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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia" *by* Y. Wang et al.

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We thank the reviewer for his/her careful reading and thoughtful and constructive comments to improve the analysis and writing of the manuscript. The page and line numbers refer to those in the tracking-change version of the revised manuscript.

1. The authors conclude that model prediction accuracy is good based on evaluation with limited observation data and on monthly scale. The authors have used monthly/annual data even at sites where weekly observations were available. It might

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be better to evaluate the model performance on weekly and monthly data separately

Response: We changed the model-observation comparison at Miyun and Beibei sites from monthly average to weekly average. Please refer to the new Figure 6 and the revised discussion in the revised manuscript (pg 14, line 296-312). The conclusion of the model's performance still holds after using the weekly average for comparison.

2. In page 24246 line 23 mention about use of satellite –derived proxies like AOD. Have the authors considered using satellite AOD data for model performance evaluation and how the SNA system impacts AOD spatial distribution over China?

Response: Ground-level PM2.5 concentrations can be derived from satellite retrieved AOD data in conjunction with chemical transport models which provide the quantitative relationship from column-based AOD to surface PM2.5 mass (van Donkelaar et al., 2006, and 2010). As our paper focuses only on one component of PM2.5–secondary inorganic aerosols, we chose not to involve satellite-derived AOD data for model evaluation here because many other factors (e.g., meteorology, optical property calculation, other chemical components in PM2.5, satellite's retrieval uncertainty) will come at play in causing the differences between modeled AOD and satellite retrievals and complex analysis will be needed in order to attribute the model's bias in total AOD to SNA. This could complicate and potentially distract from the main discussion of our manuscript. We have added the discussion on the potential of satellite-retrieved AOD in the introduction (pg 5, line 108-119). We will explore this topic in future research, particularly the reviewer's suggestion that the SNA system impact on AOD spatial distribution over China.

3. The study concludes that NH3 emissions are the primary reason for over prediction despite modeling biases in wet deposition parameters. It would be better to include sensitivity analysis based on deposition parameters to make the above conclusion more convincing.

Response: We agree. As suggested by the reviewer, we conducted a sensitivity sim-

ACPD

12, C10715–C10719, 2012

ulation in which the rate constant of nitrate wet deposition is increased by 40%. The resulting change is 20% higher wet deposition flux of nitrate and 7% higher nitrate concentrations at the surface, compared with the standard simulation. This sensitivity test suggests that the simulated nitrate concentrations are less sensitivity to wet deposition parameters than to NH3 emissions, lending support to our statement that NH3 emissions are the primary reason for nitrate over-prediction. We have added a discussion of the sensitivity simulation in the revised manuscript (pg 15, line 333-336).

Specific comments:

1. Include more details on use of potential satellite data for model evaluation in page 24246.

Response: Done, please refer to the revised paper (pg 5, line 108-119).

2. In section 2.1 add more details on GEOS-Chem/GOES model

Response: Done, please refer to the revised section 2.1.

3. In page 24249 what does poor NH3 conditions mean?

Response: We have changed 'poor NH3 conditions' to 'NH3-limited conditions'

4. Change "Februar" to "February" in page 24250 line 2

Response: Corrected.

5. In page 24251, the authors mention briefly about NO3 measurement uncertainties. Including this discussion with more details early on in the introduction section will be more insightful to the readers.

Response: We have added one paragraph at the end of section 2.2 to discuss the measurement uncertainties. Please to the revised manuscript (pg 11, line 243 – 253).

6. Better formatting of observations is recommended for clarity in Figure 4

Response: Corrected.

ACPD

12, C10715–C10719, 2012

7. In page 24253 line 24, the authors mention availability of bi-weekly observations but in introduction they mention weekly scale. Are the observations weekly or bi-weekly scale? Evaluating model with bi-weekly data will strengthen the modeling analysis.

Response: The observation data at the Miyun and Beibei sites is weekly scale. We have fixed the inconsistency and also changed model evaluation at these two sites to weekly scale. Please refer to the new Fig 6 and additions in Section 3 (pg 14, line 296-312) in the revised manuscript.

8. In page 24254 lines 7 and 19, the authors need to use caution in describing model performance as accurate as this conclusion is based on evaluating model with limited observation data

Response: Agree. We have changed “accurately” to “has some success”.

9. In page 24256 lines 7-8 the authors mention briefly about impact of high dust loading on SNA distribution. Including more details for dust impact particularly for spring season on SNA distributions will be helpful

Response: Nitric acid can react with calcium carbonate in dust to form calcium nitrate, and most of the nitrate formed by this pathway exists in the coarse mode, so this pathway will lead to reduced nitrate in the fine mode (PM2.5) which is the focus of our study. Furthermore, nitric acid uptake on coarse mode dust can also react with NO to produce NO2: HNO3 + NO → HONO + NO2, which can lead to lower HNO3 concentrations (Mochida and Finlayson-Pitts, JPC, 2000) and thus lower fine mode nitrate concentration in high dust loading regions. We found in the revision that the impact of dust on SNA cannot explain the simulated lower SNA concentrations over NC in spring, as the model version used in the manuscript does not include the module of HNO3 uptake on dust or its reaction with NO, so the discussion on dust’s impact on SNA is in fact irrelevant and should be removed. In the revised manuscript, we explain the low SNA concentrations in spring to high wind speed that favors dispersion and export of SNA to other regions (pg 20, line 461-462).

ACPD

12, C10715–C10719, 2012

10. Include the number of data points used in seasonal statistics calculation in Figure 5.

Response: Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24243, 2012.

ACPD

12, C10715–C10719, 2012