

Interactive comment on “Modeling Biogenic and Anthropogenic Secondary Organic Aerosol in China” by Jianlin Hu et al.

Anonymous Referee #1

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Hu et al. present a regional modeling study for China in 2013 focusing on contributions to secondary organic aerosol (SOA). They consider some more recently recognized pathways to SOA such as heterogeneous uptake of epoxides, dicarbonyls, and oligomerization in addition to traditional semivolatile SOA. SOA is classified in terms of its parent hydrocarbon source as well as precursor in different seasons across the domain. Model predictions of OC as well as precursor gases are compared to observations in select locations at select times and the model seems to perform reasonably.

Main comments

1. MGLY SOA: This work predicts a large role for methylglyoxal (MGLY) in forming SOA (23-28% of SOA), consistent with their previous work for the eastern United States (Ying et al., 2015). How well is this supported by laboratory and/or field work? Is the

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MGLY parameterization justified given that the uptake coefficient is based on glyoxal? More recent work by Marais et al. (2016) scaled the MGLY uptake coefficient to that of glyoxal using the relative Henry's law coefficient resulting in MGLY producing less than 1% of isoprene SOA. Mechanistic modeling by Woo and McNeill (2015) also indicate MGLY is not a dominant contributor to SOA.

2. Biogenic vs anthropogenic carbon and POA vs SOA: This work's predictions of SOA indicate a significant fraction of SOA contains modern carbon as it comes from biogenic VOCs such as isoprene and monoterpenes. Total OA in the study is however dominated by POA (SOA is ~30% of total OA, Fig S5). Other recent work such as that of Zhao et al. (2016) indicates anthropogenic VOCs (specifically semivolatile POA and IVOCs) are the major contributors to SOA in China. Can the authors reconcile their results with Zhao et al.'s results? Can the authors provide any insight as to why their large modern carbon contribution is more (or less) accurate than the anthropogenic VOC hypothesis? This affects your control strategy and which VOCs you might target (ie those important for OH interactions or those with low-volatility). Are there modern/fossil carbon measurements or POA/SOA proxies that can be compared with the model?

3. While the figures are clear and nicely presented, there could be more synthesis of information in the figures. Figure 2 for example has different dates in each panel and different vertical axis limits as well. The last figure shows some synthesis by including a pie chart along with spatial distribution. Figure S2 (locations) would be best in the main manuscript. Figure 4-5 each have 24 subplots. While the information is useful and I don't recommend removing it, it would be nice to have synthesis plots too. As an example, do underestimates in any of the precursor species correlate with underestimates in OC?

Other comments

4. Recent work by Marais et al. (2016) and Lin et al. (2016) indicate IEPOX SOA

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is mainly controlled via aerosol surface area which is linked to sulfate. The author's mechanism of IEPOX uptake may capture this phenomenon and show a relationship with sulfate. Page 15, line 5 about the model not capturing the Xu et al. relationship with sulfate should be verified.

5. Page 1, line 26 indicates SOA is highest in summer, but this seems very spatially dependent with winter perhaps having higher concentrations in a more localized area. Clarify.

6. Page 4, Model description section: Are these simulations the same as used by Hu et al. 2016?

7. Page 5, line 2: What CMAQ version served as the basis for this work?

8. Page 6, 7 and for data in general, can you provide a latitude, longitude, and sampling altitude for observations? Will observational data be made available with this manuscript for future model evaluation?

9. Page 6, line 13: Is there a reference for the PAMS method?

10. Page 6, line 28: Which species in particular are you referring to in terms of good olefin performance? OLE2 was quite high.

11. Page 7, line 7: In light of potentially large vehicle contributions to isoprene mentioned here, in your work, is isoprene attributed entirely (or mostly) to biogenic sources?

12. Page: 8, line 25, regarding underestimated OC, what about potential missing SOA sources (such as IVOCs, etc)? What role may they play? See also main comment number 2.

13. Page 19, Table 1: How do ARO1 and ARO2 map to benzene, toluene, and xylene and their respective yields (as used in CMAQ v4.7 and later)? C* should be provided with the alphas.

14. Page 19, Table 1: This table indicates the aromatic alphas were increased 13%

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while the monoterpene alphas were increased 30%. Isoprene alphas were increased by 2.2x. These numbers are all consistent with the biases in high-NO_x SOA yields reported by Zhang et al. (2014). As Zhang et al. reported the bias in yield, it is the yield not alpha that should be increased which involves refitting the yield vs organic aerosol concentration data to get the new alpha and C* parameters. Scaling the alpha alone results in an upper bound correction. The wall loss corrections have also been shown to be highly chamber specific (for example, Zhang et al. report two different toluene yield factors: 2.2 (their work) and 1.13 (another study)). Are the original parameterizations and the correction values from Zhang et al. from the same group/chamber? TERP yields in the original formulation match Carlton et al. (2010) and thus were a weighted contribution from different monoterpenes in the work of Griffin et al. (1999). Zhang et al. α -pinene+OH matches work from Chhabra et al. (2011). I suspect performing the proper correction to yield curves is unlikely to significantly change conclusions, but we should avoid propagating incorrect values.

15. Page 22, Table 4, Simulation 6: Clarify that anthropogenic VOC, NO_x, SO₂, etc were removed (not just VOC, NO_x)

16. What is the major driver for how anthropogenic emissions affect SOA? Is it through POA?

References

Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. D., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model representation of secondary organic aerosol in CMAQv4.7, *Environ. Sci. Technol.*, 44, 8553-8560, 10.1021/es100636q, 2010.

Griffin, R. J., Cocker, D. R., Flagan, R. C., and Seinfeld, J. H.: Organic aerosol formation from the oxidation of biogenic hydrocarbons, *J. Geophys. Res.*, 104, 3555-3567, 1999.

Lin, G., Penner, J. E., and Zhou, C.: How will SOA change in the future?, *Geophys.*

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Res. Lett., 43, 1718-1726, 10.1002/2015GL067137, 2016.

Marais, E. A., Jacob, D. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Krechmer, J., Zhu, L., Kim, P. S., Miller, C. C., Fisher, J. A., Travis, K., Yu, K., Hanisco, T. F., Wolfe, G. M., Arkinson, H. L., Pye, H. O. T., Froyd, K. D., Liao, J., and McNeill, V. F.: Aqueous-phase mechanism for secondary organic aerosol formation from isoprene: application to the southeast United States and co-benefit of SO₂ emission controls, *Atmos. Chem. Phys.*, 16, 1603-1618, 10.5194/acp-16-1603-2016, 2016.

Woo, J. L., and McNeill, V. F.: simpleGAMMA v1.0 – a reduced model of secondary organic aerosol formation in the aqueous aerosol phase (aaSOA), *Geosci. Model Dev.*, 8, 1821-1829, 10.5194/gmd-8-1821-2015, 2015.

Ying, Q., Li, J., and Kota, S. H.: Significant contributions of isoprene to summertime secondary organic aerosol in eastern United States, *Environ. Sci. Technol.*, 49, 7834-7842, 10.1021/acs.est.5b02514, 2015.

Zhang, X., Cappa, C. D., Jathar, S. H., McVay, R. C., Ensberg, J. J., Kleeman, M. J., and Seinfeld, J. H.: Influence of vapor wall loss in laboratory chambers on yields of secondary organic aerosol, *Proc. Natl. Acad. Sci. U. S. A.*, 111, 5802-5807, 10.1073/pnas.1404727111, 2014.

Zhao, B., Wang, S., Donahue, N. M., Jathar, S. H., Huang, X., Wu, W., Hao, J., and Robinson, A. L.: Quantifying the effect of organic aerosol aging and intermediate-volatility emissions on regional-scale aerosol pollution in China, *Sci. Rep.*, 6, 28815, 10.1038/srep28815, 2016.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-687, 2016.

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