

## **Responses to the reviewer comments on**

### **“Multiphase MCM/CAPRAM modeling of formation and processing of secondary aerosol constituents observed at the Mt. Tai summer campaign 2014” by Zhu et al.**

The authors would like to thank both reviewers for their constructive and good suggestions to improve our manuscript. We have carefully considered all the review comments and revised the manuscript. Below, we provide responses to the comments in **blue**, with changes made in the manuscript highlighted in **red**.

#### **Response to Reviewer 1:**

*The authors report a detailed multiphase chemical modeling study of the formation and processing of secondary aerosol compositions during transport to the Mt. Tai in summer 2014. The model performance of MCM/CAPRAM was evaluated against the field observations, and the day vs. night and with cloud vs. non-cloud processes were examined. The major formation pathways and key precursors of sulfate, nitrate, ammonium, and DCRCs were identified with the model. The impacts of emissions and glyoxal partitioning constants on the modeling results were also estimated by sensitivity studies. Despite an increasing number of field observational studies of secondary aerosols in recent years in China, such kind of detailed multiphase modeling study is still lacking. This study is helpful for better understanding the regional formation and processing of secondary inorganic and organic aerosols in the North China Plain. Therefore I would recommend that this manuscript can be considered for publication at ACP after the following specific comments being addressed.*

**Response:** We thank reviewer#1 for the helpful comments. Below, we address the comments and have revised the manuscript accordingly. For clarity, the reviewer's comments are listed below in *black italics*, whereas our responses and changes in manuscript are shown in **blue** and **red**, respectively. Revised Tables and Figures are put in the end.

1. P2 L58 “However, formation pathways based on measured data are still limited”:  
*rephrase this sentence.*

Response: We have rephrased this sentence as follows:

However, modeling studies that focus on understanding of DCRCs formation pathways based on field measurements are limited.

(Page 2, Line 58-59)

2. P3 L67-68: *this sentence is not clear. Do the Yangtze River Delta and Bohai Rim have a total of 410 million populations? Additionally, the commonly used three largest economic zones in China don't include the Bohai Rim.*

Response: Yes, the intended meaning was to say that both together have a population of around 410 million. In addition, we checked the China Statistical Yearbook in 2019, and found that together, the Yangtze River Delta and Bohai Rim regions had a population of more than 450 million in 2018. Therefore, we changed the sentence in the revised manuscript as follows:

Mt. Tai is located in Shandong province in the NCP, and between the Bohai Rim (BHR) and the Yangtze River Delta (YRD) regions. Together, the BHR and YRD regions had a population of more than 450 million in 2018 (China Statistical Yearbook in 2019).

(Page 3, Line 67-71)

3. P3 L83-84: *provide the standard deviations for the average temperature and RH values.*

Response: We have added standard deviations for the average temperature and RH values as follows:

$17 \pm 6.2 \text{ }^{\circ}\text{C}$

(Page 3, Line 87)

$87 \pm 13 \%$

(Page 3, Line 88)

4. P4 L98: *predicted and observed concentrations.....*

Response: We have changed “concentration” to “concentrations” in the revised manuscript as follows:

These limitations have to be kept in mind when studying deliquesced particles and comparing predicted and observed concentrations at Mt. Tai.

(Page 4, Line 113-114)

5. P4 Section 2.2: *I suggest the authors to provide the air mass cluster figures in the SI so that the readers can easily access the plot.*

Response: We have added air mass cluster Figure in the supplement as follows:

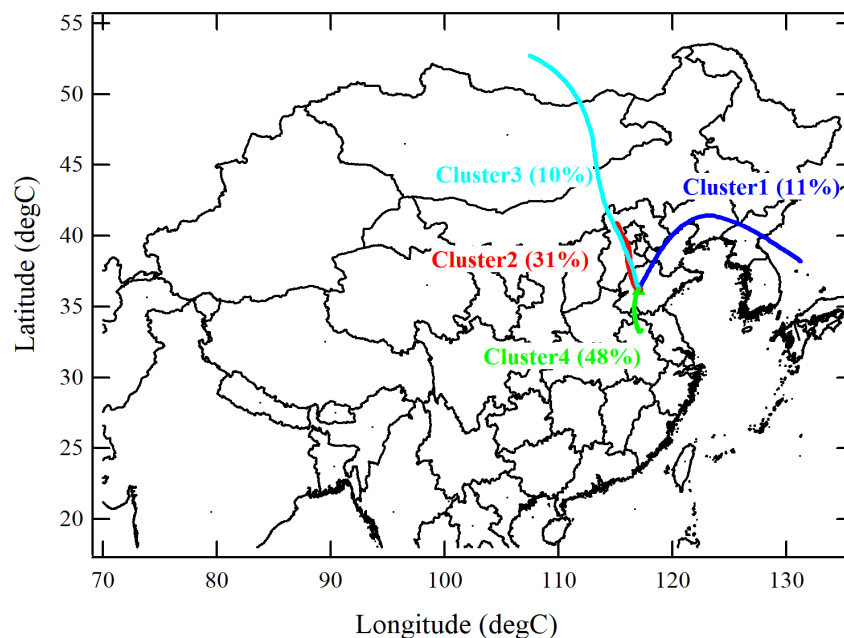


Figure S1. Three-day back-trajectories for Mt. Tai during the sampling period (green triangle: Mt. Tai).

6. P4 L111-112: *was only an important source.....*

Response: We have added “an” in the revised manuscript as follows:

Additionally, Zhu et al. (2018) have clearly shown that biomass burning was only an important source during the first half of the sampling period (June 4 – 19).

(Page 4, Line 128, Page 5, Line 129)

7. P5 L150-156: *this paragraph is a little bit redundant with the last paragraph of the Introduction (P3 L73-79). I suggest the authors may remove this paragraph.*

Response: According to the reviewer suggestion, we have removed this paragraph.

(Page 6, Line 179-185)

8. P6 L158: *replace “oxidations” by “oxidant”*

Response: We have replaced “oxidations” by “oxidant” in the revised manuscript as follows:

Due to the key role of radical and non-radical oxidant in the formation processes of secondary aerosol constituents, their concentration variations and corresponding reasons are investigated.

(Page 6, Line 187-188)

9. P6 L 168: *non-radical oxidant concentrations...*

Response: We have added “oxidant” in the revised manuscript as follows:

To our knowledge, this is the first detailed multiphase chemical modeling study examining radical and non-radical oxidant concentrations along the trajectory to the Mt. Tai under day vs. night and cloud vs. non-cloud cases.

(Page 7, Line 197-198)

10. P6 L 175: *I would suggest the authors to delete the citation here as it is only modeling results from this study.*

Response: The citations have been deleted.

(Page 7, Line 204-206)

*Section 3.2: I suggest to provide the sub-titles for “Sulfate” (e.g., 3.2.1), “Nitrate” and “Ammonium”.*

Response: We have added sub-titles for “Sulfate”, “Nitrate” and “Ammonium” subsections as follows:

3.2.1 Sulfate

(Page 9, Line 282)

### 3.2.2 Nitrate

(Page 10, Line 305)

### 3.2.3 Ammonium

(Page 11, Line 331)

11. P9 L253: replace “dominated” by “dominant”

Response: We have replaced “dominated” by “dominant” in the revised manuscript as follows:

In the nighttime cloud, aqueous-phase reaction of  $\text{HSO}_3^-$  with  $\text{H}_2\text{O}_2$  (42 %), and aqueous reaction of bisulfite with  $\text{O}_3$  (28 %) are dominant pathways for sulfate formation.

(Page 10, Line 295-297)

12. P10, L286-287 “Potential reasons are discussed below”: it is not clear where the potential reasons are discussed. Please clarify.

Response: We have changed the sentence in the revised manuscript as follows:

Potential reasons are discussed in Sect. 3.3.2.

(Page 11, Line 329-330)

13. P11 L 321: replace “shows” by “show”

Response: We have replaced “shows” by “show” in the revised manuscript as follows:

In the C2w case, Gly and MGly concentration patterns show a substantial uptake into cloud droplets.

(Page 12, Line 366)

14. P11 L348-349: I suggest to move this sentence to the beginning of this section, i.e., L319.

Response: According to the reviewer suggestion, we have moved the sentence.

(Page 12, Line 362-364)

15. P12 L363: *under-estimation*

Response: We have changed “underestimation” to “under-estimation” in the revised manuscript as follows:

The over- and under-estimation of the measured concentrations of inorganic and organic aerosol constituents could have the following reasons.

(Page 13, Line 414-415)

16. P12 L365-366: *I presume the emission data were obtained from the emission inventory, rather than model calculations.*

Response: We have changed “model calculations” to “emission inventory” in the revised manuscript as follows:

The emission data are obtained through a new anthropogenic emission inventory in Asia, which provides monthly emissions in 2010 by sector at  $0.25^\circ \times 0.25^\circ$  resolution.

(Page 13, Line 416-418)

17. P13 L401 “*reported in in above references*”: *delete one “in”.*

Response: deleted.

(Page 15, Line 456)