant next to specific pollutants.
The text has been modified.

Page 6; line 17: The 20-90% reduction refers to single measures and not to the overall mitigation potential and so nowhere 90% can be achieved for the whole agriculture. The potential is typically between 20-45% with some exceptions where structure is different, [..]
The text has been modified accordingly to the main concerned raised by the referee.

Page 8, para from line 15: Total emissions in winter are not higher than in summer! NH_3 emissions are increasing with temperature and also organic fertilizers are applied in Spring, Summer, Autumn, just as the mineral fertilizers.
We removed the wrong text and moved the discussion on seasonality on Sect.2 (where the model evaluation is discussed).

Figure 7: here the resolution for the mortality attributable to $PM_{2.5}$ is indicated as 10x10km. An explanation what data are used to develop that is needed. In general some discussion related to how coarse resolution concentration fields are used in health impact assessment would be useful [...]
The text has been extended, mentioning the issue related to the model coarse resolution.

Correction made following comments of referee # 2

1. **Please state if the ammonia reductions in this study are feasible, especially for different regions**.
   The text has been extended at the end of Sect.2, following also referee's #1 comments.
2. **Section 3.2, the aerosol pH would be determined by aerosol water, which also depends on the secondary nitrate and sulfate concentrations, relative humidity etc. Further, rich or poor ammonia in different regions should have markedly different effects on aerosol pH. Please have some discussions on them.**
   We believe this point to be already addressed in the manuscript, as all Sect.3.2 is dedicated to this argument. Therefore, no text was added.
3. **The epidemiological studies did find the secondary inorganic aerosols could have negligible effects on human health.**
   The text was augmented by adding the new references. Nevertheless we would like to point out that both references suggest a strong health

effect by sulfate (i.e. secondary inorganic aerosols) and therefore not in agreement with the point raised by the referee. We decided to leave the text unchanged, so to be coherent with the citations.

Best regards,
Andrea Pozzer (on behalf of all co-authors)

Impact of agricultural emission reductions on fine particulate matter and public health.

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Abstract. A global chemistry-climate model has been used to study the impacts of pollutants released by agriculture on fine particulate matter ($PM_{2.5}$), with a focus on Europe, North America~~and East~~, East and South Asia. Simulations reveal that a relatively strong reduction in $PM_{2.5}$ levels can be achieved by decreasing agricultural emissions, notably of ammonia (NH_3), released from fertilizer use and animal husbandry. The absolute impact on $PM_{2.5}$ reduction is strongest in East Asia, even for small emission decreases. Conversely, over Europe and North America, aerosol formation is not ~~directly~~ immediately limited by the availability of ammonia. Nevertheless, reduction of NH_3 can also substantially decrease $PM_{2.5}$ concentrations over the latter regions, especially when emissions are abated systematically. Our results document how reduction of agricultural emissions decreases aerosol pH due to the depletion of aerosol ammonium, which affects particle liquid phase and heterogeneous chemistry. Further, it is shown that a 50% reduction of agricultural emissions could prevent the mortality attributable to air pollution by ~ 250 thousands people per year worldwide, amounting to reductions of 30%, 19%~~and~~, 8% and 3% over North America, Europe~~and East~~, East and South Asia, respectively. A theoretical 100% reduction could even reduce the number of deaths globally by about 800 thousand per year.

1 Introduction

Atmospheric aerosol particles are a major constituent of ambient air and have a large impact on atmospheric chemistry, clouds, radiative transfer and climate, and also induce adverse human health effects that contribute to mortality (Stocker et al., 2013; Lelieveld et al., 2015). Particulate matter (PM) with an aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) contributes to air pollution through intricate interactions between emissions of primary particles and gaseous precursors, photochemical transformation pathways, and meteorological processes that control transport and deposition.

As shown by Lelieveld et al. (2015) and Bauer et al. (2016), agricultural emissions play a leading role in the formation of $PM_{2.5}$ in various regions of the world, for example in Central and Eastern Europe. Agricultural emissions are mostly related to animal husbandry and fertilizer use, and to a lesser extent also to the burning of crop residues (Aneja et al., 2008); around 10% of worldwide biomass burning emissions can be ascribed to agricultural activities (Doering et al., 2009b). The general importance of agricultural emissions for air quality was also previously identified by ~~Zhang et al. (2008) , Tsimpidi et al. (2007) and~~

~~Megaritis et al. (2013)~~ a number of studies (e.g. Zhang et al., 2008; Tsimpidi et al., 2007; Megaritis et al., 2013) , and recognized through environmental policies, (e.g., the establishment of ceilings for national emissions for ammonia by the European Union Clean Air Program).

The dominant trace gas emitted by agricultural activities is ammonia (NH_3). Around 80-90% of the atmospheric NH_3 emissions in industrialized regions are from the agricultural sector (Sotiropoulou et al., 2004; Lamarque et al., 2011; van Vuuren et al., 2011b, a). ~~Ammonia~~ NH_3 is formed and released during the decomposition of manure and organic matter, mostly from animal farming and the associated manure ~~processing~~storage and field application, with an additional contribution from (synthetic) nitrogen fertilizer use. ~~Ammonia~~ NH_3 is a toxic gas at very high concentrations, with a pungent smell that irritates the eyes and respiratory system. NH_3 is also a major alkaline gas in the atmosphere and plays an important role in neutralizing acids in the aerosol and cloud liquid phase, forming ammonium sulfate and ammonium nitrate (ammonium salts) (Behera et al., 2013). Therefore NH_3 contributes to secondary aerosol formation and the overall particulate matter burden, and decreases the acidity of the aerosols, which in turn increases the solubility of weak acids (e.g., $HCOOH$, SO_2). The aerosol pH plays an important role in the reactive uptake and release of gases, which can affect ozone chemistry, particle properties such as hygroscopic growth and scattering efficiency of sunlight, and deposition processes (Zhang et al., 2007; Thornton et al., 2010; Pathak et al., 2011).

Tsimpidi et al. (2007) showed that a 50% reduction of NH_3 emissions ~~leads~~ would lead to a 4% and 9% decrease in $PM_{2.5}$ over the Eastern USA in July and January, respectively. The reduction of NH_3 emissions was found to be the most effective $PM_{2.5}$ control measure for the winter period over the Eastern USA, compared to similar reductions of SO_2, NO_x and VOC emissions (Pinder et al., 2008; Tsimpidi et al., 2007, 2008; Karydis et al., 2011). Megaritis et al. (2013) and Bessagnet et al. (2014) found that over Europe the reduction of NH_3 emissions is the most effective control strategy to mitigate $PM_{2.5}$, during both summer and winter, mainly due to a significant decrease of ammonium nitrate. Further, De Meij et al. (2009), showed that reducing the NH_3 emissions ~~by~~ from agriculture by 50% ~~resulted~~ could result in a decrease of $PM_{2.5}$ concentrations up to 2.4 $\mu g/m^3$ over the Po Valley region (Italy). This confirms the finding of de Meij et al. (2006), who showed that for short-lived species like NO_x and NH_3, short-term fluctuations of the emissions play an important role in the formation of nitrate aerosol. According to Wang et al. (2011), NH_3 emissions contribute 8-11% to $PM_{2.5}$ concentrations in East China, which is comparable with the contributions of SO_2 (9-11%) and NO_x (5-11%) emissions. However, the air quality benefits of controlling NH_3 emissions could be offset by the potential enhancement of aerosol acidity. Weber et al. (2016) showed that despite the large investments in sulfur dioxide emission reductions, the acid/base gas particle system in the southeastern USA is buffered by the partitioning of semivolatile NH_3, making the pH insensitive to SO_2 controls. Several studies have been performed on the impact of NH_3 on aerosol nitrate (Pye et al., 2009; Heald et al., 2012; Schaap et al., 2004; Pinder et al., 2007; Holt et al., 2015), and sulfate (Redington et al., 2009; Paulot et al., 2016; Wang et al., 2011), mostly with a regional rather than a global view.

As $PM_{2.5}$ has been clearly associated with many health impacts, including acute lower respiratory infections (ALRI), cerebrovascular disease (CEV), ischaemic heart disease (IHD), chronic obstructive pulmonary disease (COPD) and lung cancer (LC) (Burnett et al., 2014). Due to its strong contribution to the $PM_{2.5}$ mass, control strategies in NH_3 emissions could possi-

bly reduce the mortality attributable to air pollution. Such analysis has , and air quality policy in Europe does indeed include ceilings for NH_3 emissions (Kuklinska et al., 2015) . Studies on $PM_{2.5}$ reduction due to NH_3 control have been performed regionally both for Europe (Brandt et al., 2013) and the U.S.A. (Paulot and Jacob, 2014; Muller and Mendelsohn, 2007). Further, while, a detailed analysis on the global scale was performed by Lee et al. (2015), who showed the importance of ammonia as a contributor to mortality attributable to air pollution. Nevertheless, Lee et al. (2015) assumed an ammonia reduction of 10%, and the health effects were linearized around the present-day concentrations. As the exposure-response functions, linking $PM_{2.5}$ to mortality attributable to air pollution, are highly non-linear at relatively low concentrations, the mortality reduction estimation could change drastically for strong reductions of ammonia emissions. Therefore, in this work, more aggressive reductions are studied (see Sect.2).

Furthermore, there is a need to investigate the impact of NH_3 emission reductions not only on $PM_{2.5}$ concentrations, but also account for particle acidity and aerosol composition. The goal of this work is to understand the impact of global agricultural emissions on model simulated $PM_{2.5}$ concentrations, the effects on aerosol pH and the potential consequences for human health, with a focus on four continental regions where air quality limits and guidelines for $PM_{2.5}$ are often exceeded, i.e., North America, Europe, South and East Asia. North America is defined as the region that encompasses the U.S.A and Canada, Europe is represented by the European continent (including Turkey) excluding Russia, South Asia includes India, Sri Lanka, Pakistan, Bangladesh, Nepal and Buthan, while the East Asia region includes China, North and South Korea, and Japan (see Fig.1).

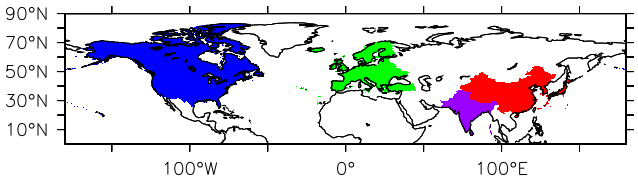

Figure 1. Regions addressed in this study, i.e., North America (blue), Europe (green), South Asia (yellowpurple) and East Asia (red).

This work may also support policy actions aimed at controlling ammonia emissions, e.g., formulated in the European Union Clean Air Program (http://ec.europa.eu/environment/air/pollutants/ceilings.htm) which sets ceilings for national emissions for sulfur dioxide, nitrogen oxides, volatile organic compounds, fine particulate matter, and also for ammonia.

2 Methodology

In this study the EMAC (ECHAM5/MESSy Atmospheric Chemistry) model version 1.9 was used. EMAC is a combination of the general circulation model ECHAM5 (Roeckner et al., 2006, version 5.3.01) and the Modular Earth Submodel System (Jöckel et al., 2005, MESSy, version 1.9), Extensive evaluation of the model can be found in Jöckel et al. (2006); Pozzer et al. (2007, 2012a); Pringle et al. (2010a); de Meij et al. (2012a). ECHAM5 has been used at the T106L31 resolution, corresponding to a horizontal resolution of $\sim 1.1 \times 1.1^{\circ}$ of the quadratic Gaussian grid, and with 31 vertical levels up to 10 hPa in the

lower stratosphere. The model set-up is the same as that presented by ~~(Pozzer et al., 2012a, b)~~ Pozzer et al. (2012a, b) and is briefly summarized here. The anthropogenic emissions are for the year 2010 from the EDGAR-CIRCE (Doering et al., 2009a, c, Emission Database for Global Atmospheric Research) database, distributed vertically to account for different injection altitudes (Pozzer et al., 2009). Bulk natural aerosol emissions (i.e., desert dust and sea spray), are treated using offline monthly emissions files based on AEROCOM (Dentener et al., 2006) and hence are independent of the meteorological conditions. The atmospheric chemistry is simulated with the MECCA (Module Efficiently Calculating the Chemistry of the Atmosphere) submodel by Sander et al. (2005, 2011), and the aerosol microphysics and gas-aerosol partitioning are calculated by the Global Modal-aerosol eXtension (GMXe) aerosol module (Pringle et al., 2010a, b). Gas / aersosol partitioning is calculated using the ISORROPIA-II model (Fountoukis and Nenes, 2007; Nenes et al., 1998a, b). Following the approach of Pozzer et al. (2012b), the year 2010 is used as reference year, and the feedback between chemistry and dynamics was switched-off, and therefore all simulations described here are based on the same (binary identical) dynamics and consequent transport of tracers.

~~The model has~~ Although Pozzer et al. (2012a) evaluated the model for the same configuration and emissions database, the emissions were referring to the year 2005 while here the emissions for the year 2010 are used. Therefore the model is re-evaluated for the species of interest (i.e. $SO4^{2-}$, $NO3^{-}$,NH_4^+ and $PM_{2.5}$). The model results of this study have been evaluated against satellite based $PM_{2.5}$ estimates (van Donkelaar et al., 2010); the results are shown in Fig.2 and are summarized in Tab.1, also focusing on the four regions focus of this study (i.e., North America, Europe, South and East Asia). Compared to global satellite derived $PM_{2.5}$ concentrations this model version, with prescribed dust emissions, consistently overestimates $PM_{2.5}$ over desert areas (see Fig.2). However, the average concentration of $PM_{2.5}$ at the surface in the regions of interest is within 30% of the observations. For Europe and South Asia, 95% of the simulated $PM_{2.5}$ concentrations are within a factor of 2 of the observations, while for North America and East Asia this is about 80%.

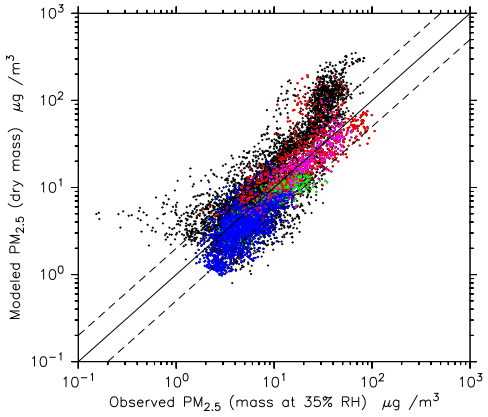

Figure 2. Scatter plot of observed and modeled yearly averaged concentrations of $PM_{2.5}$ (in $\mu g/m^3$). The colors denote the regions, i.e., blue North America, green Europe, purple ~~East~~ South Asia and red East Asia. Black are locations outside these regions.

Further, sulfate-ammonium-nitrate has been compared with station observations from different databases, such as from EPA (United States Environmental Protection Agency), EMEP (European Monitoring and Evaluation Programme) and EANET

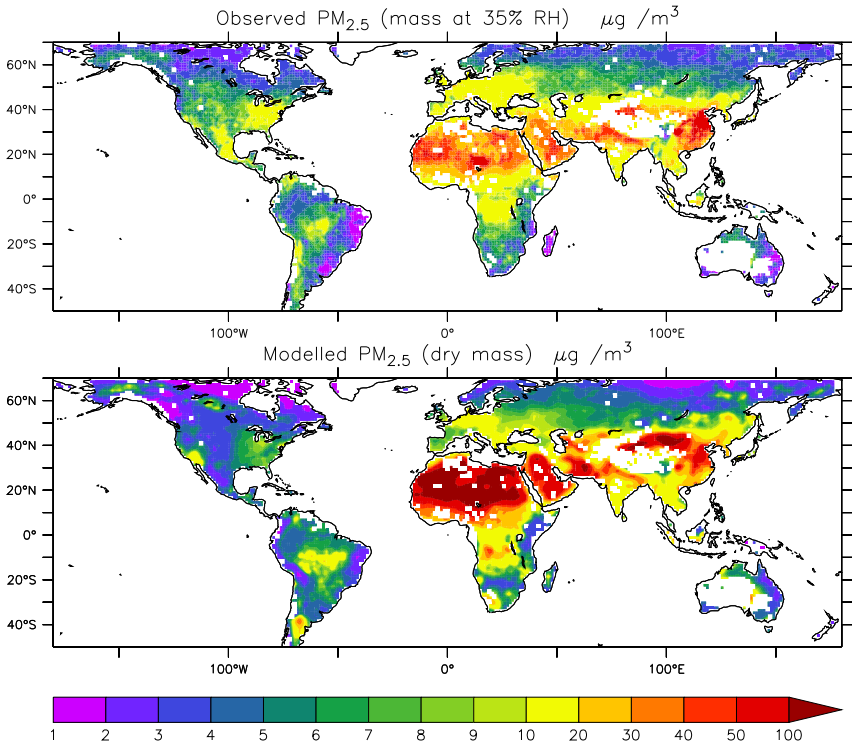

Figure 3. UPPER: observed annual mean $\mathrm{PM}_{2.5}$ from (van Donkelaar et al., 2010), LOWER: simulated annual mean $\mathrm{PM}_{2.5}$ (REF simulation), both in $\mu\,\mathrm{g/m^3}$.

Table 1. Summary of the comparison of model data to pseudo-observations of $\mathrm{PM}_{2.5}$ concentrations (van Donkelaar et al., 2010). OAM and MAM are the spatial arithmetic mean of the observations and of the model results (REF simulation), respectively (in $\mu\,\mathrm{g/m^3}$), based on the annual averages. The model results were masked in locations where no observations are available. PF2 is the percentage of model results within a factor of two of the observations.

region	MAM	OAM	MAM/OAM	PF2
Europe	9.00	11.96	0.75	0.95
North America	4.31	5.89	0.72	0.80
South Asia	24.49	24.95	0.98	0.95
East Asia	33.60	27.56	1.22	0.81
World	22.58	13.02	1.73	0.75

(Acid Deposition Monitoring Network in East Asia) for the year 2010. The results are shown in Fig. 4 and summarized in Tab.2.

While sulfate is well reproduced, with more than $\sim 85\%$ of the model results within a factor of two compared to the observations, nitrate is overestimated in North America and Europe by ~ 50 %, although nitric acid is reproduced accurately

Table 2. Summary of the comparison of model data to observations of aerosol component concentrations. OAM and MAM are the spatial arithmetic mean of the observations and the model, respectively (in $\mu g/m^3$). PF2 is the percentage of model results within a factor of two of the observations.

species	network	MAM	OAM	MAM/OAM	PF2
SO_4^{2-}	EPA	1.22	1.18	1.03	85.5
SO_4^{2-}	EMEP	1.36	1.70	0.79	86.5
SO_4^{2-}	EANET	1.54	3.30	0.46	88.8
NO_3^-	EPA	0.65	0.42	1.54	63.0
NO_3^-	EMEP	2.08	1.15	1.81	32.6
NO_3^-	EANET	1.11	1.37	0.81	68.3
NH_4^+	EPA	0.77	0.79	0.97	88.0
NH_4^+	EMEP	1.11	1.07	1.03	74.6
NH_4^+	EANET	0.77	0.96	0.79	80.6

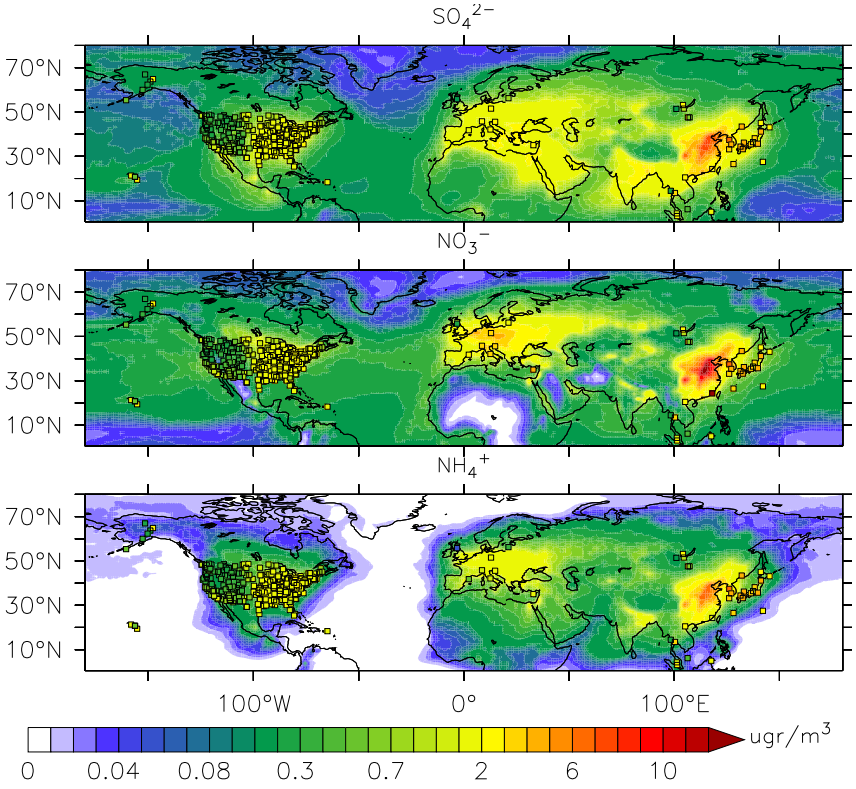

Figure 4. Simulated mean concentrations of $PM_{2.5}$ ~~component~~ components (SO_4^{2-}, NO_3^- and NH_4^+) in $\mu g/m^3$ at the surface for the year 2010, with observations from EPA, EMEP and EANET (averaged over the same period) overplotted.

by the model (based on comparison with observations from Emmons et al. (2000), see Jöckel et al. (2006)). As the nitrate concentrations seems to be on the high end of the observations, it must be acknowledged that the effect of reducing ammonia emissions from agriculture could be overestimated. On the other hand, nitrate predictions are in a good agreement with the measurements over East Asia. Further, ammonium concentrations are well captured by the model, with more than 75% of the modeled results being within a factor of 2 compared to observations. For ammonium, the annual averages estimated from model results compare well with the observations (see Tab. 2). Further, as shown by Pozzer et al. (2012a) , simulated seasonal cycle of ammonium concentrations compares well with the observed one, both for Europe and Asia (with temporal correlations between model results and observations above 0.7 and 0.5, respectively). However, this is not the case on the East Coast of the USA, where the correlation is below 0.2. As suggested by Pozzer et al. (2012a) , this is due to the wrong seasonality of the ammonia emissions, driven by an understimation of the livestock emissions, which have a maximum in summer and should account for 80% of the annual NH_3 emissions in the region (Battye et al., 2003) . The agricultural emissions of ammonia in this region in the model reproduce mostly the fertilizer application as described by Goebes et al. (2003) and therefore the real seasonality of the ammonia emissions is missing (Paulot et al., 2014) . The seasonal results over the USA should hence be taken with caution. Further evaluation can be found in Pozzer et al. (2012a, b) and de Meij et al. (2012b).

In the current analysis four simulations with the EMAC model ~~are used~~ have been performent to study the impacts on $PM_{2.5}$ components: the evaluated reference simulation (REF) and three sensitivity calculations in which the agricultural emissions have been reduced by different percentages, 50% in simulation REF_50, ~~66~~75% in simulation ~~REF_66~~ REF_75 and 100% (i.e., removing all agricultural emissions) in simulation REF_100.

The ~~total primary emitted particle mass from agricultural activities in the REF simulation is 0.4 and 1.9 Black Carbon () and Organic Carbon (), respectively, representing in both cases $\sim$ 5of their total emissions. The~~ NO_x emissions from agriculture are 0.7 $Tg(N)/yr$, i.e., only $\sim$ 1.7% of the total NO_x emissions. Most importantly, 34.3 $Tg(N)/yr$ of NH_3 are emitted by agricultural activities, such as livestock manure and N mineral fertilizers, accounting for $\sim$ 80% of the anthropogenic and $\sim$ 67% of the global total ammonia emissions. ~~Finally,~~

Agricultural waste burning is responsible for the emissions of 0.1 $Tg(S)/yr$ of SO_2 (less than 1% of the total SO_2 emissions) and 23.2 $Tg(C)/yr$ of CO ($\sim$ 5% of the total CO emissions)~~are emitted, by agricultural waste burning~~, as well as 0.4 and 1.9 $Tg(C)/yr$ of Black Carbon (BC) and Organic Carbon (OC), respectively, representing in both cases $\sim$ 5% of their total emissions.

Considering these emission magnitudes, the main effects of agricultural emissions on $PM_{2.5}$ are expected from NH_3 through gas-particle partitioning. Therefore, the ammonia emissions used in this work have been compared to other used databases, such as EDGARv4.3.1 (Emission Database for Global Atmospheric Research Crippa et al., 2016) and RCP85 (Representative Concentration Pathways van Vuuren et al., 2011b, a). These datasets differ globally by $\sim$ 15% (40.26, 47.49 and 40.62 Tg/yr for EDGAR-CIRCE, EDGARv4.3.1 and RCP85, respectively). This reflects the uncertainties in the emission estimates of ammonia which could be up to 50% on a local scale (Beusen et al., 2008). The implementation of bidirectional exchange of ammonia between soil and atmosphere may improve the emissions from livestock, although this approach is still associated

with underestimates of emissions (Zhu et al., 2015). Further, ammonia emitted from traffic is included ($\sim$ 1% of total ammonia emissions) although toward the lower end of what has been estimated by Sun et al. (2016).

As shown by Lorenz and Steffens (1997); Webb et al. (2006); Kai et al. (2008), a sustainable reduction of ammonia emissions between 20% to 90% could be achieved, depending on the emission process and the methodology applied (e.g., slurry acidification, adjustment in slurry application, under-floor drying of broiler manure in buildings, replace urea with ammonium nitrate). As the efficiencies of the abatement processes are not well established (Misselbrook et al., 2002), fixed relative reductions have been applied here to all agricultural emissions. Webb et al. (2006) showed that for the United Kingdom a moderate reduction in ammonia emission is easily affordable, while the costs are likely to increase exponentially for reductions above 25%. The same control measures would become even more difficult to apply in countries where livestock production is projected to largely increase (such as the Asia region (Delgado et al., 2001)), where these should be adopted massively.

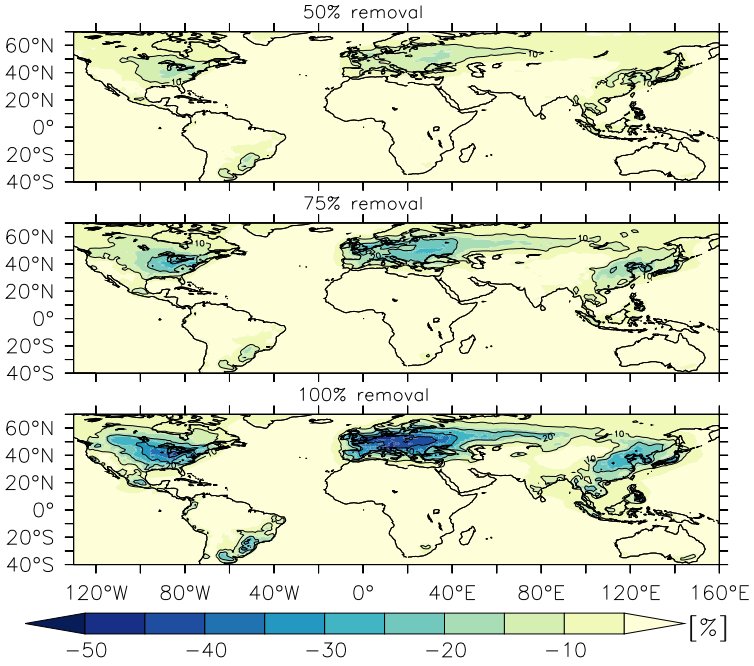

Figure 5. Relative annual average surface ~~PM2.5~~ $PM_{2.5}$ concentration changes (in %) from the three scenarios with agricultural emissions reductions of 50, ~~66~~ 75 and 100% (top, middle and bottom, respectively).

3 Results and discussion

3.1 Impact on $PM_{2.5}$

In Figure 5 the relative annual average surface $PM_{2.5}$ concentration changes between simulations REF_50, ~~REF_66~~REF_75,REF_100 and REF are presented. These simulations reflect the impact on $PM_{2.5}$ of policies imposing an overall decrease in the agricul-

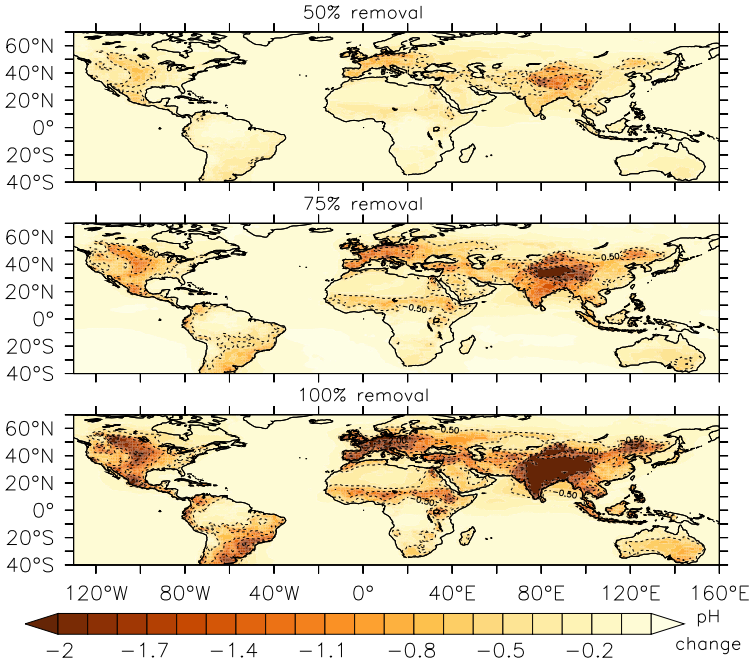

Figure 6. Absolute annual average surface aerosol pH changes (all modes) from three scenarios with agricultural emission reductions of 50, ~~66~~ 75 and 100% (top, middle and bottom, respectively).

tural emissions of 50, ~~66~~ 75 and 100%, respectively. In Tab. 3 the predicted $PM_{2.5}$ concentrations and pH for all simulations are also listed. The largest effects are found over Europe, North America and ~~over~~ China, the latter with smaller relative intensity. A 50%, ~~66~~75% and 100% reduction of ammonia emissions would reduce the annual and geographical mean $PM_{2.5}$ levels over Europe by $\sim$ 1.0 $\mu g/m^3$ (11%), 1.8 $\mu g/m^3$ (19%) and 3.1 $\mu g/m^3$ (34%), respectively, compared to the reference annual surface concentration of 8.9 $\mu g/m^3$. The same relative emission decreases in North America lead to $PM_{2.5}$ concentration reductions of 0.3 $\mu g/m^3$ (8%),0.5 $\mu g/m^3$ (12%) and 0.69 $\mu g/m^3$ (16%), respectively, compared to a reference annual surface concentration of 4.0 $\mu g/m^3$. Over East Asia the absolute decrease in the annual average $PM_{2.5}$ concentration near the surface is 1.6 $\mu g/m^3$ (5%), 2.7 $\mu g/m^3$ (8%) and 4.08 $\mu g/m^3$ (13%), respectively, for the three scenarios. Although the absolute changes in East Asia (relative to a reference value of 31.1 $\mu g/m^3$), are larger than the corresponding changes estimated over Europe and North America, the relative changes are smaller. In fact, the fraction of fine particle mass that is directly ammonia sensitive (i.e., $(NH_4^+ + NO_3^-)/PM_{2.5}$) is relatively smaller in East Asia ($\sim$13%) compared to Europe ($\sim$27%) and North America ($\sim$17%), and reduction of NH_3 emissions would mainly decrease the nitrate and ammonium components, rather than the predominant components of $PM_{2.5}$ in this part of the world. Over South Asia, this effect is even more ~~enhanced~~pronounced. The decreased emissions, in fact, have a negligible impact on annual average $PM_{2.5}$, reducing it by 0.62 (2%), 0.76 (3%) and 1.44 (6%) $\mu g/m^3$, for reductions of ammonia emissions of 50%, ~~66~~75% and 100%, respectively. The fraction of fine particle mass sensitive to ammonia, in this region, is very low (3%), since more than 90% of the aerosol mass is not formed by the

ammonium-sulfate-nitrate components, but rather by organic carbon ($\sim$ 45 % of the total mass) and dust ($\sim$ 35% of the total mass).

In all four regions considered here the impact of NH_3 emissions reduction on $PM_{2.5}$ concentrations is strongest during winter. This is related to the enhanced NH_4NO_3 partitioning in the gas phase due to the higher temperatures during summer, so that a reduction of NH_3 influences the gas phase concentrations more strongly than the particulate phase during this season. The opposite happens during the winter season. Additionally, in the REF simulation, the winter total nitrate (gas and aerosol) concentrations are somewhat higher than during the summer over Europe (5.3 vs 4.5 $\mu g/m^3$), USA (1.5 vs 1.0 $\mu g/m^3$), South Asia (10.0 vs 3.4) and East Asia (8.2 vs 4.5 $\mu g/m^3$). This is related to the lower boundary layer height in winter, and hence less dilution of the emitted tracers~~. In addition, the total emissions in winter are higher than the emissions in summer. As extensively discussed in Pozzer et al. (2012a) , this is due to a dual peak present in the emissions database, which leads to increased emissions in late winter/ beginning of spring and late summer/beginning of autumn, which most probably does not realistically reproduce the real seasonality of the ammonia emissions (Paulot et al., 2014)~~ , although in the Northern Hemisphere the ammonia winter emissions are generally lower than during summer time.

The total $PM_{2.5}$ sulfate (i.e., $SO_4^{2-} + HSO_4^-$) is not directly affected by NH_3 emission reductions since it can exist in the aerosol phase in the form of ammonium sulfate or ammonium bisulfate, depending on the ammonium concentration. However, sulfate formation in the aqueous phase is limited by high acidity. As a consequence, the SO_4^{2-} concentration in $PM_{2.5}$ decreases, annually averaged, by 11%, 23%, and ~~66~~75% over Europe, by 15%, 28% and 57% over North America, by 3%, 7% and 50% over South Asia, and by 18%, 36% and 74% over East Asia for a reduction of 50%, ~~66~~75% and 100% of agricultural emissions, respectively. This is counterbalanced by an increase of HSO_4^- concentrations.

For Europe and North America, the simultaneous decrease of nitrate and ammonium, makes the reduction of agricultural emissions very effective, especially during winter, in accord with the findings of Tsimpidi et al. (2007) and Megaritis et al. (2013). Furthermore, the relationship between ammonia and $PM_{2.5}$ concentrations is not linear, and is governed by the sulfate/nitrate ratio (Tsimpidi et al., 2007). Our EMAC simulations reveal that the $PM_{2.5}$ response to NH_3 emissions is more linear during winter (compared to summer) since the sulfate/nitrate ratio is generally lower.

Following Makar et al. (2009), the particle neutralization ratio (PNR, i.e. $(NH_4^+)/(2(SO_4^{2-} + HSO_4^-) + NO_3^-)$) calculations indicate that in Europe and East Asia (both with PNR equal to 0.20) ammonia concentrations must be decreased relatively more strongly than in North America and South Asia (PNR equal to 0.13 for both regions) to reach the ammonia limited regime, i.e., before $PM_{2.5}$ can be efficiently controlled by decreasing NH_3 emissions. On the other hand, the absolute reduction in $PM_{2.5}$ depends on the fraction of fine particulate mass that is directly ammonia sensitive. As a consequence, Europe has the overall largest potential of reducing annual averaged $PM_{2.5}$ by strongly controlling NH_3 emissions (up to 34%), followed by North America (up to 16%) and East Asia (up to 13%), while South Asia has very limited potential (up to 6%). Thus it follows that, although the emission decrease needed in Europe to reach the ammonia limited regime is larger than in North America, the effective gain of further reduction - once this regime is reached - is considerably larger. In East Asia, where $PM_{2.5}$ is not ammonia limited, even strong emission decreases would reduce the $PM_{2.5}$ mass by up to to 13% on annual average.

3.2 Impact on particle pH

In addition to the significant reductions in $PM_{2.5}$ from ammonia emission controls, which are considered beneficial for human health, we note that the aerosol pH can change substantially. This has the potential of altering the particle liquid phase and heterogeneous chemistry, including reactive uptake coefficients and the outgassing of relatively weak acids, and the pH of cloud droplets that grow on aerosols, which in turn affects aqueous phase sulfate formation. Ammonia is in fact the most abundant and efficient base that controls the aerosol composition over anthropogenically influenced regions, ~~which~~ and neutralizes sulfuric, nitric and other acids.

In the REF simulation, the particles over the focal regions are highly acidic, consisting mainly of ammonium sulfate and ammonium nitrate, as also shown by Weber et al. (2016). Figure 6 illustrates how the aerosol pH can drop due to NH_3 emission decreases. Over Europe, the calculated mean ~~pH decreases of aerosols are~~ aerosol pH decreases by 0.35, 0.62 and 1.05 pH units for the REF_50, ~~REF_66~~ REF_75 and REF_100 simulations, respectively. The calculations indicate similar decreases over East Asia (0.35, 0.62 and 1.11 pH units, respectively), smaller decreases over North America (0.16 ,0.29 and 0.51 pH units, respectively), while the largest decreases are present over South Asia (0.56, 0.99 and 1.72, pH units, respectively). Over South Asia, the impact of ammonia emissions reduction on pH is the largest (see Fig. 6) despite the relatively small impact of the same changes on $PM_{2.5}$. This is due to the high sulfate concentrations, which are neutralized in decreasing order by the presence of ammonium in the three sensitivity simulations. The pH of $PM_{2.5}$ is therefore more sensitive to ammonia emissions (and its atmospheric concentrations) than sulfate, as shown by Weber et al. (2016). This increase of acidity for reduced ammonia emissions would have strong influence on halogen activation and aerosol-gas equilibrium of weak acids in the atmosphere.

Contrary to what was found for $PM_{2.5}$, the reduction of pH is larger during summer than during winter. This is due to the lower concentrations of ammonia in the aerosol phase during summer (see Sect. 3.1), i.e., with relatively low neutralization capability in this season. Therefore, any further reduction of ammonia emissions would strongly reduce the neutralization potential, and therefore increase even more drastically the acidity of the particles.

It should be mentioned that in the present calculations the chemical impact of alkaline desert dust is not taken into account, which can contribute significantly to $PM_{2.5}$ over areas downwind of the deserts (Karydis et al., 2016), e.g., over the Indian subcontinent in the dry season and over East China during spring (Wang et al., 2013), so that the pH effect described here is probably an upper limit. This topic is subject of a publication in preparation.

3.3 Impact on public health

From the simulated $PM_{2.5}$ concentrations, the mortality attributable to air pollution has been calculated following the method of Lim et al. (2013) ~~using the~~ and described in detail in Lelieveld et al. (2015) . The exposure-response functions of Burnett et al. (2014) is used, which shows how fine particulate matter is associated with health impacts, through chronic obstructive pulmonary disease (COPD), acute lower respiratory infections (ALRI), cerebrovascular disease (CEV), ischaemic heart disease (IHD), and lung cancer (LC). Here mortality attributable to $PM_{2.5}$ at 50% relative humidity has been estimated, thus not accounting for ozone related mortality through COPD, which is $\sim 5\%$ of the total mortality attributable to air pollution (Lelieveld

Table 3. Average concentration of $PM_{2.5}$ and $PM_{2.5}$ components (in $\mu g/m^3$). SO_4^{2-} represent total sulfate (i.e., SO_4^{2-} and HSO_4^-). pH average values are also listed.

Region	REF simulation					REF_50 simulation					REF_75 simulation					REF_100 simulation				
	NH_4^+	NO_3^-	SO_4^{2-}	$PM_{2.5}$	pH	NH_4^+	NO_3^-	SO_4^{2-}	$PM_{2.5}$	pH	NH_4^+	NO_3^-	SO_4^{2-}	$PM_{2.5}$	pH	NH_4^+	NO_3^-	SO_4^{2-}	$PM_{2.5}$	pH
	All year																			
Europe	0.94	1.80	1.25	8.95	2.04	0.72	1.32	1.20	7.93	1.68	0.53	0.92	1.19	7.22	1.42	0.09	0.27	1.19	5.89	0.98
North America	0.27	0.45	0.56	4.07	1.60	0.20	0.30	0.55	3.73	1.43	0.15	0.21	0.54	3.58	1.31	0.06	0.11	0.54	3.38	1.09
South Asia	0.50	0.39	1.41	23.27	2.87	0.46	0.25	1.41	22.65	2.31	0.42	0.18	1.41	22.51	1.88	0.16	0.12	1.40	21.83	1.15
East Asia	1.56	2.43	2.51	31.12	1.95	1.12	1.47	2.49	29.50	1.59	0.77	0.80	2.49	28.43	1.33	0.14	0.10	2.49	27.04	0.83
World	0.10	0.21	0.32	9.23	1.84	0.08	0.16	0.32	9.05	1.75	0.06	0.13	0.32	8.98	1.68	0.02	0.10	0.32	8.89	1.53
	Summer																			
Europe	0.90	1.02	1.89	7.74	2.26	0.74	0.70	1.88	7.07	1.80	0.60	0.45	1.91	6.72	1.50	0.11	0.10	1.87	5.70	1.04
North America	0.22	0.13	0.68	5.51	1.93	0.18	0.08	0.68	5.32	1.73	0.14	0.06	0.66	5.29	1.59	0.06	0.05	0.67	5.23	1.34
South Asia	0.17	0.19	0.75	16.76	2.96	0.16	0.18	0.74	16.49	2.44	0.14	0.17	0.74	16.44	2.07	0.06	0.16	0.73	16.16	1.40
East Asia	1.21	0.98	3.00	19.33	1.87	0.89	0.50	2.98	18.40	1.57	0.61	0.19	2.94	17.69	1.36	0.09	0.02	2.93	17.04	0.95
World	0.09	0.13	0.36	7.39	1.94	0.07	0.11	0.36	7.25	1.85	0.05	0.09	0.36	7.23	1.78	0.01	0.08	0.36	7.20	1.64
	Winter																			
Europe	1.08	2.48	0.80	11.12	1.90	0.80	1.96	0.75	9.84	1.59	0.55	1.46	0.74	8.86	1.35	0.06	0.47	0.74	6.94	0.90
North America	0.43	1.01	0.48	3.98	1.39	0.29	0.68	0.45	3.36	1.22	0.20	0.47	0.44	2.98	1.10	0.06	0.19	0.43	2.48	0.87
South Asia	0.71	0.57	1.75	29.63	2.95	0.64	0.33	1.75	28.65	2.40	0.58	0.20	1.75	28.48	1.90	0.24	0.11	1.75	27.54	1.03
East Asia	2.07	4.25	2.00	40.16	2.18	1.53	2.96	1.91	37.96	1.73	1.06	1.84	1.90	36.27	1.39	0.18	0.21	1.93	33.61	0.72
World	0.13	0.33	0.30	11.39	1.78	0.10	0.25	0.29	11.14	1.68	0.07	0.19	0.29	11.02	1.60	0.02	0.12	0.29	10.85	1.43

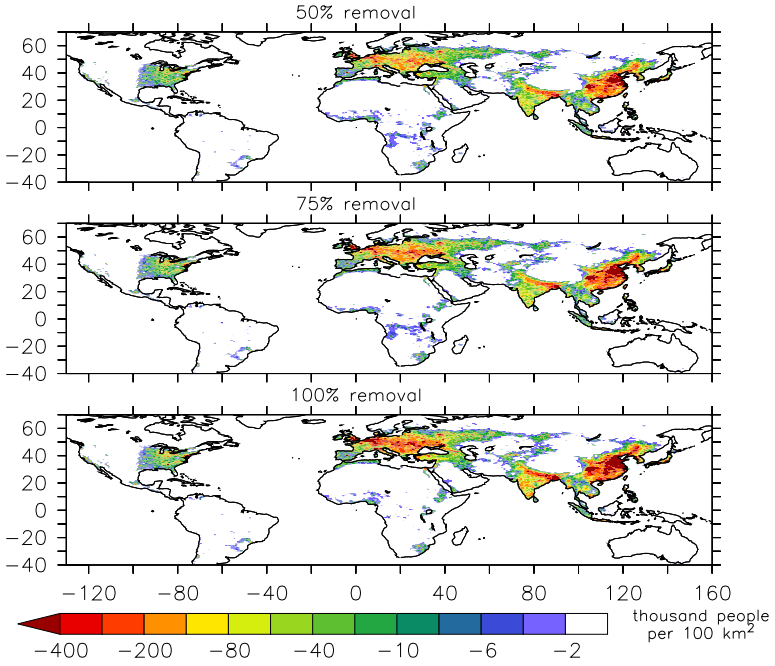

Figure 7. Annual average mortality attributable to $PM_{2.5}$ concentration changes (in $\mathrm{thousands\,people/100\,km^2}$) from the three scenarios with agricultural emissions reductions of 50, ~~66~~ 75 and 100% (top, middle and bottom, respectively).

et al., 2015). The model results where interpolated to the finer grid of the population map (Center for International Earth Science Informatio due to the coarse model resolution used in this study, it is expected to have an underestimation of exposure in urban areas. As discussed in the supplement of Lelieveld et al. (2015) , an uncertainty range of about ±50% is estimated for the mortality attributable to air pollution. The results, presented in Tab. 4 and Fig.7, show that a reduction of 50% in agricultural emissions could have a large impact on air pollution related mortality, reducing it worldwide by ∼ 8% i.e., affecting 250 (95% confidence interval (CI): 148-290) thousand people/year. North America would benefit from a large relative change, reducing the number of deaths by ∼ 30% (16 (95% CI: 10-19) thousand people/year), followed by Europe (∼ 19%, 52 (95%CI: 41-53) thousand people/year) and East Asia (∼ 8%, 105 (95% CI: 53-116) thousand people/year), while almost no effects are found over South Asia (∼ 3%, 25 (95% CI: 14-33) thousand people/year). The relatively large effect in North America is explained by the shape of the integrated response function (Burnett et al., 2014), which predicts a steep change in the attributable fraction at relative low $PM_{2.5}$ concentrations. If it were possible to fully phase out agricultural emissions, the global reduction of $PM_{2.5}$ related mortality would reduce by about 801 thousand people per year (95% CI: 417-984). In Europe the number would be reduced by about 222 thousand (95% CI: 139-249), in North America by 40 thousand (95% CI: 17-61), in East Asia by about 343 thousand per year (95% CI: 159-401) and in South Asia by 82 thousand per year (95% CI: 45-110) (Table 4).

Ammonia reduction policies should consider the intricate and non-linear effects through gas-aerosol partitioning and multi-phase chemistry (including aerosol pH changes) and therefore a coherent decrease of ammonia, nitrogen and sulfur emissions

is recommended. A coupled reduction of NH_3 and acid precursor emissions (e.g., SO_2) cannot only limit the decrease in aerosol pH but can also lead to a more efficient reduction of $PM_{2.5}$ concentrations than ~~the~~ an NH_3 emission control alone, as shown by (Tsimpidi et al., 2007). In the electronic supplement, a table showing the changes in mortality for the top 100 most populated countries is presented. Consistently with the results of Lee et al. (2015), Central and East European countries would benefit strongly from agricultural emission reductions , decreasing drastically the per capita air pollution related mortality. This can be seen also in Fig. 5, as the strongest relative changes in $PM_{2.5}$ due to agricultural emissions reduction are found in Central and East Europe, where a 50% emission reduction would decrease mortality attributable to air pollution by $\sim$ 15-20%.

It must be emphasized that, although many epidemiological studies have linked long term $PM_{2.5}$ exposure to public health outcome, it is yet unclear if any particular aerosol components and/or source categories are predominantly responsible for air pollution related mortality. The debate is open and firm conclusions of a specific relationship have not been reached (Harrison and Yin, 2000; Reiss et al., 2007), although it is expected that some aerosol components may be more toxic than others ~~(Shiraiwa et al., 2012)~~ (Shiraiwa et al., 2012; Mar et al., 2006; Ito et al., 2006) .

Table 4. Mortality attributable to air pollution in thousands people per year. In parenthesis the minimum-maximum range.

Region	*REF*		*REF*_50		*REF*_75		*REF*_100	
	average	range	average	range	average	range	average	range
Europe	277	(148 - 414)	225	(107 - 361)	176	(66 - 313)	55	(9 - 165)
North America	54	(21 - 100)	38	(11 - 81)	26	(6 - 65)	14	(4 - 39)
South Asia	778	(410 - 1140)	753	(396 - 1107)	750	(395 - 1102)	696	(365 - 1030)
East Asia	1381	(607 - 1929)	1275	(553 - 1812)	1195	(514 - 1719)	1037	(447 - 1527)
World	3155	(1523 - 4603)	2905	(1375 - 4313)	2739	(1280 - 4123)	2353	(1106 - 3619)

4 Conclusions

Pinder et al. (2007) showed that in North America emission controls of SO_2 and NO_x are likely to be very costly and probably less efficient than decreasing agricultural emissions. Therefore, the regulation of ammonia emissions from agricultural activities offers the possibility of relatively cost-effective control policies for $PM_{2.5}$. Our model simulations indicate that a 50% decrease of ammonia emissions could reduce the annual, geographical average near-surface $PM_{2.5}$ concentrations up to $\sim$ 1.0 (11%), 0.3 (8%), 1.6 (5%) and 0.6 (2%) $\mu g/m^3$ in Europe, North America, East Asia and South Asia, respectively. The reduction can even be larger during winter (up to $\sim$ 1.3 (11%), 0.6 (15%), 2.2 (5%) and 1.0 (3%) $\mu g/m^3$, respectively) when particulate ammonium nitrate concentrations are typically higher than in summer.

Our model simulations underscore that strong non-linearity plays a role in the sulfate-nitrate-ammonia system, which affects the efficiency of $PM_{2.5}$ controls, especially during summer when the sulfate/nitrate ratio is high. A strong reduction of $PM_{2.5}$ in response to NH_3 emission regulation is expected once the ammonia-limited regime is reached. As a result, the possible $PM_{2.5}$ reduction could be as large as $\sim$34% and $\sim$17% in Europe and North America, respectively. Our results also suggest that ammonia emission controls could reduce the particle pH up to 1.5 pH units in East Asia during winter, and more than 1.7

pH units in South Asia, theoretically assuming complete agricultural emission removal, which could have repercussions for the reactive uptake of gases from the gas phase and the outgassing of relative weak acids.

Furthermore, the global mortality attributable to $PM_{2.5}$ could be reduced by ~ 250 (95% CL: 148-290) thousand people/year worldwide by decreasing agricultural emissions by 50%, with a gain of 16 (30%), 52 (19%), 105 (8%) and 25 (3%) thousand people/year in Europe, North America South Asia and East Asia, respectively. A total phase-out of agricultural emissions would even reduce the mortality attributable to air pollution worldwide by about 801 (25%) thousand people/year, in Europe by about 222 (80%) thousand people/year, in North America by about 40 (74%) thousand people/year, in South Asia by about 82 (10%) thousand people/year and in East Asia by about 343 (25%) thousand people/year. These strong impacts are related to the non-linear responses in both the sulfate-nitrate-ammonia system and the exposure-response functions at relatively low $PM_{2.5}$ concentrations.

Therefore, emission control policies, especially in North America and Europe, should involve strong ammonia emission decreases to optimally reduce $PM_{2.5}$ concentrations, as well as further reduction in sulfur and nitrogen oxides emissions to avoid strong acidification of particles.

Data availability. The data from all model integrations are available from the authors upon request.

Competing interests. No competing interests are present.

Acknowledgements. V. Karydis acknowledges support from a FP7 Marie Curie Career Integration Grant (project reference 618349). A. P. Tsimpidi acknowledges support from a DFG Individual Grant Programme (project reference TS 335/2-1).

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