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 <mark>in red color and highlighted</mark>.

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 for wind, temperature, and water vapor was used in all vertical layers; in the second and third domain, the same nudging scheme 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 192-194, 362-365, and 369-372).

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. NOx-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 298-301).

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 over China (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 parameterizations 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 568-571 and 852-856).

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) and Robinson et al. (2007). Modification are made in Line 390-393.

Table 2 and related text on the design of the sensitivity experiment: overall, I think the sensitivity experiment could be explained more clearly. I was not able to understand what was the goals of the sensitivity experiments and how those goals relate to the parameters in Table 2.

Reply: Because the size distribution of primarily emitted particles and the volatility distribution of POA have substantial impacts on the simulation of particle number concentration over areas influenced by anthropogenic sources, the sensitivity experiments were designed to investigate the impacts of these factors on our study results. The description of sensitivity experiments and their relations with the parameters in Table 2 are described more clearly in the revised manuscript (seen in Sect.3.2).

Lines 382- 383: "...the China Atmosphere Watch Network... Zhang et al. (2008)": What year(s) were the measurements? The writing of this sentence seemed to suggest that the measurements were from 2010, which cannot be possible.

Reply: Yes, the year of OC measurements in China was 2006. "2010" is changed to "2006" in the revised manuscript (seen in Line 414).

Lines 437 to 439: "The number concentration of particles from 100 nm to 1000 nm ...correlation coefficient being 0.70.': Figure 2 uses a different unit for particle size (micrometer), and I do not see the diameter extending to 1000 nm. Also, how were the normalized bias and the correlation coefficient calculated? Did the authors calculated only the bias and correlation for the time series of the total number concentration (which is not shown)? Or did they calculated a mean bias and correlation for the entire PNSD spectrum? If the latter, how was this done, and did the statistics entail a preferential weighting of the smaller particles?

Reply: Yes, the shaded figure (Fig.2) did not extending to 1000 nm. Because the size bins of observation and the model are different, the particle number size distribution (PNSD) of observation is mapped to the size bins of the model when comparing the particle number size distribution in Fig.2. The number concentration of particles from 100 nm to 1000 are calculated by adding the number concentration in the size bin from 100 nm to 1000 nm. Due to the limitation of detection size range (15-685nm) of the measurement, the diameter shown in Fig.2 was not extended to 1000 nm. In the community, "100 nm to 1000 nm" is commonly used as a proxy of accumulation mode and the particles from 100 nm to 685 nm can account for the most of particles number concentration from 100 nm to 1000 nm. Due to these reasons, "100 nm to 1000 nm" was used in our study. However, this expression is inaccurate. In the revised manuscript, "100 nm to 1000 nm" are changed to the size range of measurements and the comparison method of simulated particle number concentration with observations is expressed clearly (seen in Line 412-414 and 477).

Figure 4: The measurements were too small and unreadable in this figure. Please enlarge and circle with a black or white outline.

Reply: The circles are made clear with a black outline in the revised manuscript (seen in Fig.4).

Figure 4: Also, the OC measurements in China all appeared to be much, much higher

than the simulated concentrations. This is inconsistent with what was shown in Figure 1, where the authors indicated that the model was able to represent the observed OA concentrations in the one site in Beijing. Please resolve this inconsistency or provide more discussion in the text. The discrepancy between the measurements and the simulated concentrations in Figure 4c is large enough that, I do not think the difference in the year could explain it. Also, the symbols were too small to read.

Reply: The emission data for 2010 simulation (Fig.4) was different from the emission used in the simulation during the period from August 22 to September 30, 2015. 2015 case used the multi-resolution emission inventory for China in 2015 (http://www.meicmodel.org). 2010 simulation used a publicly available datasets for 2010 (https://edgar.jrc.ec.europa.eu/htap_v2/index.php). The difference of BC and OC emission between 2010 and 2015 over China is displayed in Fig.R1. Compared with the emission in 2010, a reduced emission of BC and OC in main source regions can be seen in 2015 over China. In our study, the total OC emission in 2010 over China was 3.54 Tg C yr^{-1}. Using the GEOS-Chem and a emission of 3.95 Tg C yr^{-1} for OC, Fu et al. (2012) found the model underestimated OC at most observation sites, particularly in January in 2006. Their top-down estimation for total OC emission was 6.67 Tg C yr^{-1}, greatly higher than the OC emission in our study. Studies indicated that the uncertainties of BC and OC emission can be higher than 200% (Zhao et al., 2013; Li et al., 2015). We agree that the underestimation of OC in 2010 could not be attributed only to the uncertainties in emission. However, the difference of emission is a major factor responsible for the different model performance in simulating OC concentration between 2010 and 2015. According to the suggestions of the reviewer, some discussions on the inconsistent model performance between 2010 and 2015 are added (seen in Line 545-549). In addition, the explanations for OC underestimation are presented (seen in Line 536-545). The symbols in Fig.4 are made clear to read.
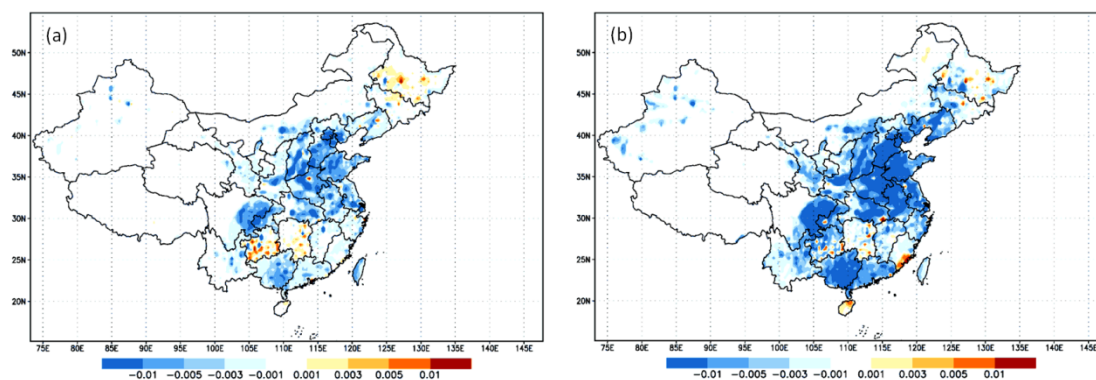
Fig.R1 Differences of (a) BC and (b) OC emission rates between 2010 and 2015
(2015-2010)

Fu, T. M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., Qu, W. J., Han, Z., Zhang, R., Wang, Y. X., Chen, D., and Henze, D. K.: Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution, Atmospheric Chemistry and Physics, 12, 2725-2746, 10.5194/acp-12-2725-2012, 2012.

Li, M., Liu, H., Geng, G. N., Hong, C. P., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H. Y., Man, H. Y., Zhang, Q., and He, K. B.: Anthropogenic emission inventories in China: a review, Natl Sci Rev, 4, 834-866, 10.1093/nsr/nwx150, 2017.

Zhao, Y., Zhang, J., and Nielsen, C. P.: The effects of recent control policies on trends in emissions of anthropogenic atmospheric pollutants and CO2 in China, Atmospheric Chemistry and Physics, 13, 487-508, 10.5194/acp-13-487-2013, 2013.

Zhang, X. Y., Wang, J. Z., Wang, Y. Q., Liu, H. L., Sun, J. Y., and Zhang, Y. M.: Changes in chemical components of aerosol particles in different haze regions in China from 2006 to 2013 and contribution of meteorological factors, Atmospheric Chemistry and Physics, 15, 12935-12952, 10.5194/acp-15-12935-2015, 2015.

Lines 499-500: "Overall, the model explained most of the observations." I really did not see this in Figure 4c. Please revise and provide an estimate of the bias.

Reply: Yes, the model underestimated the OC concentration over China. Nevertheless, the spatial variations of OC at different observation sites, particularly the west-east gradient, were well reproduced. The OC concentration in the United States were also reasonably simulated (Fig.4a). According to the comment, the inappropriate expressions are modified (seen in Line 531-534). The underestimation of OC at sites in China can be explained by the following reasons: (1) the underestimation of OC emission; (2) representativeness difference between observation and simulation results; (3) uncertainties in model mechanism calculating OC concentration.

The study of Zhao et al. (2012) and Li et al. (2017) pointed out that the emission of OC over China did not change much. The difference of OC concentration between 2006 and 2010 is also small (Zhang et al., 2015). The OC emission (3.54 Tg C yr^{-1}) in our study is 47% lower than the estimated emission (6.67 Tg C yr^{-1}) in Fu et al.

(2012). In addition, in the VBS module used in our study, POA emissions were distributed to the five volatility bins, which can lead to an underestimation of OC and OA from POA emission (Donahue et al., 2009). Following the recommendation of existing study (Tsimpidi et al., 2010; Shrivastava et al., 2011), we did the experiment increasing the existing POA emission by a factor of 3. Although the underestimation of OC in China was reduced, the OC concentrations in the United States were overestimated (Fig.R3). Considering the substantial contribution of intermediate volatility organic compounds (IVOC) to SOA and OA (Zhao et al., 2016; Yang et al., 2019), and the underestimation of SOC fraction in China, the IVOC emissions used in our study may be underestimated. Based on these facts, the underestimation of OC emission over China should largely be responsible for the underestimated OC in China.

However, the underestimation of SOC fraction also indicates to the deficiency of SOA formation mechanism. Although studies (Zhao et al., 2016; Yang et al., 2019) have suggested that IVOCs have a large contribution to SOA and OA over China, the emissions of IVOCs were not included in the traditional emission inventory, which make it difficult to estimate SOA from IVOCs. The underestimation of their emission and production yields to SOA can lead to considerable underestimation of SOA and OA. Aqueous-phase formation processes of SOA have an evident influence on the particle properties and total SOA mass (Ervens et al., 2011). Nevertheless, aqueous-phase processes of SOA was not included and it may cause an underestimation of SOA and OA.

According to the reviewer's comment, the reasonable description, necessary discussion, and the model bias were added in the revised manuscript (seen in Line 532-534, 545-549, and 586-590).
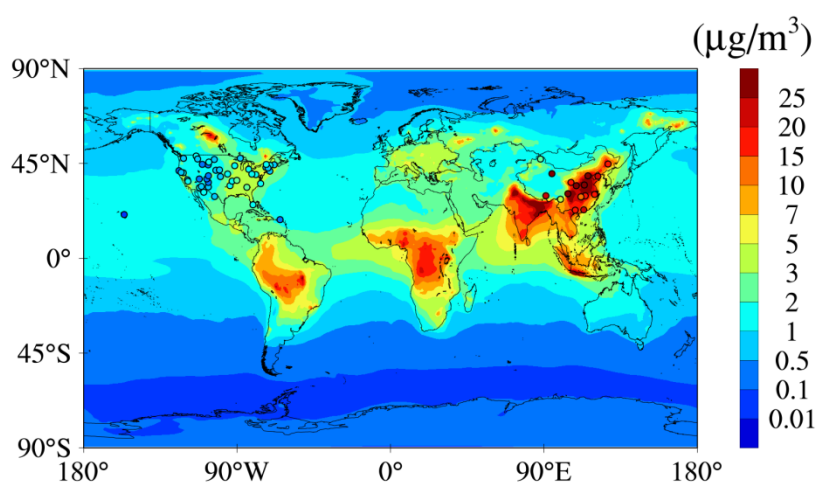

Fig.R2 Surface layer horizontal spatial distributions of organic carbon concentrations. The shaded circles denote the observed concentrations.

Donahue, N., Robinson, A., and Pandis, S.: Atmospheric organic particulate matter: From smoke to secondary organic aerosol, Atmospheric Environment, 43,

94-106, 10.1016/j.atmosenv.2008.09.055, 2009.

Ervens, B., Turpin, B. J., and Weber, R. J.: Secondary organic aerosol formation in cloud droplets and aqueous particles (aqSOA): a review of laboratory, field and model studies, Atmos. Chem. Phys., 11, 11069–11102, https://doi.org/10.5194/acp-11-11069-2011, 2011.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmospheric Chemistry and Physics, 11, 6639-6662, 10.5194/acp-11-6639-2011, 2011.

Tsimpidi, A. P., Karydis, V. A., Zavala, M., Lei, W., Molina, L., Ulbrich, I. M., Jimenez, J. L., and Pandis, S. N.: Evaluation of the volatility basis-set approach for the simulation of organic aerosol formation in the Mexico City metropolitan area, Atmospheric Chemistry and Physics, 10, 525-546, DOI 10.5194/acp-10-525-2010, 2010.

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.

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

Figure 4: Are there also observational constraints on what fraction of the measured OC was secondary, e.g. using the EC-tracer method? I think this was in Zhang et al. (2008) and can be shown in Figure 4d for comparison.

Reply: SOC fractions in Zhang et al. (2008) are shown in Fig.4d.

Lines 509-511: "However, our simulations show in Fig. 5 ... large anthropogenic emissions." How does this statement relate to, or can be used to explain the finding in Fig 4, i.e., the model severely underestimated the observed OC, particularly over

China? Lines 518-519: "In the second ... over China": How does this statement relate to, or can be used to explain the finding in Fig 4, i.e., the model severely underestimated the observed OC, particularly over China?

Reply: In the case of lower simulated OC concentration than observation, the underestimation of SOC in major anthropogenic source regions indicated that the model underestimated the formation of anthropogenic SOA (ASOA) in these regions. Underestimation of AOA should be responsible for the underestimation of OA over China. The exact contribution of ASOA to SOA would be greater than estimated in our simulation. The importance of ASOA is still true though our model underestimated the absolute concentration of OC, particularly over China. However, the exact contribution of ASOA should be refined by incorporating the precise emission including non-traditional emission and improving the representation of SOA formation in the VBS module in our future study. The relations between the underestimation of OC and the higher concentration of AOA than BOA are added and the influences of the OC underestimation, particularly over China, are added in our revised manuscript (seen in Line 586-590).

Figure 7: Again, all of the symbols for the observed values were way too small and unreadable. Please revise.

Reply: revised (seen in Fig.7).

Figure 9 caption, last line: "over the first domain (top panel) and second domain (bottom panel)": should be 'left panel' and 'right panel', respectively.

Reply: revised (seen in line 909).

Reference Jo et al. (2019), Geosci. Model Dev., 12, 2983-3000.