

## Response to Referee #1

The authors thank the reviewer for the comments that improve the quality of the paper. The detailed responses are given as follows. The reviewer comments are shown in italic font, the responses are in regular font, and the revised text is in bold font.

*The authors present model results using the WRF/CMAQ for SOA formation in China. The model is updated for the partitioning of water vapor to the organic aerosol and the non-ideality of the organic phase. A comparison of the model to observations is performed for multiple sites. SOA enhancement during the summer and winter is discussed for the different China domains. The effect of aerosol liquid water on SOA formation and aerosol optical depth are presented. Correlation of the calculated OA hygroscopicity based on the k-Köhler theory to the O:C ratio is performed to show seasonal and multicity variations. Although the publication could provide valuable insights into the factors that govern SOA formation in China it currently lacks a detailed discussion and validation of the presented results. Therefore, the publication requires major revisions suggested below.*

### Major Comments

**Comment 1:** *The manuscript is hard to read. Discussion and Results are not well separated in the manuscript. Many phrases are not clear and require more elaboration and better use of English. The table and most figures are poorly made and the science is hard to follow. Examples are given below.*

**Response 1:** We thank the reviewer for pointing this out. We have updated all the figures and tables by showing monthly-averaged results instead of the daily maximum average as used in the previous version. Also, the text has been revised carefully to make it clear and easy to follow.

**Comment 2:** *A major drawback in this work is that the model is not capturing the SOA formation during the winter that has been shown to be the dominant organic aerosol source in multiple publications for different domains of China. The authors only in a sentence discuss that the conversion of the POA to SOA may be the reason for these discrepancies but they have no observations to back this up.*

**Response 2:** We agree with the reviewer that there is no direct observational evidence for this particular episode that POA aging played a significant role in the SOA formation. However, this process is one of the important missing

sources of SOA in several models, field and chamber studies (Robinson et al., 2007; Shrivastava et al., 2008; Zhao et al., 2016; Jimenez et al., 2009; Hodzic et al., 2010; Murphy et al., 2017). Also, organic compounds with intermediate volatility (IVOCs) between SVOCs and traditional VOCs, especially from combustion sources might contribute to SOA as well (Robinson et al., 2007; Shrivastava et al., 2008; Tkacik et al., 2012; Zhao et al., 2014; Zhao et al., 2016; Hodzic et al., 2010), but with high uncertainties in the emission inventory and SOA yields (Shrivastava et al., 2008; Tkacik et al., 2012). We performed an additional simulation using the latest version of the CMAQ model (v5.3.1), which includes a parameterization of these processes. The predicted SOA indeed increases significantly (Figure R1). We further analyzed the contribution of traditional POA (as semi-volatile POA in CMAQv5.3.1), SOA from the oxidation of the semi-volatile POA, traditional SOA, and a new SOA surrogate (pcSOA) representing missing SOA from IVOC oxidation, multigenerational aging of VOC oxidation products, and underestimate of SOA yield due to chamber wall losses (Murphy et al., 2017), finding pcSOA dominates in Beijing and Guangzhou (Figure R2) as well as the whole domain (Figure R1). The averaged SOA/POA ratio in Beijing is increased to 1.53, which is more consistent with field measurements (Zhao et al., 2019; Sun et al., 2016; Sun et al., 2013). However, the emission factors and oxidation rate of pcSOA precursors are highly uncertain and the contribution of pcSOA requires more observational constrains (Murphy et al., 2017).

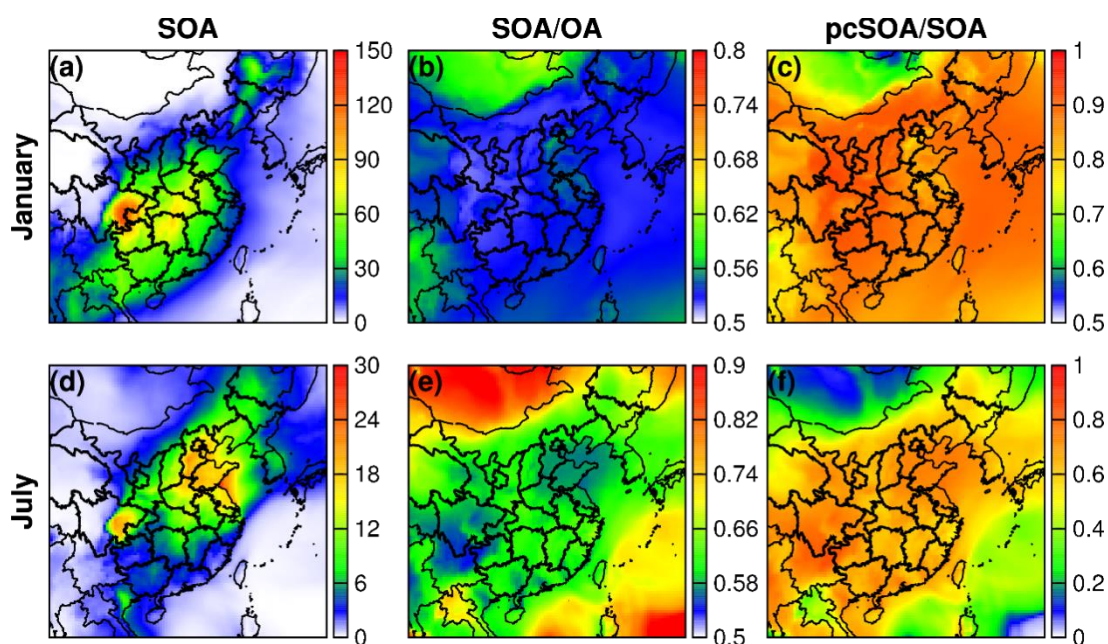
We also examined the sensitivity of SOA and organic liquid water ( $ALW_{org}$ ) to pcSOA and POA in an offline calculation in Beijing, Guangzhou, Jinan, and Nanjing. POA has the same properties as we used in the model. Non-volatile isoprene SOA is taken to represent pcSOA as their similarities in saturation vapor pressure and O:C ratio. We found that both SOA and  $ALW_{org}$  are positively correlated with pcSOA, increased by 2-5 times in different locations when pcSOA increased by 2 times. The impacts of water partitioning into OPM and non-ideality of the organic-water mixture by including the above process should be explored in a future study.

We extend the discussion of potential reasons for underestimated SOA in the revised text (L309-326):

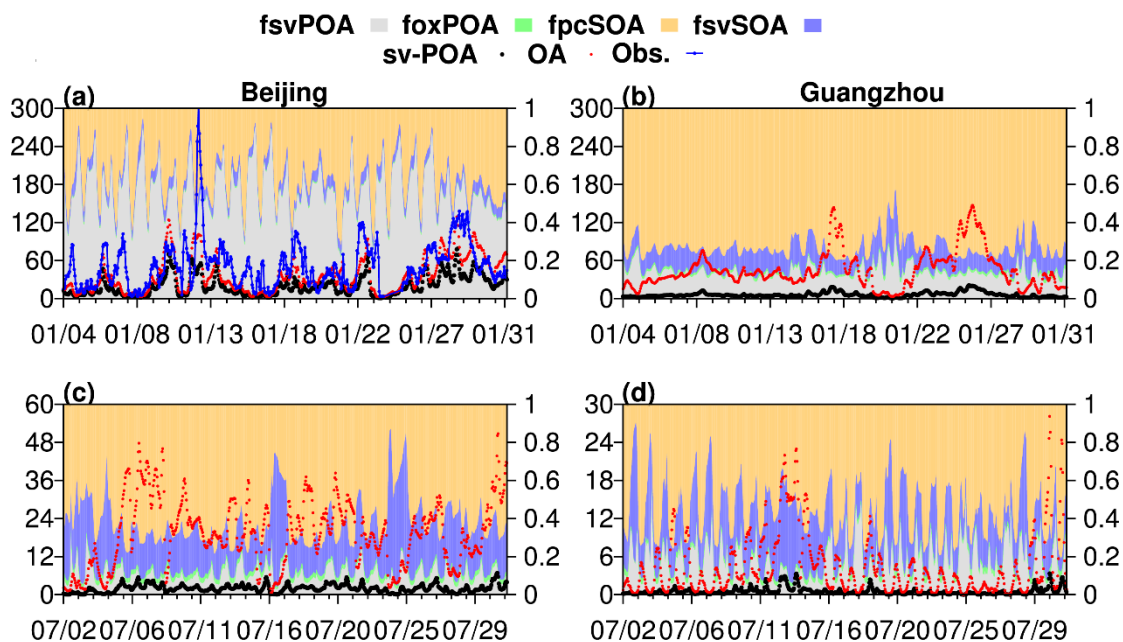
**“Again, no apparent changes of SOA nor OA are observed between case S3 and BS (not shown), since POA is predicted to be the primary contributor to OA at Beijing in winter in the current model, with an averaged SOA/POA ratio of 0.12. This ratio is much lower than the field observation of about 0.45-1.94 (Zhao et al., 2019; Sun et al., 2013; Sun et al., 2016). The bias might be due to the missing SOA converted by partitioning and aging of semi-volatile POA as well as oxidation from**

intermediate volatile organic compounds (IVOCs) and VOC oxidation products. Those pathways are shown to be important for SOA formation by modeling, field and chamber studies (Hodzic et al., 2010; Jimenez et al., 2009; Murphy et al., 2017; Robinson et al., 2007; Shrivastava et al., 2008; Tkacik et al., 2012; Zhao et al., 2014; Zhao et al., 2016a).

A sensitivity test was performed by using the newest CMAQ model version 5.3.1 that includes all the above processes in the aerosol module. The SOA/POA ratio in Beijing is improved greatly to be 1.53 in winter. However, high uncertainties still exist in the emissions of the involved precursors and characterization of SOA formation through these processes, needing further constrains by observations. Their influences on water partitioning into OPM and non-ideality of the organic-water mixture on SOA will be evaluated in a future study.”



**Figure R1.** Mean SOA, SOA/OA, and pcSOA/SOA ratio predicted during January and July of 2013 by CMAQv5.3.1.



**Figure R2.** Modeled concentration of semi-volatile POA (sv-POA), OA and fraction of each organic aerosol component fsvPOA (sv-POA), foxPOA (oxidation of sv-POA), fpcSOA (pc-SOA) and fsvSOA (traditional SOA) in Beijing (a, c) and Guangzhou (b, d). Observations of OA in January 2013 at Beijing (Obs.) are also included in (a). The left axis is for the concentrations ( $\mu\text{g m}^{-3}$ ) and the right axis is the fraction of OA components.

**Comment 3:** *There is no discussion on the influence of nitrate on aerosol liquid water. What fraction of the ALW is related to nitrate and what to organics? How could the ALW related to nitrate influence the partitioning of organics?*

**Response 3:** In the current model, we separately treated the liquid water associated with organics ( $ALW_{org}$ ) and inorganics ( $ALW_{ing}$ ) in the condensed phase. Nitrate is assumed to only affect the inorganic aerosols and  $ALW_{ing}$  as the interactions between inorganic and organic phases are not considered currently. This is the same approach used by Pankow et al. (2015). When considering the interactions between inorganic and organic aerosols in a CMAQ model, Pye et al. (2017) found an increase in SOA, which impacts are less significant than the separate treatment of the two phases. However, the interactions among condensed organics, i.e. the polarity of organics in the aerosol were ignored in their study. The interactions between inorganic and organic phases on ALW and SOA are beyond the scope of the current study and will be investigated in the future. We added a statement in the revised text (L207-L208) to make it clear:

**“In the current model, we assumed no interactions between the inorganic and organic phases.”**

**Comment 4:** *A comparison of the model to observations should be performed and presented in the main text for both seasons in more detail. The effect of the improvements performed for the SAPRC-11 model is not discussed. The processes added e.g., the heterogeneous formation of nitrate and sulfate on the particle surface, SOA from isoprene, and dicarbonyls surface-controlled reactive uptake are not discussed. What is the effect of these added processes to the overall performance of the model? Each addition and the effect should be discussed in detail in order to support the importance of including them.*

**Response 4:** The contribution of heterogeneous formation of nitrate and sulfate are not related to SOA formation. Papers documenting these changes have already been cited in the manuscript (Ying et al., 2014; Hu et al., 2016), and the impacts of the heterogeneous chemistry on nitrate and sulfate were discussed in those and another study (Zheng et al., 2015). The predicted nitrate and sulfate have been extensively compared with observations (Shi et al., 2017; Qiao et al., 2018). The improvements in the modeled SOA such as the reactive uptake of dicarbonyls and the isoprene generated epoxydiols have been discussed in previous studies by Ying et al. (2015), Li et al. (2015), Hu et al. (2017) and Liu et al. (2020) and have been shown to greatly increase the predicted SOA concentrations. The focus of this work is partitioning of water into OPM and the polarity of condensed organic compounds on SOA formation in China, which have not been examined so far. We have revised the manuscript to make it clear (L149-L161):

**“Model configurations were largely based on that used by Hu et al. (2016) as summarized below. Firstly, SAPRC-11 was expanded for a more detailed treatment of isoprene oxidation and tracking dicarbonyl products (glyoxal and methylglyoxal) from different groups of major precursors (Ying et al., 2015). Secondly, SOA from isoprene epoxydiols (IEPOX), methacrylic acid epoxide (MAE) and dicarbonyls through surface-controlled irreversible reactive uptake were added (Hu et al., 2017; Li et al., 2015; Liu et al., 2020; Ying et al., 2015). Thirdly, the heterogeneous formation of secondary nitrate and sulfate from NO<sub>2</sub> and SO<sub>2</sub> reaction on the particle surfaces (Ying et al., 2014) were added, which is an important source of secondary inorganic aerosols (Zheng et al., 2015) and improves model estimates of nitrate and sulfate (Qiao et al., 2018; Shi et al., 2017). Fourthly, SOA yields were corrected for vapor wall loss (Zhang et al., 2014). Impacts of the above updates on model performances have been extensively discussed in the cited work and will not be further investigated in the current study.”**

**Comment 5:** *MEGAN has been shown to overestimate the isoprene emissions. Would this have a major effect on SOA formation in this work?*

**Response 5:** The MEGAN model has been shown to overestimate emissions of isoprene in the eastern and southeastern US (Wang et al., 2017; Kota et al., 2015), which is mainly due to databases of emission factors used. While our previous study indeed showed up to 5 times higher isoprene concentrations compared to the observations in Nanjing, the isoprene oxidation products (MACR and MVK) agree well with observations. Another study of Southern China also showed up to 2 times higher of MEGAN compared to measured isoprene fluxes (Situ et al., 2014). Thus, it is still inconclusive whether isoprene was indeed overestimated. If this isoprene overestimation was prevalent throughout the country and the isoprene SOA changed linearly with isoprene emissions, the actual SOA concentration and impacts on SOA due to water partitioning will decrease by 40-50% and 20-30%, respectively. Emissions of isoprene and other biogenic emissions are low during winter so that little or no impact is expected for winter.

**Comment 6:** *Table 1: There is no discussion of the table in the main text. Abbreviations are not included in the caption. What is MB, GE, Num? Discussion on more statistically relevant values would be beneficial, e.g. what is the R2 of the comparisons? What are the presented values? Medians? Means? What is the domain of the model and how much do the values fluctuate around the domain? What are the uncertainties of the measurements and the model? A figure of the comparison of temperature and relative humidity of obs. vs model with the associated errors and linear regression analysis with the statistics would be informative.*

**Response 6:** Table 1 shows the mean observation (OBS), mean prediction (PRE), mean bias (MB), gross error (GE), and the number of valid data of temperature and relative humidity in 8 sub-regions of the domain as shown in Figure S1. The table has been revised with explanations of all the abbreviations. A new table (Table S5) has been added to explain each region in Table 1. Since there are too many observation sites in the domain to show the uncertainties and regression information, we added R in the revised Table 1 and updated the table. More information about measurement methodology and uncertainties associated with measurements can be found at the NCDC data website. We discussed Table 1 in the original text (L245-L249):

“Table 1 shows the comparison of WRF predictions and observations in 8 sub-regions of the domain (Figure S1). Observed data are accessible from the National Climatic Data Center at <ftp://ftp.ncdc.noaa.gov/pub/data/noaa>. Temperature and RH are well captured by WRF in YRD, the Pearl River Delta (PRD), and central regions of China (the major regions of eastern China).”

We expanded this discussion in the revised manuscript in L277-L288:

**“Temperature and RH are the two meteorological factors that affect SOA formation. Table 1 lists model statistics of mean observation (OBS), mean prediction (PRE), mean bias (MB), gross error (GE) and correlation coefficient (R) based on WRF and observations at monitoring sites located in 8 sub-regions of the domain (Figure S1) during January and July of 2013. The benchmarks for the MM5 model (another meteorology model) of 4-12km horizontal resolution suggested by Emery et al. (2001) are also listed in the table. Details of monitoring sites in the 8 sub-regions are listed in Table S5. Overall, WRF tends to underestimate both temperature and RH. The model shows better agreement with observed temperature as R is higher than that of RH. Both temperature and RH are well captured by the model in YRD, the Pearl River Delta (PRD), and the central regions of China (the major regions of eastern China). In these regions, MB and GE of temperature are -1.2~0.7 K and 1.8~2.6 K, respectively, which are -11.8~5.6% and 9.2~16.8% for RH, respectively.”**

**Comment 7:** *Figure 1: (1) The current Figure has no information regarding the season the measurements are performed. A comparison of both seasons should be made and a figure like Fig1(a) should be made for each season. The time series should include the same site for the two seasons. (2) Why is the base case exactly the same as the S3 in Figure 1(b)? (3) Add errors to the measurements.*

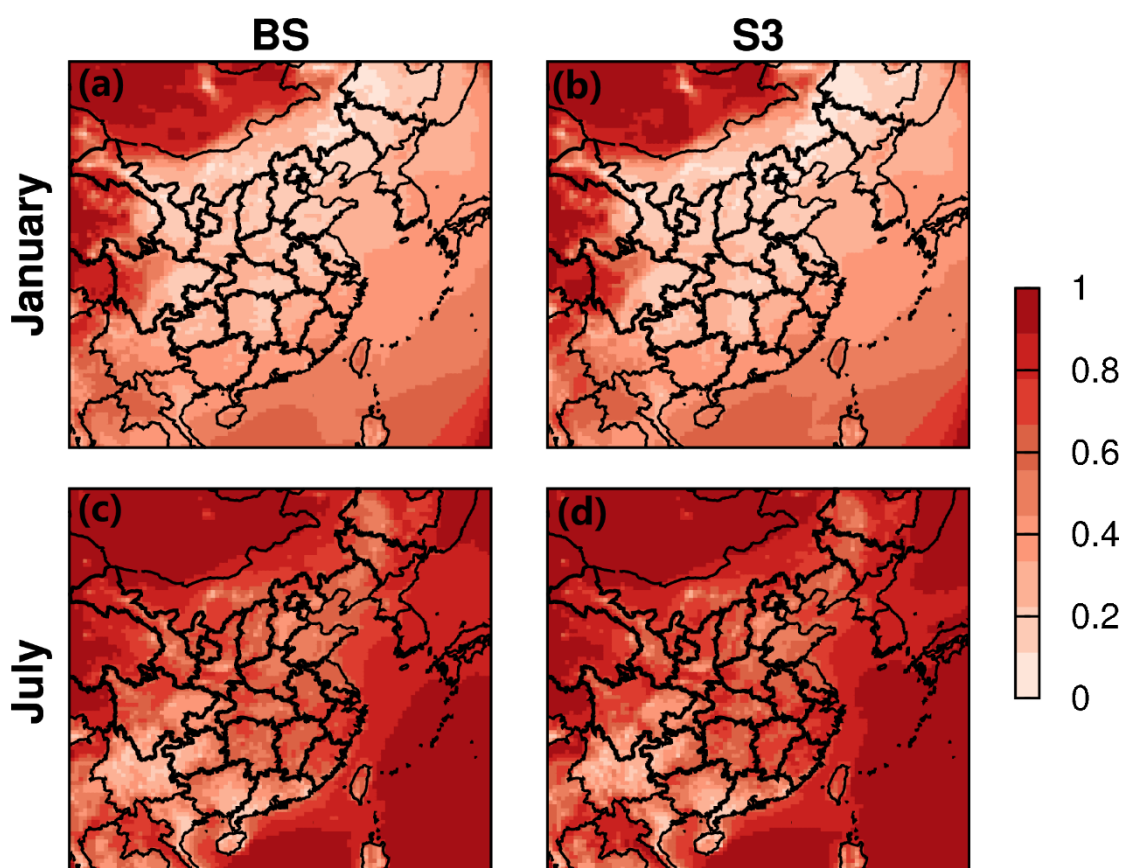
**Response 7:** (1) Due to limited observations of the simulated episode, we only have OC and OA measurements in January of 2013 at these sites. We used surface PM<sub>2.5</sub> alternatively to evaluate model performances in July of 2013. As a significant fraction of PM<sub>2.5</sub> in July is secondary, this still provides an indirect assessment of the model prediction of the oxidation capacity of the atmosphere, which is import for SOA formation. (2) The insignificant difference between BS and S3 in Figure 1(b) is likely due to a much smaller fraction of SOA compared to POA at this location predicted in the current model. Related discussions can refer to Response 2. (3) Unfortunately, there is no error information available for those measurements at this point.

**Comment 8:** *Figure 2: Why do you use in (a) the base case and not the S3 case? I would consider promoting the updated S3 on the left and the changes on the middle and right panels.*

**Response 8:** We have replaced case BS with S3 in Figure 2(a) and (d) and showed monthly-averaged results instead of averaged daily maximum in (b)-(c) and (e)-(f) in Figure 2.

**Comment 9:** *Figure 3: I don't see the point in presenting the ratio of LWC<sub>org</sub> to SOA. If both are expected to increase during pollution episodes then the ratio might stay the same therefore providing no valuable information. I would plot the SOA to OA as an alternative option or the SOA alone.*

**Response 9:** One effect of water in the organic phase is that it decreases the average molecular weight of the absorbing organic phase, which could affect the subsequent partitioning of other semi-volatile organic compounds (see Eq 2). The ALW<sub>org</sub>/SOA allows the readers to see the importance to consider ALW<sub>org</sub> in the partitioning calculation as it can account for a significant fraction of SOA and lead to a reduced average molecular weight. We agree with the reviewer that this ratio might not be very different between clean and polluted episodes. However, it is nonetheless useful in assessing the importance of including ALW<sub>org</sub> in SOA modeling. SOA/OA is certainly very useful information as well. Since POA is still significantly higher than SOA, especially during the winter month, SOA/OA ratio did not change significantly when ALW<sub>org</sub> is considered as shown in Figure R3. We have added this figure in the revised supplemental materials as Figure S13.



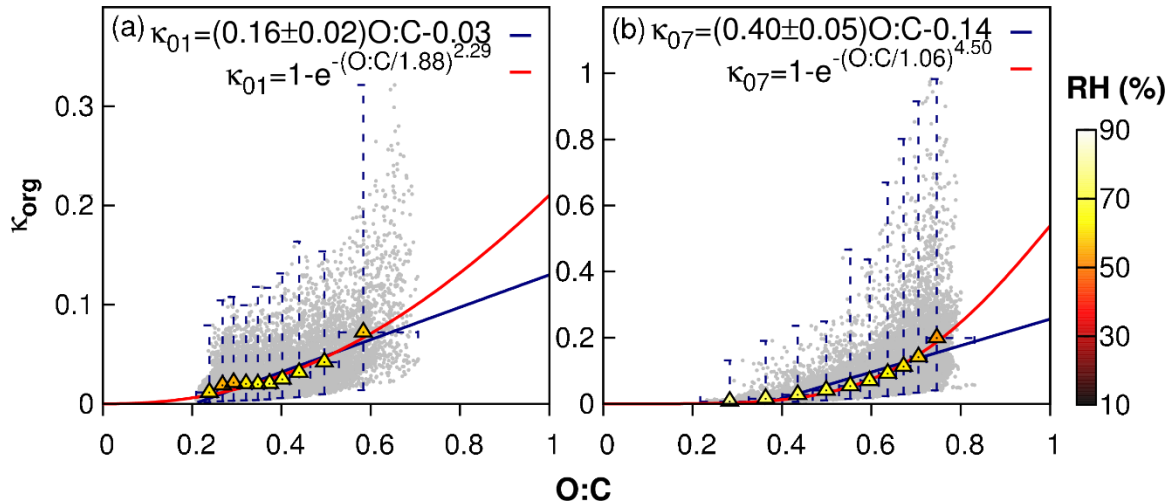
**Figure R3.** Averaged SOA/OA ratio from case BS and S3 during January and July of 2013.

**Comment 10:** *Figure 4: The data are really hard to observe. Please change*



colors and increase font size.

**Response 10:** Figure 4 (as shown below) has been revised to show all the data from each city in each month. Detailed results of each city are shown in Figure S9 and S10.



**Figure 4.** The correlation of hygroscopicity of organic aerosol ( $\kappa_{org}$ ) and O:C ratio at 9 representative cities including Shenyang (SS), Beijing (BJ), Jinan (JN), Zhengzhou (ZZ), Xi'an (XA), Nanjing (NJ), Shanghai (SH), Chengdu (CD), and Guangzhou (GZ) in January (a) and July (b) of 2013. O:C ratios are categorized into 10 bins. In each bin, the ranges of O:C and  $\kappa_{org}$  are represented by bars.

The mean values of O:C and  $\kappa_{org}$  are represented by triangles colored by the averaged RH of each bin. The relationship between  $\kappa_{org}$  and O:C is fitted by a linear function with reduced major axis regression (blue lines) and an exponential function (red lines), respectively.  $\kappa_{01}$  and  $\kappa_{07}$  represent the fitted correlation for January and July, respectively.

**Comment 11:** Figure 7: The graph is not clear. In the main text, the authors discuss that the daily maximum of SOA occurs when RH is greater than 70% in both cities. The RH is higher than 70% all the time. The time of the day is up to 24 hours and not 25. The markers and boxes for (c) are not discussed whether they represent the left or right axis.

**Response 11:** We have removed this figure to avoid confusion.

### Specific Comments

**Comment 12:** Line 34: Please elaborate more

**Response 12:** Now the text reads:

**“However, the models typically assume that the organic particulate matter (OPM) is an ideal mixture and ignore the partitioning of water vapor to OPM.”**

**Comment 13:** *Line 39: Please define generally with a statistical value that has meaning.*

**Response 13:** The text has been revised in L40-42 as follows:

**“The modified model can generally capture the observed surface organic carbon (OC) with a correlation coefficient R of 0.7, and the surface OA with the mean fractional bias (MFB) and mean fractional error (MFE) of -0.28 and 0.54, respectively.”**

**Comment 14:** *Line 91: Please elaborate more on the “purer condensed organics” for non-experts.*

**Response 14:** This should be condensed organics and has been revised accordingly.

**Comment 15:** *Line 99: “neglect” instead of “neglected”.*

**Response 15:** The text has been revised as instructed.

**Comment 16:** *Line 99: Please elaborate more on 1).*

**Response 16:** The text has been revised in L98-100 as following:

**“1) the molecular structures and interactions of functional groups (-OH, -C=O, -COOH, etc.) of condensed organics (non-ideality);”**

**Comment 17:** *Line 109: The sentence is missing a verb.*

**Response 17:** The sentence has been revised in L109-111 as following:

**“Laboratory and field studies have observed water absorbed by SOA from a variety of precursor VOCs (Lambe et al., 2011; Zhao et al., 2016b; Asa-Awuku et al., 2010; Varutbangkul et al., 2006).”**

**Comment 18:** *Line 128-131: The sentence is hard to read. Please rephrase.*

**Response 18:** The sentence has been revised in L126-129 as follows:

**“Using UNiversal Functional Activity Coefficient (UNIFAC) method (Fredenslund et al., 1975) for calculating activity coefficients of the organic-water mixture, it was found that in the eastern U.S., where biogenic SOA dominated the OA, considering  $ALW_{org}$  leads to a**

significant increase in predicted SOA (Pankow et al., 2015; Jathar et al., 2016).”

**Comment 19:** *Line 138: Which region?*

**Response 19:** The region refers to China. The text has been revised in L134-135 as follows:

**“Previous modeling studies in China indicate that SOA was underpredicted (Lin et al., 2016; Jiang et al., 2012)”**

**Comment 20:** *Line 163: (1) Don't change a line. (2) Also, why acidic conditions? Please, elaborate more. (3) Why is the reactive uptake of dicarbonyls, IEPOX and MAE in the “non-volatile” category?*

**Response 20:** (1) This is a mistake due to file format conversion and has been corrected. (2) This is a typo. SOA formed by isoprene oxidation under acidic conditions refers to IEPOX and MAE SOA based on chamber experiments (Lal et al., 2012; Lin et al., 2012; 2013). We have removed “isoprene oxidation under acidic conditions” in the revised text. (3) In the current model, we assume that the reactive uptake of dicarbonyls, IEPOX and MAE is irreversible, as an upper-limit estimation of SOA from these precursors. That’s why they are classified as non-volatile SOA. The text has been revised in L165-173 as following:

**“SOA from dicarbonyls, IEPOX, and MAE were formed by irreversible reactive uptake and categorized as NV-SOA in the current model as well. Some studies investigated SOA from glyoxal, methylglyoxal, and IEPOX using detailed reactions and reversible pathways in models or observed as reversible processes in chamber experiments, leading to a relatively lower SOA yield compared to the surface-controlled irreversible uptake (Lim et al., 2013; Knote et al., 2014; Galloway et al., 2009; El-Sayed et al., 2018; Budisulistiorini et al., 2017). The non-volatile assumption used in this paper allows an upper-limit estimation of the importance of these additional SOA formation pathways.”**

**Comment 21:** *Line 164: change to “was mostly”*

**Response 21:** This sentence has been moved to L162 in the revised manuscript and now it reads as follows:

**“The SOA module mostly follows Pankow et al. (2015).”**

**Comment 22:** *Line 165: Please elaborate more on the non-ideality calculation of the organic-water mixture for non-experts.*

**Response 22:** This sentence has been moved to L199-200 in the revised manuscript and now it reads as follows:

**“POA is also involved in the calculation of activity coefficients for the**

organic-water mixture.”

**Comment 23:** *Line 170: Is this the absorbing organic phase?*

**Response 23:** Yes. The text has been revised to “the absorbing organic phase”.

**Comment 24:** *Line 196: Change to “as water condenses”.*

**Response 24:** The text has been revised as instructed.

**Comment 25:** *Line 200: Please elaborate more on the “Kelvin effect neglected” for non-experts.*

**Response 25:** The text has been revised in L210-211 as following:

**“Based on the  $\kappa$ -Köhler theory with linearly additive hygroscopic behavior of each component of the mixed particle”**

**Comment 26:** *Line 204: Change to “can be estimated”.*

**Response 26:** The text has been revised as instructed.

**Comment 27:** *Line 242: Observations in 8 sub-regions of the domain during which period?*

**Response 27:** The text has been revised in L278-281 as follows:

**“Table 1 lists model statistics of mean observation (OBS), mean prediction (PRE), mean bias (MB), gross error (GE) and correlation coefficient (R) based on WRF and observations at monitoring sites located in 8 sub-regions of the domain (Figure S1) during January and July of 2013.”**

**Comment 28:** *Line 251: No significant improvements observed when applying the above additions means that the model is still missing a significant pathway to SOA formation, especially since OA in both seasons are dominated by SOA based on observations. This should be discussed in detail and in the context of previous studies and findings from AMS measurements in China.*

**Response 28:** We have revised the text and added a discussion about the underestimation of SOA in the current model. Please refer to Response 2 for more details.

**Comment 29:** *Line 258: Does it capture the observed diurnal variation? What is the  $R^2$  or  $R$  of the timeseries of the modeled to observed values? What is the ratio of the two? In many cases, it seems that the difference is higher than a factor of 4. Is that a usual discrepancy? If so, how is much is it improved when incorporating the detailed SOA models?*

**Response 29:** Our model can capture the diurnal variation. The mismatching

of several peak values might be due to uncertainties in the emission inventory and the underestimate of SOA in the current model. The R of the modeled to observed OA is 0.55. The ratio of the averaged prediction to observation is 0.75. Since the model predicts a very small ratio of SOA to POA, the improvement from the detailed SOA model is insignificant. The small SOA/POA ratio might be due to the missing SOA from other pathways including POA aging and oxidation from IVOCs and VOC oxidation products. This has been explained in Response 2.

We revised the text in L301-304 to expand more discussion of the modeled and observed OA comparison:

**“CMAQ can well capture the observed diurnal variation of OA in Beijing during wintertime, except for the underestimates of peak values. The correlation coefficient of modeled to observed OA is 0.55. We find a 25% underestimate of OA on average.”**

*Comment 30: Line 259: It would be great if “better” was described with statistical terminology. A way to describe the data and the comparison to modeled values would be to generate box and whiskers of the ratio of observations to modeled values for non-polluted and polluted days, respectively.*

**Response 30:** We did a mistake in the MFB and MFE calculation for OA, which should be -0.28 and 0.54, respectively. We also calculated the biases on polluted and non-polluted days of OA. MFB and MFE of polluted days are -0.38 and 0.64, which are -0.26 and 0.52 for non-polluted days. The text has been revised in L306-309 as follows:

**“The mean fractional bias (MFB) and mean fractional error (MFE) of polluted days are -0.38 and 0.64, respectively, which are worse than that of the non-polluted days (-0.26 for MFB and 0.52 for MFE). The overall MFB and MFE of OA during January are -0.28 and 0.54, within the criteria (MFB $\leq$ ±0.6; MFE $\leq$ 0.75) suggested by EPA (2007).”**

*Comment 31: Line 262-263: POA is not the primary contributor to OA in Beijing in winter. Many studies show that SOA is the major contributor and the path towards SOA formation is currently unknown and strongly dependent on LWC in the particles. Aging of POA not treated in the model is not guaranteed to be the main source of SOA.*

**Response 31:** Please refer to Response 2.

*Comment 32: Line 280: Here only one season is provided in terms of timeseries comparison of the model and obs. Please provide both seasons.*

**Response 32:** Unfortunately, detailed chemical composition measurements for

aerosols are very limited in China during 2013. We only have observations of OC and OA in January of 2013 and PM<sub>2.5</sub> in July of 2013 available for model evaluation.

**Comment 33:** *Line 285: Figure S5 shows the anthropogenic SOA and not the dicarbonyl SOA. Please separate the contributions and discuss them in the main text. Identifying the contribution of different compounds to SOA formation in China would be of great interest to the scientific community.*

**Response 33:** The contribution of each precursor to SOA of this episode has been shown in Hu et al. (2017) and will not be discussed in detail in the current study. The text has been revised in L344-346 as following:

**“Anthropogenic emissions are the major sources of SOA (Figure S6), such as dicarbonyl products from the oxidation of xylene and toluene (Hu et al., 2017).**

**Comment 34:** *Line 312: What about particulate nitrate?*

**Response 34:** The interactions between water-inorganics and water-organics are treated separately in the current model. We only focus on the water-organic interaction in the current study.

**Comment 35:** *Line 380: RH is higher than 70% all the time. What is the meaning of this sentence?*

**Response 35:** We have removed this figure to avoid confusion.

## References:

- Hodzic, A., Jimenez, J. L., Madronich, S., Canagaratna, M. R., DeCarlo, P. F., Kleinman, L., and Fast, J.: Modeling organic aerosols in a megacity: potential contribution of semi-volatile and intermediate volatility primary organic compounds to secondary organic aerosol formation, *Atmos. Chem. Phys.*, 10, 5491-5514, 10.5194/acp-10-5491-2010, 2010.
- Hu, J., Chen, J., Ying, Q., and Zhang, H.: One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system, *Atmos. Chem. Phys.*, 16, 10333-10350, 10.5194/acp-16-10333-2016, 2016.
- Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525-1529, 10.1126/science.1180353, 2009.
- Kota, S. H., Schade, G., Estes, M., Boyer, D., and Ying, Q.: Evaluation of MEGAN predicted biogenic isoprene emissions at urban locations in Southeast Texas, *Atmospheric Environment*, 110, 54-64, 2015.
- Lal, V., Khalizov, A.F., Lin, Y., Galvan, M.D., Connell, B.T., Zhang, R., 2012. Heterogeneous reactions of epoxides in acidic media. *J. Phys. Chem. A* 116, 6078e6090.
- Li, J., Cleveland, M., Ziemba, L. D., Griffin, R. J., Barsanti, K. C., Pankow, J. F., and Ying, Q.: Modeling regional secondary organic aerosol using the Master Chemical Mechanism, *Atmos. Environ.*, 102, 52-61, <https://doi.org/10.1016/j.atmosenv.2014.11.054>, 2015.
- Lin, Y.H., Zhang, Z.F., Docherty, K.S., Zhang, H.F., Budisulistiorini, S.H., Rubitschun, C.L., Shaw, S.L., Knipping, E.M., Edgerton, E.S., Kleindienst, T.E., Gold, A., Surratt, J.D., 2012b. Isoprene epoxydiols as precursors to secondary organic aerosol formation: acid-catalyzed reactive uptake studies with authentic compounds. *Environ. Sci. Technol.* 46, 250-258.
- Lin, Y.-H., Zhang, H., Pye, H. O. T., Zhang, Z., Marth, W. J., Park, S., Arashiro, M., Cui, T., Budisulistiorini, S. H., Sexton, K. G., Vizuete, W., Xie, Y., Luecken, D. J., Piletic, I. R., Edney, E. O., Bartolotti, L. J., Gold, A., and Surratt, J. D.: Epoxide as a precursor to secondary organic aerosol formation from isoprene photo-oxidation in the presence of nitrogen oxides, *P. Natl. Acad. Sci. USA*, 110, 6718-6723, doi:10.1073/pnas.1221150110, 2013.
- Liu, J., Shen, J., Cheng, Z., Wang, P., Ying, Q., Zhao, Q., Zhang, Y., Zhao, Y., and Fu, Q.: Source apportionment and regional transport of anthropogenic secondary organic aerosol during winter pollution periods in the Yangtze River Delta, China, *Sci. Total Environ.*, 710, 135620, <https://doi.org/10.1016/j.scitotenv.2019.135620>, 2020.
- Murphy, B. N., Woody, M. C., Jimenez, J. L., Carlton, A. M. G., Hayes, P. L., Liu, S., Ng, N. L., Russell, L. M., Setyan, A., Xu, L., Young, J., Zaveri, R. A., Zhang, Q., and Pye, H. O. T.: Semivolatile POA and parameterized total combustion SOA in CMAQv5.2: impacts on source strength and partitioning, *Atmos. Chem. Phys.*, 17, 11107-11133, 10.5194/acp-17-11107-2017, 2017.

Pankow, J. F., Marks, M. C., Barsanti, K. C., Mahmud, A., Asher, W. E., Li, J., Ying, Q., Jathar, S. H., and Kleeman, M. J.: Molecular view modeling of atmospheric organic particulate matter: Incorporating molecular structure and co-condensation of water, *Atmospheric Environment*, 122, 400-408, <http://dx.doi.org/10.1016/j.atmosenv.2015.10.001>, 2015.

Pye, H. O. T., Murphy, B. N., Xu, L., Ng, N. L., Carlton, A. G., Guo, H., Weber, R., Vasilakos, P., Appel, K. W., Budisulistiorini, S. H., Surratt, J. D., Nenes, A., Hu, W., Jimenez, J. L., Isaacman-VanWertz, G., Misztal, P. K., and Goldstein, A. H.: On the implications of aerosol liquid water and phase separation for organic aerosol mass, *Atmos. Chem. Phys.*, 17, 343-369, 10.5194/acp-17-343-2017, 2017.

Qiao, X., Ying, Q., Li, X., Zhang, H., Hu, J., Tang, Y., and Chen, X.: Source apportionment of PM<sub>2.5</sub> for 25 Chinese provincial capitals and municipalities using a source-oriented Community Multiscale Air Quality model, *Sci. Total Environ.*, 612, 462-471, <https://doi.org/10.1016/j.scitotenv.2017.08.272>, 2018.

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.

Shi, Z., Li, J., Huang, L., Wang, P., Wu, L., Ying, Q., Zhang, H., Lu, L., Liu, X., Liao, H., and Hu, J.: Source apportionment of fine particulate matter in China in 2013 using a source-oriented chemical transport model, *Science of the Total Environment*, 601, 1476-1487, <http://dx.doi.org/10.1016/j.scitotenv.2017.06.019>, 2017.

Shrivastava, M. K., Lane, T. E., Donahue, N. M., Pandis, S. N., and Robinson, A. L.: Effects of gas particle partitioning and aging of primary emissions on urban and regional organic aerosol concentrations, 113, 10.1029/2007jd009735, 2008.

Situ, S., Wang, X., Guenther, A., Zhang, Y., Wang, X., Huang, M., Fan, Q., and Xiong, Z.: Uncertainties of isoprene emissions in the MEGAN model estimated for a coniferous and broad-leaved mixed forest in Southern China, *Atmos. Environ.*, 98, 105-110, <https://doi.org/10.1016/j.atmosenv.2014.08.023>, 2014.

Sun, Y., Du, W., Fu, P., Wang, Q., Li, J., Ge, X., Zhang, Q., Zhu, C., Ren, L., Xu, W., Zhao, J., Han, T., Worsnop, D. R., and Wang, Z.: 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.

Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.: Aerosol composition, sources and processes during wintertime in Beijing, China, *Atmos. Chem. Phys.*, 13, 4577-4592, 10.5194/acp-13-4577-2013, 2013.

Tkacik, D. S., Presto, A. A., Donahue, N. M., and Robinson, A. L.: Secondary Organic Aerosol Formation from Intermediate-Volatility Organic Compounds: Cyclic, Linear, and Branched Alkanes, *Environmental Science & Technology*, 46, 8773-8781, 10.1021/es301112c, 2012.

Wang, P., Schade, G., Estes, M., and Ying, Q.: Improved MEGAN predictions of biogenic isoprene in the contiguous United States, *Atmospheric Environment*, 148, 337-351, <http://dx.doi.org/10.1016/j.atmosenv.2016.11.006>, 2017.

Ying, Q., Cureño, I. V., Chen, G., Ali, S., Zhang, H., Malloy, M., Bravo, H. A., and Sosa, R.: Impacts of Stabilized Criegee Intermediates, surface uptake processes and higher aromatic secondary organic aerosol yields on predicted PM<sub>2.5</sub> concentrations in the Mexico City Metropolitan Zone, *Atmos. Environ.*, 94, 438-447, <https://doi.org/10.1016/j.atmosenv.2014.05.056>, 2014.

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.

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.

Zhao, J., Qiu, Y., Zhou, W., Xu, W., Wang, J., Zhang, Y., Li, L., Xie, C., Wang, Q., Du, W., Worsnop, D. R., Canagaratna, M. R., Zhou, L., Ge, X., Fu, P., Li, J., Wang, Z., Donahue, N. M., and Sun, Y.: Organic Aerosol Processing During Winter Severe Haze Episodes in Beijing, *J. Geophys. Res.*, 124, 10248-10263, 10.1029/2019jd030832, 2019.

Zhao, Y., Hennigan, C. J., May, A. A., Tkacik, D. S., de Gouw, J. A., Gilman, J. B., Kuster, W. C., Borbon, A., and Robinson, A. L.: Intermediate-Volatility Organic Compounds: A Large Source of Secondary Organic Aerosol, *Environ. Sci. Technol.*, 48, 13743-13750, 10.1021/es5035188, 2014.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, *Atmos. Chem. Phys.*, 15, 2031-2049, 10.5194/acp-15-2031-2015, 2015.