

This study focused on an important and interesting topic of the rapid sulfate formation pathway in hazes. The modelling work in this study is sound and clearly described. In addition, the paper is clearly written and well-organized. However, discussion on the results is relatively weak. In my view, major drawback of this study is that, it spent too much effort discussing the model performance, while too little about the underlying reasons. The following concerns should be addressed before it can be published.

Major Comments:

1. In the abstract and conclusions etc., the authors simply claimed that doubling the ammonia emissions can improve the model performance, without any justification of doing so. As the predicted NH₃ in base scenario is significantly underestimated (Page 9, Line 6), increasing the emissions seems reasonable, but the authors should clarify on this issue.

2. Although doubling the ammonia emissions will result a higher NH₃ in models than observations (NMB of 34%; Page 9, Line 7), the predicted NH₄⁺ is still much lower than observation (NMB of -31%, Page 11, Line 5). As a result, doubling the ammonia emissions may generate a reasonable total ammonia (NH₃ + NH₄) concentrations? If so, it would further support the model modification of doubling ammonia emissions.

3. The causal relationship among aerosol pH and the contribution of SO₂+NO₂ reactions seems confusing in this study. Prediction of aerosol pH in ISORROPIA model is based on the aerosol compositions. However, the model underestimation of nitrate and ammonium (section 3.2.2) is more significant than that of sulfate (section 3.2.1). This will surely result in a lower aerosol pH in models than that predicted based on observation results. A comparison of model-based and observation-based aerosol pH should be conducted and discussed. For the aerosol compositions required in ISORROPIA while is not observed here (i.e., Na⁺, Ca²⁺, K⁺, Mg²⁺), the model predicted concentrations can be used.

4. Adding the SO₂ + NO₂ heterogeneous reactions alone (noHet vs. Het) cannot obviously improve the sulfate, which may be actually caused by the much lower aerosol pH predicted by the models than the actual (observation-based) pH. Doubling the ammonia emissions may push the predicted pH closer towards the actual pH, which is the actual reason why noHet_2NH₃ performs the best. These discussions should be added.

5. In the parameterizations of k₀, the influence of RH is actually the influence of aerosol water (see discussions in Cheng et al. (2016)), which depends on the average aerosol compositions. The difference of Beijing – Shanghai aerosol hygroscopicity (or growth curves) should be considered.

Minor Comments:

1. Page 1, Line 20: simulation of specific area “for the first time” is not the major contribution of this study, and should not be highlighted. The same issue applies for description in the Introduction (Page 3, Line 24).

2. Page 6, line 19: The classification of clean, transition and polluted conditions following the

Beijing scheme is weird. The authors should classify the schemes by local PM_{2.5} time series to classify the background, small peaks and large peaks (e.g., as described in Zheng et al. (2015b)), or by air quality standard (e.g., Zheng et al. (2016)).

3. Page 6, Line 26: The citation should be Cheng et al. (2016). Also the pattern is reported in Zheng et al. (2015b), not Zheng et al. (2015a).

4. Page 11, Line 32: citations to the references are not in the proper format.

5. Page 12, Line 4: many observation-based ISORROPIA predictions is against the conclusion that “aerosol pH is always acidic” (e.g., Shi et al., 2017) . The authors should be more through citing the references.

References

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