

We thank the referees for their thoughtful and constructive comments. We have addressed the comments (numbered, below), with referee comments in quotes and italics, and our responses in plain text.

Referee #1

Major comments:

1. *“Pg 3, line 19: The authors offered clear explanation for the decreasing tendency of SO₂ and NO_x, which is a result of regulation. However, it seems less clear to me why NH₃ is increasing, although the authors have tied NH₃ emissions with population growth previously. It would be better to explicitly state that the increase of NH₃ emissions is due to the increase of farming activities and fertilizer applications, in order to support the growth of population. I would also suggest adding something about the potential increase of ammonia emission due to global warming, such as the study of Skjøth and Geels 2013. Skjøth, C., and Camilla Geels. "The effect of climate and climate change on ammonia emissions in Europe." Atmospheric Chemistry and Physics 13 (2013): 117-128.”*

To emphasize the relationship between NH₃ emission and population due to food production, we have revised Page 3 Line 11 to “Given that fertilizer usage supports food production for about half the global population (Erisman et al., 2008), NH₃ emissions are linked to world population and so expected to increase into the 21st century (Gerland et al., 2014).” In the introduction, we have added a sentence citing the suggested publication, “Higher temperatures resulting from global warming can also potentially enhance NH₃ emissions (Skjøth and Geels, 2013).”

2. *“Pg 5, line 5. “With high NH₃ concentration, it is somewhat representative of northwestern Europe.” I would suggest the authors to provide additional evidence for this claim. Perhaps, some reference which indicate that northwestern Europe is normally have high NH₃ concentration. Or, maybe provide the averaged NH₃ concentration value on northwestern Europe and compared it with the averaged NH₃ concentration in Cabauw.”*

We have provided more information and noted that Cabauw has high NH₃ levels; “Northwestern Europe has fairly high NH₃ concentrations with yearly averages ranging from 1 to 14 $\mu\text{g m}^{-3}$ (median as 4.2 $\mu\text{g m}^{-3}$) for the Netherlands in 2013, reported by the Measuring Ammonia in Nature (MAN) network (Lolkema et al., 2015). Satellite-derived 14 years average for the western Europe is 3 ppbv ($\sim 2.3 \mu\text{g m}^{-3}$) (Warner et al., 2017). Cabauw was somewhat higher due to intensive agriculture in the region with observed yearly NH₃ average of $7.3 \pm 6.0 \mu\text{g m}^{-3}$ (~ 10 ppbv).”

3. *“Pg 6, line 15. “Inorganic ions are also assumed to be only in the aqueous phase.” Does the model assume that all aerosol species are in the aqueous phase or it also consider some of the species in solid state? Please clarify.”*

Yes, we ran the model assuming that all ions are in the aqueous phase. We have revised as “Inorganic ions are also assumed to be only in the aqueous phase (i.e., no solid precipitates)” to minimize confusion.

4. *“Pg 6, line 29. Other studies show that existence of organic phase could also impact the NH₃ and NO₃ partition as some SOA could react with NH₃ and reduce the NH₃ concentration. Add comments. Zhu, S., Horne, J.R., Montoya-Aguilera, J., Hinks, M.L., Nizkorodov, S.A. and Dabdub, D., Modeling reactive ammonia uptake by secondary organic aerosol in CMAQ: application to continental US.”*

We have revised to “This is confirmed by the good agreement between measured and ISORROPIA-II predicted NH₃-NH₄⁺ partitioning without considering organic acids or other organic species (see section 3.2). Although recent modeling study has suggested that ambient NH₃ concentration can be decreased by as much as 31% in winter and 67% in summer in the US, due to the reactive uptake of NH₃ by secondary carbonyl compounds (Zhu et al., 2018), this process doesn’t appear to have an impact on NH₃-NH₄⁺ partitioning and predicted pH for the locations in this study.”

With the above said, it is also important to note that even if NH_{3(g)} were reduced by 30-60% by reactions with the organic phase, the impact on aerosol acidity would be modest (change of about 0.1-0.2 pH units) given that an order of magnitude change in ambient NH₃ concentration is required for pH levels to be changed by one unit (Guo et al., 2017b).

5. *“Pg 7, line 4-5. The authors used two “discussed below” in this sentence. It would be better to give the exact section or location of the discussion instead. Does it refer to the first paragraph of 2.3? Actually, there is research showing that different mixing assumption could have significant impact on NO₃- and NH₄⁺ partition, especially on NO₃-: Zhu, S., Sartelet, K., Zhang, Y. and Nenes, A., 2016. Three-dimensional modeling of the mixing state of particles over Greater Paris. Journal of Geophysical Research: Atmospheres, 121(10), pp.5930-5947.”*

We have provided the sections that we refer to, “(discussed below in Section 2.3)...(discussed below in Section 3.2 and also see Table S1)”. Thanks for pointing out this paper, we have cited it in the manuscript. We have not discussed it in detail since it follows the less quantitative approach of assuming nitrate only forms once sulfate is fully neutralized (i.e., NH₄⁺/SO₄²⁻ ratios above 2), instead of a rigorous thermodynamic analysis, the focus of this paper.

6. *“P11, line 15. The authors should provide more details regarding to the nature of “particle artifacts in the gas collection system” that is affecting the measurement of HNO₃ and HCl.”*

We have edited this, removing speculation on the cause. It now reads, “However, for unknown reasons, gas-phase components of these two species showed significant discrepancies (R² of 0.13 to 0.17). We note that it may be associated with the very low gas phase concentrations of these species, in contrast to NH₃.”

7. *“Pg 17, line 19. Since the calculations are based on site measurement in this study, does it suggest that the pH calculated here is closer to the reality than the one calculated by Pozzer et al., (2017). Or, on the other hand, is it possible that the measurements site is not representative enough for the larger domain used in the global model calculation due to its coarse resolution? Are there any regional simulation results that is consistent with the pH prediction presented here?”*

This is a good point. Pozzer et al. (2017) did not publish any NH_3 concentration, which could be used for comparison with observations in Cabauw. Other relevant regional simulated pH could also not be found for the same region. Pozzer et al. (2017) published an average pH for Europe of ~ 2 , which is 1.7 units lower than the one-year pH in Cabauw. This may indicate that the Cabauw sampling site is not representative of Europe in general (the Pozzer’s paper), but a detailed comparison between the two model inputs is necessary to understand the cause of the pH difference. As noted above (and in the revised manuscript), the Cabauw site has higher NH_3 concentrations, which will increase particle pH. Holding all other model inputs constant, a factor of 10 lower NH_3 decreases pH by roughly one unit (as shown in Figure 3). Based on satellite derived 14-year NH_3 average (3 ppb and $2.3 \mu\text{g m}^{-3}$) for western Europe as an example (Warner et al., 2017), the pH in Cabauw decrease to around 3 for the one-year average and winter average, and below 3 for summer, according to the linear fitting lines in Figure 3. Meteorological conditions and particle composition also contribute to the difference in pH prediction mentioned above. Although it has been found that inaccurate treatment of nonvolatile cations may cause overestimation of particle pH in regional models (Vasilakos et al., 2018), it doesn’t explain the above pH difference since the modeled pH in Pozzer et al. (2017) (from global modeling) is the lower one and the levels of nonvolatile cations are low in Cabauw. Despite of the difference in pH, we believe the findings of Pozzer et al, (2017) are consistent with the framework established in this study, that is a critical pH of ~ 3 . Since the Pozzer’s European pH is 2, below 3, controlling NH_3 emission is suggested as an effective way to reduce particle mass.

8. *“Pg 18, line 13-14. This conclusion looks not very convincing to me. Since the particle composition is so different between SE US and NE US, the author should justify how the SE US could be a representative case for the eastern US in the summer, and how the NE US could be representative case for the eastern US in the winter before drawing such a conclusion. Or latest explain the cause of such a high sulfate composition (76%) in the SE US case.”*

We have clarified the statement as suggested by the reviewer. The SE US simulation is only representative of the SE US; the same for the NE US simulation. Now the text becomes “Therefore, it is more effective to control NH_x in winter in the NE US and SO_4^{2-} in summer in SE US, a finding consistent with previous studies (Duyzer, 1994; Tsimpidi et al., 2007).” The large fraction of sulfate is a result of the small fraction of nitrate. In such situations, ammonium basically tracks sulfate. Due to the difference in molecular weight, sulfate is the dominant inorganic mass. We have added a sentence explaining the reason, “A small fraction of nitrate aerosol is typically observed in the southeast in summer (Hidy et al., 2014) due to the high temperature and low particle pH.”

Minor comments:

9. "Pg 5, line 13, the word "alternatively" here is confusing. Do you mean it is the first hour measurement is for PM1 and the next hour will be for PM2.5? In that case the measurement interval will be 2 hours for either PM1 or PM2.5, is that the case? Please clarify."

A clarification is made. "...alternatively between PM₁ and PM_{2.5}, each size sampled hourly (i.e., a two-hour interval for one size; a one-hour interval for gas)."

10. "Pg 7, line 9. "In Cabauw, it has been reported..." Could reference be provided for this report?"

The reference "(Schlag et al., 2017)" was there in the middle of the sentence. Since it is not obvious, we have moved it to the end.

11. "Pg 7, line 25. It would be better to specify the "coarse mode salts" that HNO₃ evolved into."

We have added examples as "coarse mode salts (e.g., NaCl and CaCl₂)".

12. "Pg 9, line 14. "0.987x10⁻¹⁴ is a unit conversion factor" I would better to specify which units are being converted with this factor."

We have added explanation as "where 0.987×10^{-14} is a unit conversion factor (from converting atm and μg to SI units)".

13. "Pg 10, line 3. Could the authors be more specific on how the "approximately 0.6" nonideality shifts are calculated? Or provide a reference S curve without the non-ideality effect?"

The 0.6 unit pH difference is provided by comparing nitrate partitioning S curves calculated assuming $\gamma_{\text{NO}_3^-} \gamma_{\text{H}^+} = 1$ (ideal) and 0.24 (non-ideal; from ISORROPIA). More specifically, compare pH₅₀ values for the two S curves. A figure is provided to visualize the difference and added to supplemental material as the new Fig. S2.

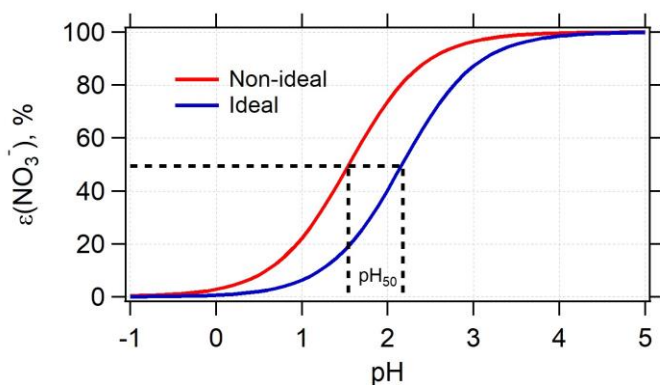


Figure. Predicted particle phase fraction of total nitrate, $\epsilon(\text{NO}_3^-)$, versus pH for one-year average condition in Cabauw based on Eq. (4). The red and blue lines are based on $\gamma_{\text{NO}_3^-}\gamma_{\text{H}^+} = 0.24$ and 1, respectively.

14. *“P11, line 17. Could the authors provide the references for those “previous studies” mentioned here?”*

A reference has been added.

15. *“P12, line 2. Could the authors provide the exact hour ranges used in this study to define “night” and “daytime”?”*

We define “night” and “day” by sunrise and sunset. However, we don’t have solar radiation data to plot a diurnal profile. Since sunrise and sunset time can vary substantially from summer to winter, we cannot provide exact hour ranges. For example, daytime is from 05:24 to 22:03 on June 1 2013 and from 08:48 to 16:38 on Dec 31 2013. To minimize confusion, we have revised the text to exact hours which are not as affected by seasonal changes in sunrise and sunset, “A diurnal pattern of ambient particle pH is observed in Cabauw, similar to other studies (Guo et al., 2015). For example, for the nighttime period of 1 am to 7 am, the average pH is 3.9, whereas for the daytime period of 1 pm to 6 pm the pH is 3.5. The difference is mainly driven by the diurnal variation in liquid water content (see Fig. S1)”.

16. *“Pg 12, line 13. I found it confusing that the authors keep changing between “NE US” and “WINTER” for the Guo et al., (2016) case, for example, “WINTER” is used in Figure 2, but “NE US” is used here in the text. I suggest the authors use more consistent expression.”*

We have revised the Figure 2 legends to separate the SOAS and WINTER studies. The SOAS study is under the “Southeastern US” and the WINTER study is under the “Northeastern US”. Hopefully, this clarifies the issue.

17. *“Pg 19, line 4. The previous discussions in this paragraph are based on Cabauw winter and Beijing, while the 19%“(NH₄⁺) value used here are from one-year Cabauw, would you explain why?”*

Thanks for pointing it out! We have replaced “19%” with “27%”, which was the right number for Cabauw winter average.

18. *“Pg 20, line 12. What does “further from the actual ambient particle pH” referred for? Do you mean the region 2 of the curve is further from the ambient particle pH?”*

Yes, we mean region 2 or pH₅₀. We have revised to “This is explained by a shift in the HNO₃-NO₃⁻ partitioning ($\epsilon(\text{NO}_3^-)$) curve to lower pH in winter and pH₅₀ (where $\epsilon(\text{NO}_3^-) = 50\%$) further from the actual ambient particle pH.”.

Referee #2

Major comments:

1. *“The authors introduce a new conceptual framework to explain seasonal and regional differences in the sensitivity of particulate matter to ammonia emissions. This has potential policy implications and it would be useful for the authors to compare with other techniques that have been used previously to highlight potential differences.*

*In particular, previous studies have used the gas-ratio from Ansari and Pandis to interpret global model results (see for instance Pinder et al. (2007, 2008), Paulot (2016), Pozzer et al., 2017) $GR = (TNH4 - 2*TSO4)/TNO3$ with $0 < GR < 1$ indicating sensitivity to NH_x and $GR > 1$ indicating sensitivity to NH_3 .*

Obviously, this cannot directly address variations associated with seasonality. However, based on the information provided in Table S1, $GR < 1$ only for SE US, Virginia, and Pasadena. In other words the weak sensitivity of nitrate to ammonia emissions at the other sites could be inferred simply from concentrations, which is consistent with the findings of the studies mentioned earlier.

In addition, many global models do not use ISORROPIA but simpler (cheaper) aerosol thermodynamic models (see for instance Bellouin et al (2011), Hauglustaine (2014)). Such schemes, which do not explicitly account for aerosol pH, will also simulate a nonlinear response of ammonium nitrate to changes in ammonia emissions (see equation A8 in Bellouin et al (2011)). It would be useful for the authors to show how different the response of nitrate and ammonium to changes in ammonia/ NO_x emissions (i.e., Fig 5) would be using such approach.

In particular, this would help strength the case for thinking in terms of aerosol pH rather than simply in terms of concentrations.”

The reviewer raises an important and very broad question. First we note that global models use aerosol thermodynamic modules of all levels of complexity (some not at all). All these models would predict some degree of nonlinearity because one of the precursors, NH_3 or HNO_3 , become limiting. Our point is that using pH to look at the sensitivity of nitrate to the precursors is new. It makes things simpler and provides a more fundamental understanding of the processes involved. Furthermore, even if the models have the correct thermodynamics they can still get the sensitivity wrong due to a biased predicted pH, as we note with the reference to Vasilakos et al. (2018). We feel that the degree to which each implementation differs, and how it compares with the usage of pH as a control parameter requires a dedicated publication in itself.

We have added some text to try and clarify these points. The next now reads:

“Large-scale models to assess effectiveness of NH_3 control requires good predictions of a range of pertinent emissions and sinks (NH_3 , NO_x , SO_2 , and nonvolatile cations), and accurate representation of their applicable atmospheric chemical processes. Thermodynamic modules

of different levels of complexity are then applied to determine sensitivities to the precursors (e.g., NH_3 , HNO_3). In some cases (Pozzer et al., 2017), the aerosol pH is explicitly determined with an embedded thermodynamic model, such as ISORROPIA-II (Fountoukis and Nenes, 2007). Due to the complexities from all these factors, chemical transport model-predicted responses to changing emissions may not align with observations. For example, the sensitivity of $\text{PM}_{2.5}$ pH in the Community Multiscale Air Quality (CMAQ) simulations to the mass of crustal material apportioned to the $\text{PM}_{2.5}$ size range can have important effects on anticipated responses to these changing emission trends. Vasilakos et al. (2018) have shown that including too much crustal material in $\text{PM}_{2.5}$ results in a predicted increasing trend in both aerosol pH and concentrations of NH_4NO_3 , which is counter to observations (Weber et al., 2016).

Overall, calculating aerosol pH is a more accurate approach that provides a fundamental understanding of the factors controlling HNO_3 - NO_3^- partitioning and therefore enables a direct evaluation of different studies. Furthermore, it is also useful to determine aerosol pH since it has broad application to many other important aerosol processes. For instance, pH is a mediator of many heterogeneous chemical processes, including various acid-catalyzed reactions (Jang et al., 2002; Eddingsaas et al., 2010; Surratt et al., 2010), gas-particle partitioning of species other than HNO_3 and NH_3 , such as organic acids and halogens (Fridlind and Jacobson, 2000; Young et al., 2013; Guo et al., 2017a; Nah et al., 2018), and solubility of metals and other nutrient species (Meskhidze et al., 2003; Nenes et al., 2011; Longo et al., 2016; Stockdale et al., 2016; Fang et al., 2017).

In this study, we apply a more direct approach, where measured gas and particle concentrations and the thermodynamic model ISORROPIA-II are used directly in a sensitivity analysis to evaluate the effectiveness of NH_3 emission controls on fine particle mass relative to NO_x control. Contrasts are made between sites that have a wide range in NH_3 concentrations and aerosol composition, ...”

2. *“I am not convinced by the current discussion of the impact of NH_3 emissions controls on nitrogen deposition. The authors argue that lowering aerosol pH (via lower NH_3 emissions) will modify the ratio of reduced to oxidized nitrogen deposition. However, it is unclear why this is important (no reference is given), especially considering the benefits of lower NH_x deposition and the existence of other removal pathways (wet deposition) that may not exhibit the same sensitivity to the NH_4/NH_3 partitioning. A longer discussion is needed given that this conclusion is highlighted in the abstract.”*

We were only focusing here on effects on dry deposition since the paper discusses relative gas and particle concentrations and we note the large differences in gas/particle deposition velocities. Discussing effects of N deposition due to wet processing is beyond the scope of this paper. However, the reviewer's point that it may be more complicated is well taken. We have modified the text to be more precise and note complicating effects of wet removal processes.

In the abstract, it has been revised to “Finally, controlling NH_3 emissions to increase aerosol acidity and evaporate NH_4NO_3 will have other effects, beyond reduction of $\text{PM}_{2.5}$ NH_4NO_3 , such as increasing aerosol toxicity and potentially altering the deposition patterns of nitrogen and trace nutrients.”

In the section 3.5, “Lowering particle pH through NH₃ reductions will decrease overall reduced nitrogen deposition but may results in more localized oxidized nitrogen dry deposition if the lower pH results in NO₃⁻ evaporation and higher HNO₃ concentrations. Deposition due to wet removal processes are not considered here.”

3. *“the authors focus on seasonal averages. It would be interesting to discuss whether the sensitivity of particulate matter to NH₃ emissions is different depending on the concentration of NO₃ and whether this would affect the probability distribution of PM under the different emission reduction scenarios shown in Fig. 5. This may be important for policy makers as some standards are based on 24hr averages (https://www3.epa.gov/ttn/naaqs/standards/pm/s_pm_history.html)”*

We understand the point raised. The main issue with this suggestion is that the sensitivity of particulate matter to NH₃ (or HNO₃) emissions is directly determined by the aerosol pH. Otherwise, the sensitivity can take a wide range of values for constant NO₃⁻, as the pH can still vary considerably. We believe that this shift in approach (first looking at pH and then seeing how that affects aerosol sensitivity to emissions), is one of the most important messages of the paper. Towards that, a simpler approach, the HNO₃-NO₃⁻ S curve (in Section 2.3), is provided to roughly estimate the effectiveness of NH₃ control.

Technical comments:

4. *“p4 line 5 NH₃ can also enhance the in-cloud oxidation of SO₂ by O₃. See for instance Wang (2011) or Paulot (2017)”*

Thanks for bringing attention to these references. We have revised the text to “Reduction in NH₃ also reduces the amount of NH₄⁺ associated with sulfates and lowers the pH-dependent sulfate production rate, such as in cloud SO₂ oxidation by O₃ (Wang et al., 2011; Cheng et al., 2016; Paulot et al., 2017), and the interplay between the two species may drive much of the sensitivity of PM_{2.5} to NH₃ and NO_x reductions (e.g., (Vasilakos et al., 2018))”.

5. *“p17 line 15 I believe livestock emissions are likely to dominate ammonia emissions in summer.”*

We had thought that as well, but literature studies do not seem to agree. Based on Figure 7 in Zhang et al. (2018), livestock waste dominates in winter rather than summer. The annual emissions from fertilizer and livestock waste are quite similar (5.05 vs 5.31 Tg a⁻¹).

6. *“dash black line Fig. 4 not defined”*

We apologize for this oversight. The black dash lines in the pH figures identifies the critical pH value of 3, and now has been noted in the caption.

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