

We thank the reviewers for the effort to review the manuscript and to provide constructive comments and good suggestions to improve our manuscript. Our replies to the comments and our actions taken to revise the paper (in blue) are given below (the original comments are copied here).

The modifications corresponding to the comments are labeled **in red color and highlighted**.

The language and grammar in the revised manuscript have been edited carefully and polished native English speakers by according to the reviewers' comments. (labeled in red color)

Referees' comments:

Referee #2

This manuscript presented a significant effort to couple OA formation pathways and microphysical processes to a global and regional chemical transport model, with the goal of simulating the impacts of OA physics/chemistry to particle number size distribution and mixed particle composition. The work coupled a 1.5-D VBS module and the APM microphysics module to the IAP-AACM chemical transport module. The authors also presented some preliminary comparisons with the observations, and overall the model appeared able to capture the global OA concentrations and the CN10 concentrations. The amount of work done was impressive, and the methods were mostly valid and up-to-date. Overall, I think the paper may be published after clarifying some missing details and improving the figure representation.

Reply: Thanks to the reviewer for the great effort to review our manuscript and to provide constructive suggestions to improve our manuscript.

Section 2.1 Host model: What meteorological data is the IAP-AACM driven by? I see in lines 342 to 350 that the model was driven by meteorological parameters from WRF, but maybe that information can be moved here. Also, how were global meteorological fields obtained from WRF? Did the authors run Global WRF? If so,

then additional references for Global WRF should be included, e.g., Zhang et al. (2012). What was the spatial resolution of the meteorological fields, and was interpolation used? What was the temporal resolution of meteorological fields? I.e., how often were the meteorological fields updated. How was the nudging performed in the three nested domains?

Reply: The IAP-AACM+APM was driven by the global WRF. The essential reference (Zhang et al., 2012) is added in the revised manuscript. The IAP-AACM used the same domain and horizontal grid (i.e., 1° for D01, 0.33° for D02, and 0.11° for D03) as for the global WRF; thus, only vertical interpolation of the meteorological fields of the global WRF was performed to drive the IAP-AACM+APM. The meteorological fields were updated hourly in IAP-AACM+APM. In the first domain, a nudging coefficient of 0.0003 was used in all vertical layers; in the second and third domain, the same nudging coefficient was used in vertical layers except those in boundary layer, where nudging was not used. These necessary description has been added in the revised manuscript (seen in Line 193-194, 360-363, and 367-370).

Zhang, Y., Hemperly, J., Meskhidze, N., and Skamarock, W. C.: The Global Weather Research and Forecasting (GWRf) Model: Model Evaluation, Sensitivity Study, and Future Year Simulation, Atmospheric and Climate Sciences, 02, 231-253, 10.4236/acs.2012.23024, 2012.

Section 2.3 VBS module: I would like to see the model's representation of the relationship between oxidation state and volatility expressed more clearly. Did the authors simply move the oxidation products of POA and IVOC into a volatility bin that is one magnitude lower? What about the fragmented products during the oxidation, i.e., the smaller molecular weight products?

Reply: Considering a single oxidation step would hardly provide enough carbon number reduction required to move the oxidation products of POA and IVOC into a volatility bin that is one magnitude lower. In the 1.5D VBS module (Koo et al., 2014; Yang et al., 2019) used in our study, the POA aging process is approximated by using a "partial conversion" to OOA: Oxidation products of POA are represented as a mixture of POA and OPOA in the next lower volatility bins. For IVOC, lower SOA mass yields are assumed to consider this process. Fragmentation in the VBS module is

implicitly considered through reduction in carbon number of the oxidation products. NO_x-dependent product mass yields from oxidation of hydrocarbon precursors were determined based on smog chamber data (Murphy and Pandis, 2009; Hildebrandt et al., 2009). The necessary information has been added in the revised manuscript (seen in Line 278-281, 285-287, and 296-299).

Murphy, B., and Pandis, S.: Simulating the Formation of Semivolatile Primary and Secondary Organic Aerosol in a Regional Chemical Transport Model, Environmental science & technology, 43, 4722-4728, 2009.

Hildebrandt, L., Donahue, N.M., Pandis, S.N.: High formation of secondary organic aerosol from the photo-oxidation of toluene. Atmos. Chem. Phys. 9, 2973-2986, 2009.

Koo, B., Knipping, E., and Yarwood, G.: 1.5-Dimensional volatility basis set approach for modeling organic aerosol in CAMx and CMAQ, Atmospheric Environment, 95, 158–164, 10.1016/j.atmosenv.2014.06.031, 2014.

Yang, W., Li, J., Wang, W., Li, J., Ge, M.-F., Sun, Y., Chen, G., Ge, B., Tong, S., Wang, Q., and Wang, Z.: Investigating secondary organic aerosol formation pathways in China during 2014, Atmospheric Environment, 213, 10.1016/j.atmosenv.2019.05.057, 2019.

Section 2.3 VBS module: Also, a recent paper (Jo et al., 2019) indicated that the VBS representation of SOA formation from isoprene is incorrect because the reactive uptake pathway dominates SOA formation from isoprene. Please discuss this point, the lack of reactive update pathways in this model, and the implication for the present model results.

Reply: Yes, based on the comparison of full-chemistry calculation and VBS simulation, the VBS representation could not capture the physicochemical dependencies of SOA formation on dominant pathway from isoprene and VBS may underestimate the biogenic SOA from isoprene (Jo et al., 2019). Nevertheless, the biogenic SOA could not explain the SOA concentration (Spracklen et al. 2011; Matsui et al., 2014; Lin et al., 2016). Therefore, the important roles of anthropogenic SOA revealed in our study still hold true and the major conclusions would not change. However, the fixed parameters in VBS make it difficult to represent the real formation pathway of SOA and capture the response of SOA to emission changes. More accurate parameters considering the key physicochemical dependencies should be incorporated to update the VBS module in our model. This is critical to (1) accurately quantify the contribution of biogenic and anthropogenic sources to OA, (2) evaluate the effectiveness of control measures to reduce OA concentration in order to improve

air quality, and (3) explore the aerosol-climate-vegetation interactions. Thank the reviewer for this valuable comment and notice for our future work. The necessary discussions have been added in our revised manuscript (seen in Line 543-547 and 821-825).

Jo, D. S., Hodzic, A., Emmons, L. K., Marais, E. A., Peng, Z., Nault, B. A., Hu, W. W., Campuzano-Jost, P., and Jimenez, J. L.: A simplified parameterization of isoprene-epoxydiol-derived secondary organic aerosol (IEPOX-SOA) for global chemistry and climate models: a case study with GEOS-Chem v11-02-rc, Geoscientific Model Development, 12, 2983-3000, 10.5194/gmd-12-2983-2019, 2019.

Lines 359-360: "In the LV_POA and HV_POA experiment, quartiles of the abovementioned distribution factors are used". Not sure what this meant. Looking at Table 2, I do not see the use of 'quartiles of the above-mentioned factors'. What different factors were used in the LV_POA and HV_POA experiments, respectively?

Reply: The different factors used in the LV_POA and HV_POA experiments were provided in the "Volatility distribution" column in Table 2. The quartiles of POA volatility factor are taken from May et al. (2003a, b, c). Modification were made in Line 383-384.