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Interactive comment

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

Anonymous Referee #2

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This manuscript presents in detail the predicted secondary organic aerosol in China by a regional CTM model. Model performance is evaluated by comparing the predicted VOC, EC, and OC concentrations to the observations at several urban sites. In general, the paper is well-written. However, it is well known that SOA is complex and the parameterization of various pathways is highly uncertain. There are limited discussion and sensitivity tests on the uncertainty of the presented pathways as well as the potential contribution of unaccounted pathways. The results of the paper are also lack of observational constraints. I suggest the following comments to be considered for revision.

Specific comments:

(1) Uncertainty of SOA pathways:

– GLY/MGLY uptake is found to a major contributor to SOA in this study. The simulation is however based on irreversible uptake, whilst some studies have shown the process may also be reversible (Fu et al., 2009). Large uncertainties remain in the uptake mechanism (Galloway et al., 2011). A short summary of laboratory findings is needed along with the discussion about the potential impact on the model predictions.

– Isoprene seems extremely important given the predictions (contributing 61% in the summertime). The predicted isoprene however doesn't match well with the observations (seems being over-predicted). The fact that better agreements were found for MACR and MVK makes the case more complicated (i.e., possibly underestimation of OH). I think it is important to confirm that the predicted isoprene peak is not due to errors in the model. Either comparison to other locations with the same model or other model predictions using the same version of MEGAN is needed.

– The SOA predictions are lack of observational constraints. Intensive work has been done in major cities in China for example by AMS, which provides details about various OA types and estimates of the oxidation state (Hu et al., 2016; Huang et al., 2013; Sun et al., 2016). There are also off-line filter measurements, e.g., Yang et al. (2016) and He et al. (2014) for SOA tracers in China. Direct comparisons to such observations may be difficult to make, but rough comparisons may provide constraints to tell if the model predictions make sense, and if different studies are consistent. For example, the oxidation state of the OA as well as the organosulfate concentration may not support a high contribution of oligomers as well as GLY/MGLY/IEPOX SOA.

– How could the unaccounted processes change the predictions? For example, the conversion between POA to SOA (Robinson et al., 2007), the gaseous (Donahue et al., 2012) and heterogeneous aging of SOA (Kroll et al., 2015).

(2) Page 4, Line 30-32: The isoprene SOA yield does not vary much for dry or wet conditions (Carlton et al., 2009) as well as solid or liquid seed particles (Kuwata et al., 2012). Acidity is a key factor that regulate the production of SOA from isoprene (Kuwata

et al., 2015). In fact, the yields from Kroll et al. (2006) along with other chamber studies represent both contributions of traditional partitioning and reactive uptake, although the contribution of reactive uptake might be much smaller compared to acidic conditions.

(3) Page 5, Line 7: Which version of MEIC inventory (or the base year) has been used? Isn't MEIC only for the mainland China but not for the surrounding areas in East and Southeast Asia?

(4) Descriptions about the observations are needed in Section 2, particularly for unpublished data. In Sect. 3.1.1., does the VOC data come from GC-MS measurements? What is the time resolution? Have the data been processed? For the EC/OC data, what is the time resolution? Are the data from different studies obtained and processed in the same way?

(5) Page 6, Line 24: What kind of measurement errors?

(6) Page 8, Line 13-15: This is somewhat misleading. EC can't represent all primary emissions.

(7) Page 8, Line 30-35: It is well known that POC themselves can be semi-volatile and form SOA quickly (Robinson et al., 2007). I think the data are too limited to achieve a conclusion that POC or OC is well estimated by the model.

(8) Page 9, Line 6-7: Please clarify whether the open biomass burning emissions contribute directly more SOA precursors or more POC that causes more gas-to-particle condensation.

(9) Page 10, Line 10-15: It is also because the aging of SOA is not considered in the model.

Technical Remarks:

Page 10, Line 10: "sptial" should be "spatial".

Reference:

Carlton, A. G., Wiedinmyer, C., and Kroll, J. H.: A review of secondary organic aerosol (SOA) formation from isoprene, Atmos. Chem. Phys., 9, 4987-5005, 2009.

Donahue, N. M., Henry, K. M., Mentel, T. F., Kiendler-Scharr, A., Spindler, C., Bohn, B., Brauers, T., Dorn, H. P., Fuchs, H., Tillmann, R., Wahner, A., Saathoff, H., Naumann, K. H., Mohler, O., Leisner, T., Muller, L., Reinnig, M. C., Hoffmann, T., Salo, K., Hallquist, M., Frosch, M., Bilde, M., Tritscher, T., Barmet, P., Praplan, A. P., DeCarlo, P. F., Dommen, J., Prevot, A. S. H., and Baltensperger, U.: Aging of biogenic secondary organic aerosol via gas-phase OH radical reactions, Proc. Natl. Acad. Sci. U. S. A., 109, 13503-13508, 10.1073/pnas.1115186109, 2012.

Fu, T. M., Jacob, D. J., and Heald, C. L.: Aqueous-phase reactive uptake of dicarbonyls as a source of organic aerosol over eastern North America, Atmos. Environ., 43, 1814-1822, 10.1016/j.atmosenv.2008.12.029, 2009.

Galloway, M. M., Loza, C. L., Chhabra, P. S., Chan, A. W. H., Yee, L. D., Seinfeld, J. H., and Keutsch, F. N.: Analysis of photochemical and dark glyoxal uptake: Implications for SOA formation, Geophys. Res. Lett., 38, L17811, 10.1029/2011gl048514, 2011.

He, Q. F., Ding, X., Wang, X. M., Yu, J. Z., Fu, X. X., Liu, T. Y., Zhang, Z., Xue, J., Chen, D. H., Zhong, L. J., and Donahue, N. M.: Organosulfates from pinene and isoprene over the Pearl River Delta, South China: Seasonal variation and implication in formation mechanisms, Environ. Sci. Technol., 48, 9236-9245, 10.1021/es501299v, 2014.

Hu, W. W., Hu, M., Hu, W., Jimenez, J. L., Yuan, B., Chen, W. T., Wang, M., Wu, Y. S., Chen, C., Wang, Z. B., Peng, J. F., Zeng, L. M., and Shao, M.: Chemical composition, sources, and aging process of submicron aerosols in Beijing: Contrast between summer and winter, J. Geophys. Res., 121, 1955-1977, 10.1002/2015jd024020, 2016.

Huang, X. F., Xue, L., Tian, X. D., Shao, W. W., Sun, T. L., Gong, Z. H., Ju, W. W., Jiang, B., Hu, M., and He, L. Y.: Highly time-resolved carbonaceous aerosol characterization

in Yangtze River Delta of China: Composition, mixing state and secondary formation, Atmos. Environ., 64, 200-207, 10.1016/j.atmosenv.2012.09.059, 2013.

Kroll, J. H., Lim, C. Y., Kessler, S. H., and Wilson, K. R.: Heterogeneous oxidation of atmospheric organic aerosol: Kinetics of changes to the amount and oxidation state of particle-phase organic carbon, J. Phys. Chem. A, 119, 10767-10783, 10.1021/acs.jpca.5b06946, 2015.

Kuwata, M., Zorn, S. R., and Martin, S. T.: Using elemental ratios to predict the density of organic material composed of carbon, hydrogen, and oxygen, Environ. Sci. Technol., 46, 787-794, 10.1021/es202525q, 2012.

Kuwata, M., Liu, Y., McKinney, K. A., and Martin, S. T.: Physical state and acidity of inorganic sulfate can regulate the production of secondary organic material from isoprene photooxidation products, Phys. Chem. Chem. Phys., 17, 5670-5678, 10.1039/C4CP04942J, 2015.

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J. R., and Pandis, S. N.: Rethinking organic aerosols: Semivolatile emissions and photochemical aging, Science, 315, 1259-1262, 10.1126/science.1133061, 2007.

Sun, Y. L., Du, W., Fu, P. Q., Wang, Q. Q., Li, J., Ge, X. L., Zhang, Q., Zhu, C. M., Ren, L. J., Xu, W. Q., Zhao, J., Han, T. T., Worsnop, D. R., and Wang, Z. F.: Primary and secondary aerosols in Beijing in winter: sources, variations and processes, Atmos. Chem. Phys., 16, 8309-8329, 10.5194/acp-16-8309-2016, 2016.

Yang, F., Kawamura, K., Chen, J., Ho, K. F., Lee, S. C., Gao, Y., Cui, L., Wang, T. G., and Fu, P. Q.: Anthropogenic and biogenic organic compounds in summertime fine aerosols (PM2.5) in Beijing, China, Atmos. Environ., 124, 166-175, 10.1016/j.atmosenv.2015.08.095, 2016.

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

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