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 NOx, 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 NH_3 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), NH_3 emissions are linked to world population and so expected to increase into the 21th 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 NH_3 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 NH_3 levels; “Northwestern Europe has fairly high NH_3 concentrations with yearly averages ranging from 1 to 14 $\mu g\ m^{-3}$ (median as 4.2 $\mu g\ m^{-3}$) 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 (~2.3 $\mu g\ m^{-3}$) (Warner et al., 2017). Cabauw was somewhat higher due to intensive agriculture in the region with observed yearly NH_3 average of 7.3 ± 6.0 $\mu g\ m^{-3}$ (~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 NH_3-NH_4^+ partitioning without considering organic acids or other organic species (see section 3.2). Although recent modeling study has suggested that ambient NH_3 concentration can be decreased by as much as 31% in winter and 67% in summer in the US, due to the reactive uptake of NH_3 by secondary carbonyl compounds (Zhu et al., 2018), this process doesn't appear to have an impact on NH_3-NH_4^+ 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 NH_3 concentration is required for pH levels to be changed by one unit (Guo et al., 2017).

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. Threeâ˘AŘ 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., NH_4^+/SO_4^{2-} 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 new reads, "However, for unknown reasons, gas-phase components of these two species showed significant discrepancies (R^2 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 NH_3."

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 µg 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 PM_1 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 $CaCl_2$)".

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 $\gamma_{NO_3^-}\gamma_{H^+} = 1$ (ideal) and 0.24 (non-ideal; from ISORROPIA). More specifically, compare pH_{50} values for the two S curves. A figure is provided to visualize the difference and added to supplemental material as the new Fig. S2.

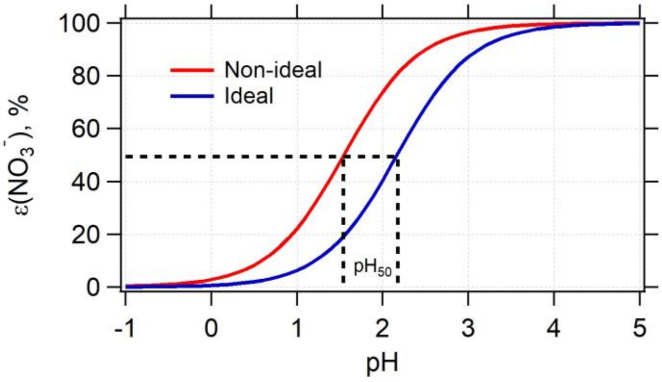

Figure. Predicted particle phase fraction of total nitrate, ε(NO_3^-), versus pH for one-year average condition in Cabauw based on Eq. (4). The red and blue lines are based on $\gamma_{NO_3^-}\gamma_{H^+} =$ 0.24 and 1, respectively.

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 pH_{50}. We have revised to "This is explained by a shift in the HNO_3-NO_3^- partitioning (ε(NO_3^-)) curve to lower pH in winter and pH_{50} (where ε(NO_3^-) = 50%) further from the actual ambient particle pH.".

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