

Interactive comment on “Seasonal variation of fine and coarse-mode nitrates and related aerosols over East Asia: Synergetic observations and chemical transport model analysis” by Itsushi Uno et al.

Anonymous Referee #2

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The authors investigated inorganic particulate nitrate, ammonium, sulfate, and BC over an observation station at Fukuoka in East Asia using data from a 14-month observation and the corresponding GEOS-Chem model simulation. They analyzed the data over daily to seasonal variations and in fine and coarse mode particles, particularly for particulate nitrate. They also include the precursor gas tracers of NH₃ and HNO₃ in the discussion of their aerosol counterparts NH₄⁺ and NO₃⁻, respectively. This is an interesting and valuable study. However, the analysis could be more logical and concise. The scientific accuracy of its statistical analysis is problematic. A major revision

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is required before the paper is published in ACP.

Major Comments It would be better to re-organize the discussion content and clearly point out the unique information shown by each figure. I feel it is a bit difficult to follow the discussion that jumps back forth among figures. For example, section 4.3.4 (monthly variation in total NO₃ and NH₃) starts with a discussion on Figure 8 and follows up with Figure 5d, Figure 6a, and Figure 4a, c, d. I suggest merging Figure 3 and 4 together for daily variation and Figure 5 and 6 together for monthly variation. Please use monthly data to give seasonal and statistical findings and daily data to add any new findings on a daily time scale.

Another major concern is the scientific accuracy of the method used by the authors in explaining the relationship between model and observation based on the regression equation. For example, the authors concluded that the modeled PM_{2.5} value is approximately 58% of the observed value because the data regression gives $PM_{2.5}(\text{model}) = 0.58PM_{2.5}(\text{observation}) + 1.16$. From my understanding, a slope of 0.58 does not necessarily lead to that conclusion. A slope that is less than 1 could be a natural phenomenon owing to the representation issue of the model and observation. The GEOS-Chem model data is an average value over a grid box of $0.5^\circ \times 0.667^\circ$, while the observation is conducted over a station. Consequently, it is hard for the model to capture the maximum and minimum observational values. In other words, the simulated model results tend to be underestimated at the high end and overestimated at the low end, which results in a slope less than 1. The authors need to modify their explanation of the model-observation relationship throughout the manuscript.

Specific Comments 1. Abstract, lines 19-20 and lines 24-26 are duplicates. 2. Page 4 line 19: What are the secondary inorganic aerosols considered for this study? 3. Page 5 line 27: Do NH₃ emissions from agriculture and domestic animals belong to the emission of “anthropogenic emissions” mentioned here? 4. Page 6 line 10: How does GEOS-Chem convert coarse-mode sea salt to Na⁺ concentration? 5. Page 6 line 23 to Page 10 line 15: Reorganize the analysis as “Daily variation in PM_{2.5},

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PM10, aerosol compositions and CO” and “Monthly variation in PM2.5 and aerosol composition”. 6. Page 6 line 24: Please explain optical BC. What is its relationship with BC mass concentration? 7. Page 6 line 25: What does “(2h)” mean here? 8. Page 7 lines 9-14: Why do the authors show CO data in this paper? Is there any way to combine the analyses of CO and aerosols? 9. Page 7 line 12: Why does the high CO a product of long-range transport? 10. Page 7 lines 21-25: Merge Figure 5 and 6 since they are both for monthly average data and NH4 related to both SO4 and NO3. 11. Page 8 line 1: What does “intermittent high” mean here? 12. Page 8 lines 3-4: What is the SF1? Why do the authors mention SF1 event specifically here? Figure 4a shows that the model completely misses the observed peak SO4. 13. Page 9 lines 3-4: Why is cNO3 higher during fall? 14. Page 9 line 28: In sentence “. . . and NH3 gas is also higher even in winter”, “higher” than what? 15. Page 9 lines 29-31: How do T and precipitation influence NH3? 16. Page 10 line 4: Using “decreased bias” here may be misleading. My understanding is that the missing emission in model results in an increased emission bias. 17. Page 10 lines 4-9: How do you know that the HNO3-high (-low) and NH3-low (-high) relationship may retain the same equilibrium constant? Is there any evidence to support the authors’ conclusion? There are series of aqueous reactions between gases (i.e. NH3 and HNO3) and solid aerosol (i.e. NH4NO3). In NH3 poor regime, NH3 emission cloud matters. 18. Figure 4b and c: Which of the two observation data shown in the figure does “Obs” in the regression equation refer to?

Technique Corrections 1. Abstract lines 21-24: Please change this to “Observational data confirmed that coarse NO3 (cNO3) made up the largest proportion (i.e. 40-55%) of total nitrate (defined as the sum of fNO3, cNO3, and HNO3) during the winter, while HNO3 gas constituted approximately 40% of total nitrate in summer. fNO3 peaked during the winter.” 2. Abstract line 24: Change “A” to “The”. 3. Page 4 line 14: Delete “the” before “a” 4. Page 10 line 1: add “(Fig. 5d)” after “NH3” and add “(Fig. 6a)” after “HNO3”. 5. Page 10 line 2: There is no figure Fig. 4d.

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