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 #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 NHx and GR>1 indicating sensitivity to NH3.*

   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 a 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/NOx 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 $PM_{2.5}$ pH in the Community Multiscale Air Quality (CMAQ) simulations to the mass of crustal material apportioned to the $PM_{2.5}$ 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 $PM_{2.5}$ 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., 2017; 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 NH3 emissions controls on nitrogen deposition. The authors argue that lowering aerosol pH (via lower NH3 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 NHx deposition and the existence of other removal pathways (wet deposition) that may not exhibit the same sensitivity to the NH4/NH3 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 $PM_{2.5}$ 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 NH_3 reductions will decrease overall reduced nitrogen deposition but may results in more localized oxidized nitrogen dry deposition if the lower pH results in NO_3^- evaporation and higher HNO_3 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 NH_3 (or HNO_3) emissions is directly determined by the aerosol pH. Otherwise, the sensitivity can take a wide range of values for constant NO_3^-, 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 HNO_3-NO_3^- S curve (in Section 2.3), is provided to roughly estimate the effectiveness of NH_3 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 NH_3 also reduces the amount of NH_4^+ associated with sulfates and lowers the pH-dependent sulfate production rate, such as in cloud SO_2 oxidation by O_3 (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 NH_3 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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