

Dear scholar:

Thank you very much for your constructive comments. We have addressed them one by one below. Thank you again.

--

Regards,

Steve

YIM, Hung-Lam Steve, Ph.D.

Assistant Professor
Department of Geography and Resource Management

The Chinese University of Hong Kong, Shatin, Hong Kong

Tel: (852) 3943 6534

Fax: (852) 2603 5006

Email: steveyim@cuhk.edu.hk

GRMD@CUHK: <http://www.grm.cuhk.edu.hk/eng/>

Dear authors,

I have some short questions here:

- 1. When you refer to the emission inventory, you cited Gu and Yim et al. (2016). There are two pieces of literature in the reference: Gu Y. and Yim, S. H. L.: The air quality and health impacts of domestic trans-boundary pollution in various regions of China, Environ. Int., 97, 117–124, doi:10.1016/j.envint.2016.08.004, 2016a. Gu, Y. and Yim, S. H. L.: The air quality and health impacts of domestic trans-boundary pollution in various regions of China, , 97, 117–124 doi:10.1016/j.envint.2016.08.004, 2016b. Actually, the inventory and its limitation were very briefly described in Gu and Yim et al. (2016), and the reviewer's comments were not shown. So would you please explain in detail the inventory and its limitation? I believe this is also very important and fundamental for the manuscript. And what about the natural dust part of the PM2.5 if it is not included in the inventory at all?**

Respond: In Gu and Yim et al. (2016), the detailed emission inventory, including the source information, vertical and temporal allocation, and chemical speciation were discussed in the section 3 of supporting information (SI). Assumptions involved in the emission making were also described in the SI. For natural dust, we adapted a physical-based model to simulate mineral dust emissions, in consideration of land cover, wind speed, soil information, air density, etc.

2. **Can you get the components of the PM_{2.5}? I think there are more elements other than SO₄²⁻ /NO₃⁻ in the PM_{2.5} and the components of the PM_{2.5} may partly help to explain the seasonal variations.**

Respond: Thanks for this question. We did consider other PM_{2.5} components when analyzing the seasonal variations such as ammonium, black carbon and organic carbon. Since we aim to assess acid deposition, SO₄²⁻ and NO₃⁻ are focused.

3. **Would you please also explain in greater detail the chemistry process regarding the PM_{2.5} and its wet deposition which was claimed to be the significant part of the deposition?**

Respond: For the formation of PM_{2.5}, our air quality model involved a number of chemistry processes. In addition to primary species, PM_{2.5} could be formed from inorganic precursors, e.g. SO₂, NO_x, NH₃, through gas-phase oxidation and aqueous-phase chemistry, and can be ultimately coagulated and deposited to secondary particulates. Also, some organic precursors, including VOCs and HCs, could be oxidized by O₃ and OH to form secondary organic aerosols, which can become a major component of PM_{2.5}. Refer to Binkowski, (1999) for details.

Reference:

Binkowski, F. S.: AEROSOLS IN MODELS-3 CMAQ, [online] Available from: https://www.cmascenter.org/cmaq/science_documentation/pdf/ch10.pdf, 1999.

4. **Have you ever traced the PM_{2.5} backward to see the trajectories and the origin from a Lagrangian perspective? If you did so, do the results agree with your current conclusions?**

Respond: Thanks for your suggestion. It is true that the method of backward trajectories may to some extent explain the origin. However, the secondary particulates such as SO₄²⁻ and NO₃⁻ can form through chemical reactions that the backward trajectories can reflect these processes, especially heterogeneous reactions are critical for PM_{2.5} formation and acid depositions in East Asia. We have cited several back trajectory studies of TAP in this region, and although those studies are typically limited to short-term episodic events, indeed our results align reasonably well (Lee et al., 2013) (Lee et al., 2011).

References

- Lee, S., Ho, C.-H., Choi, Y.-S., 2011. High-PM₁₀ concentration episodes in Seoul, Korea: Background sources and related meteorological conditions. *Atmospheric Environment* 45, 7240–7247. <https://doi.org/10.1016/j.atmosenv.2011.08.071>
- Lee, S., Ho, C.-H., Lee, Y.G., Choi, H.-J., Song, C.-K., 2013. Influence of transboundary air pollutants from China on the high-PM₁₀ episode in Seoul,

Korea for the period October 16-20, 2008. *Atmospheric Environment* 77, 430–439.