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 0.83, 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 0.83 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."

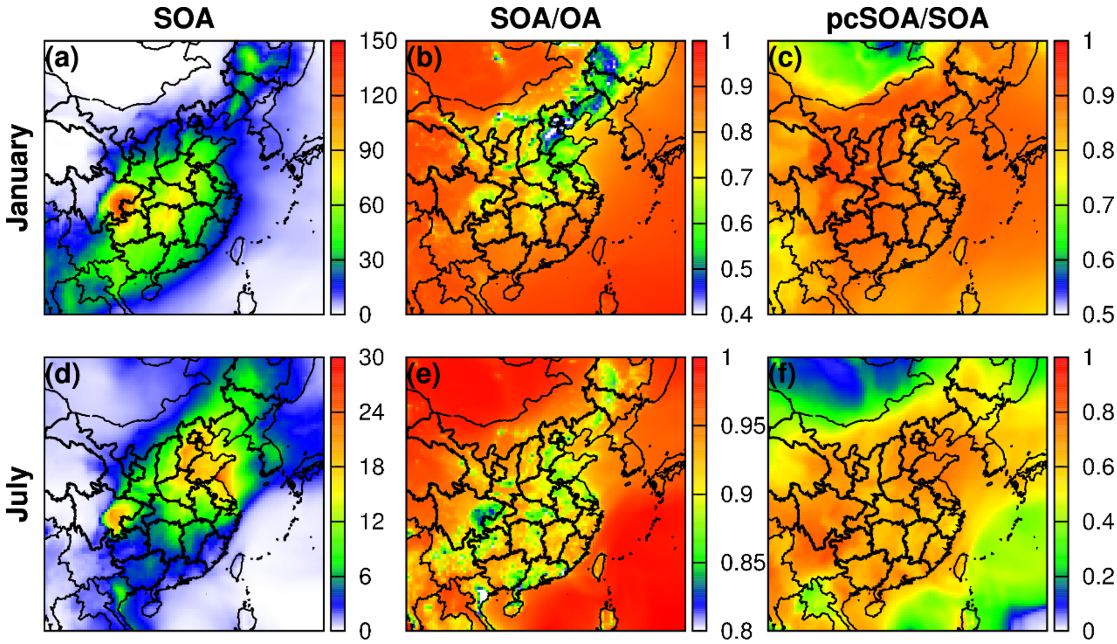

Figure R1. Mean SOA, SOA/OA, and pcSOA/SOA ratio predicted during January and July of 2013 by CMAQv5.3.1.

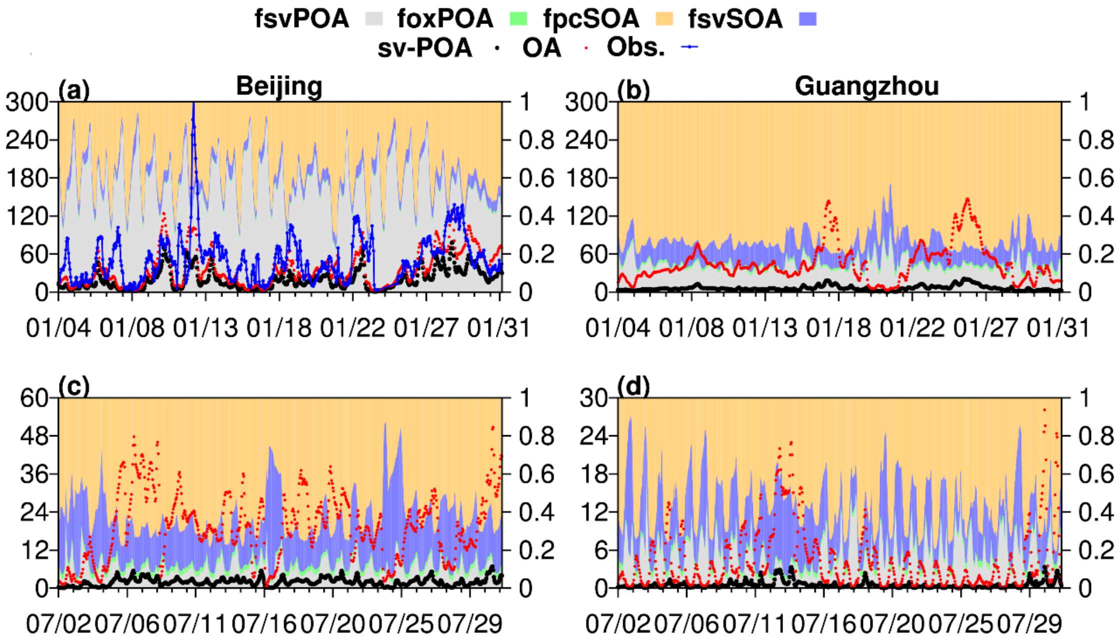

Figure R2. Modeled concentration of semi-volatile POA (sv-POA), OA and fraction of each organic aerosol component fsvPOA (sv-POA), foxPOA (oxidation of sv-POA), fpcSOA (pc-SOA) and fsvSOA (traditional SOA) in Beijing (a, c) and Guangzhou (b, d). Observations of OA in January 2013 at Beijing (Obs.) are also included in (a). The left axis is for the concentrations ($\mu g\ m^{-3}$) 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 NO2 and SO2 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 $PM_{2.5}$ alternatively to evaluate model performances in July of 2013. As a significant fraction of $PM_{2.5}$ 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 LWCorg 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_{org}/SOA allows the readers to see the importance to consider ALW_{org} 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_{org} 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_{org} is considered as shown in Figure R3. We have added this figure in the revised supplemental materials as Figure S13.

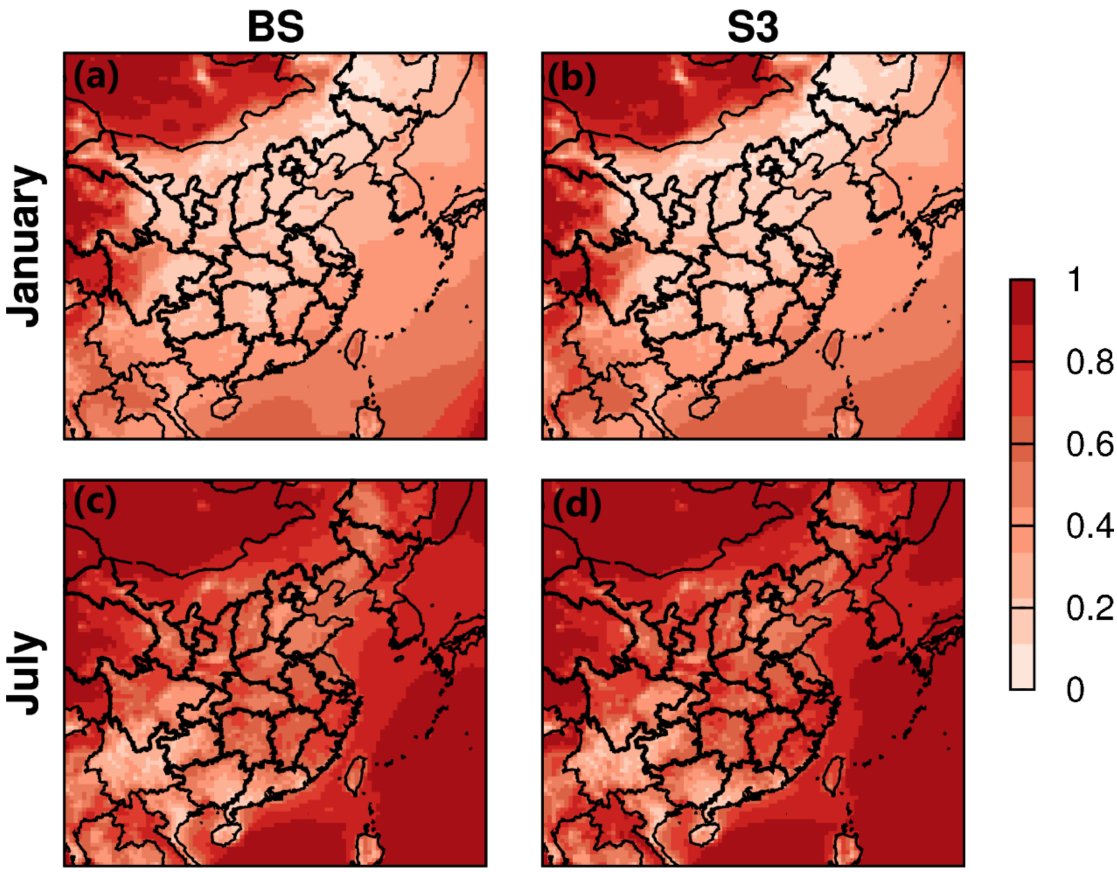

Figure R3. Averaged SOA/OA ratio from case BS and S3 during January and July of 2013.

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.

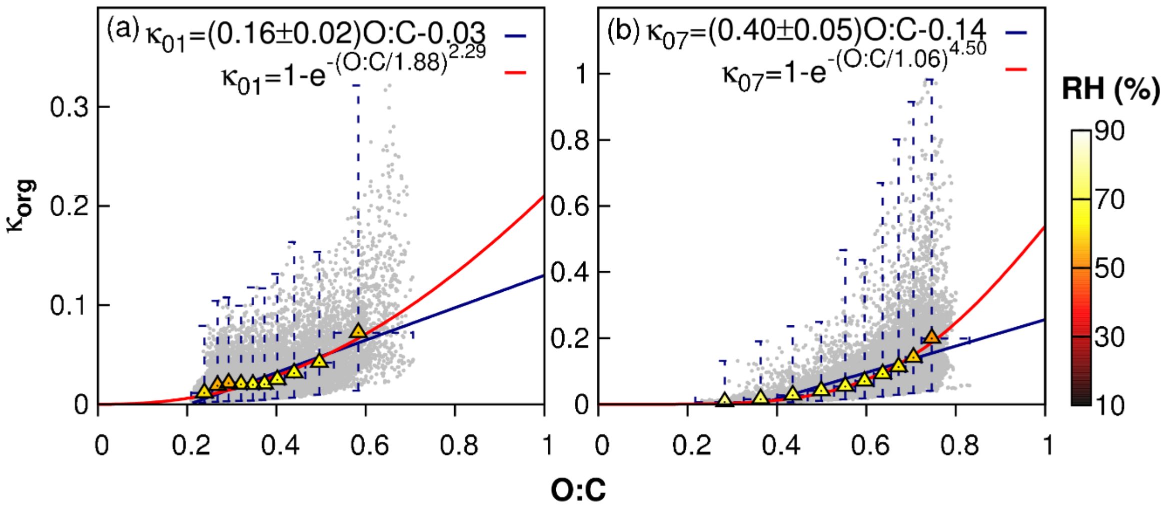

Figure 4. The correlation of hygroscopicity of organic aerosol (κ_{org}) and O:C ratio at 9 representative cities including Shenyang (SS), Beijing (BJ), Jinan (JN), Zhengzhou (ZZ), Xi'an (XA), Nanjing (NJ), Shanghai (SH), Chengdu (CD), and Guangzhou (GZ) in January (a) and July (b) of 2013. O:C ratios are categorized into 10 bins. In each bin, the ranges of O:C and κ_{org} are represented by bars. The mean values of O:C and κ_{org} are represented by triangles colored by the averaged RH of each bin. The relationship between κ_{org} and O:C is fitted by a linear function with reduced major axis regression (blue lines) and an exponential function (red lines), respectively. κ_{01} and κ_{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 κ-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 R2 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≤±0.6; MFE≤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 $PM_{2.5}$ 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.

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Response to Referee #2

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 fonts, the responses are in regular font, and the revised text is in bold font.

In this paper, Li et al. have modified the CMAQ model to take into account the impacts of water partitioning and polarity of organic compounds on SOA formation. The model was applied over Eastern China to estimate the regional and seasonal impacts of these modifications on SOA and the aerosol water content. This study may have potential to contribute in the organic aerosol modeling field but major revisions needs to be done before publication. In particular, I have several concerns regarding the validity of the scientific methodology used and the presentation of the study. Therefore, I would recommend publication only if these comments will be addressed and fundamental changes will be contacted.

Major comments

Comment 1: *Page 4 line 94: The majority of current CTMs have replaced the 2-product model with the VBS approach. Please make this clear and refer to the 2-product model of Odum et al. (1996) for historical reasons.*

Response 1: The text has been revised in the manuscript (L88-95) to make this clear:

"**The formation of condensed organic products is commonly represented by lumped surrogate SVOCs in a 2-product model with volatilities and SVOC yields fitted to chamber experiments (Odum et al., 1996). To better represent the volatility of primary organic aerosol (POA) and the multi-generation oxidation of SVOCs to a wider range, Donahue et al. (2006) proposed the volatility basis set (VBS) model in which the mass yields of SVOCs are fitted to a fixed number of volatility bins (usually 0.01-105 μg m-3). The VBS model has been adopted by several CTMs (such as WRF-Chem, GEOS-Chem, etc.).**"

Comment 2: *Page 6, line 162: Please add the appropriate references to support the nonvolatile nature of the products by these oxidation pathways.*

Response 2: Formation of these non-volatile SOA was traditionally treated in the CMAQ model, except for dicarbonyls, IEPOX, and MAE that are assumed to form SOA by irreversible reactive uptake in our model, as an upper-limit estimation of SOA from these precursors. The text has been revised to make this clear (L162-173 in the manuscript):

"The SOA module mostly follows Pankow et al. (2015). Two types of SOA as traditionally treated in CMAQ were considered, "semi-volatile" (SV) portion that formed via equilibrium absorption-partitioning of SVOCs, and

"non-volatile" (NV) portion that includes the oligomers and SOA formed via direct oxidation of aromatics at low-NOx. 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 3: *Page 6 line 149: There is no discussion in the methodology about the observations and the statistical analysis metrics used to evaluate the model performance. Especially for the OA observations, there is no reference provided or description of the methods used.*

Response 3: Thank you for pointing this out. We have included details of observation data, statistical analysis metrics for both meteorology and aerosols in the revised manuscript:

In L265-276

"**The meteorological inputs and emissions have been used in several previous publications. Model performance on meteorological parameters (temperature and RH), gaseous species and gas and aerosol concentrations have been extensively evaluated (Hu et al., 2016; Hu et al., 2017; Qiao et al., 2018; Shi et al., 2017). A summary of the model performance related to this study is provided below. Observed meteorological data were obtained from the National Climatic Data Center (ftp://ftp.ncdc.noaa.gov/pub/data/noaa). Observations of OC at two urban locations, Beijing (Cao et al., 2014; Wang et al., 2015) and Guangzhou (Lai et al., 2016) and OA in Beijing (Sun et al., 2014) during January of 2013 as well as surface $PM_{2.5}$ at several monitoring sites during July of 2013 from China National Environmental Monitoring Center (http://113.108.142.147:20035/emcpublish/) were used to evaluate model estimates of aerosols. Details of measurement methodology and uncertainties of observations are listed in the corresponding references.**"

In L278-282

"**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."

In L294-296

"**Overall, the mean fractional bias (MFB) and mean fractional error (MFE) of OC are -0.20 and 0.27, within the criteria (MFB≤±0.6; MFE≤0.75) suggested by EPA (2007).**"

In L308-309:

"**The overall MFB and MFE of OA during January are -0.28 and 0.54, within the criteria (MFB≤±0.6; MFE≤0.75) suggested by EPA (2007).**"

Comment 4: *Page 7 line 179: More details are needed here. (1) How the model defines the low NOx and high NOx conditions? Which compounds each of the lump species represent? What is the difference between the lumped species of the same precursor (e.g., BNZ1, BNZ2, BNZ3)? (2) Can you include the aerosol yields for each lumped species in tables S1 and S2? Are these aerosol yields NOx-dependent? (3) What does the SVP stands for in Tables S1 and S2?*

Response 4: (1) SOA formation in CMAQv5.0.2 is based on the frame of a previous version 4.7.1. All the details about "high" and "low" NO_x conditions (based on chamber experiments of corresponding VOCs), lumping species and method of each precursor, and the yields of precursors from parent VOCs have been documented by Carlton et al. (2010) and summarized in the revised supplemental materials as following:

"**The CMAQ model treats high and low NOx SOA formation pathways during OH oxidation by allowing the lumped RO2 radical to competitively react with HO2 and NO. Using the lumped ARO1 species as an example, an SOA formation specific RO2 radical ARO1RO2 is added as a gas phase reaction product with OH:**

ARO1 + OH → ARO1RO2 + products

The ARO1RO2 can react with both HO2 and NO, as shown in the following two reactions:

ARO1RO2 + HO2 → HO2 + TOLNRXN; k1

ARO1RO2 + NO → NO + TOLHRXN; k2

Details of the determination of the rate constants can be found in Carlton et a. (2010). The TOLNRXN and TOLHRXN are counter species that track how much ARO1 is reacted through low NOx and high NOx pathways, respectively, in one gas chemistry time step. The concentrations of these counter species are passed into the aerosol module to calculate the formation semi-volatile products (TOL1 and TOL2) in the high NOx pathway and non-volatile products (TOL3) in the low NOx pathway, using the mass-specific yields, as listed in Table S1 and S2. Equilibrium partitioning of TOL1 and TOL2 in the gas phase and their counterparts ATOL1 and ATOL2 in the organic phase are affected by temperature and the amount of absorbing organics in the aerosol phase. Similar

treatments are applied to the other lumped aromatic compounds ARO2, with xylene as a representative and most abundant species in that group, and to benzene. SOA formation from lumped long-chain alkene species ALK5, and isoprene and monoterpenes are not considered as NO_x dependent and are represented by equilibrium partitioning of one or two semi-volatile oxidation products. Details of the mass-specific yields of semi-volatile products and other related parameters can be found in Table S1 and S2."

We revised the text in (L188-190) to make it clear:
"**More details about the lumped precursors such as formation conditions ("high" and "low" NOx), lumping species and method, and yields from parent VOCs can be found in Carlton et al. (2010) and summarized in SI.**"

(2) In the CMAQ model, the amount of SOA can form after a precursor reacts with OH, O_3 or NO_3 depends on the volatility of the products, which is temperature dependent, and the amount of the absorbing organics. The mass yields of the semi-volatile or non-volatile products are included in Table S1 and S2 in the revised supplementary materials. For more details, we refer the readers to Carlton et al. (2010) and the references therein.

(3) SVP is the saturation vapor pressure. We have explained this in the corresponding tables.

Comment 5: *Page 8, lines 194-198: (1) Does the absorbing phase of equation 1 includes only the water associated with the organics (from eq. 3) or it includes the total water (including the water associated with the inorganic aerosol components)? (2) Under high RH (higher than the organic/inorganic phase separation RH, SRH), the aerosol organic phase is well mixed with the inorganic salts and, therefore, the aerosol water associated with the inorganic constituents can also contribute to the SOA absorbing medium (Pye et al., 2017). Please clarify what you have assumed here and add the relative discussion.*
Response 5: (1) The absorbing phase of equation 1 only includes organic and water associated with organics when considering water-organic interactions. We explained this in the original text L195-196:
"In addition to organic compounds, water partitioning into OPM is enabled according to Eq 1 and Eq 2. In such a case, the absorbing phase in Eq 1 includes both organic aerosols and water partitioning into OPM."

(2) We didn't consider the mixing of organic and inorganic phase in this study and assumed that they are always two distinct aerosol phases without direct interactions. The phase separation RH (SRH) depends on the OM/OC ratio of the organic phase. Unlike the conditions modeled by Pye et al. (2017) for the

southeast US where SOA is often the dominant OA component, the winter episode we modeled is dominated by primary emitted organic aerosols thus with a relatively low OM/OC (~1.4-1.6). The SRH based on equation (7) of Pye et al. (2017) is ~97%-99%. The summer episode has more contributions of SOA to OA, with OM/OC~1.8, which corresponds to an SRH of 87%. Thus, we don't expect interactions of organic and inorganic phases to occur in high frequency to greatly influence the model results. We assumed no interactions between inorganic and organic phases in the current model. We have also revised text in L207-208:
"**In the current model, we assumed no interactions between the inorganic and organic phases.**"

Comment 6: *Page 8, Equation 3: This equation gives the volume of water associated with the organic fraction of the aerosol. However, ALW on the left-hand side refers to the mass of water. Please correct.*
Response 6: The equation has been corrected in the revised text L212-214:
"

$$\boldsymbol{ALW_{org} = \rho_w V_{org} \kappa_{org} \frac{a_w}{1 - a_w}} \quad \textbf{(Eq3)}$$

where ρ_w is the density of water (assumed to be 1 g cm^{-1}), V_{org} is the volume concentration of organics, and a_w is the water activity (assumed to be the same as RH)."

Comment 7: *Page 8, lines 203-204: How do you estimate the hygroscopicity?*
Response 7: We used Eq3 to estimate the hygroscopicity. This has been clarified in the revised text in L214-216:
"**Since ALW_{org} in this study is calculated mechanistically using the partitioning theory, κ_{org} can be estimated by rearranging Eq3:**

$$\boldsymbol{\kappa_{org} = \frac{ALW_{org}}{\rho_w V_{org}} \times \frac{1 - a_w}{a_w}} \quad \textbf{(Eq4)}$$

"

Comment 8: *Page 8, lines 204-205: How do you calculate the ALW? Are you using κ_{org} and the eq3? Do you use the kappa hygroscopicity to calculate the ALW? If so, how you estimate the kappa?*
Response 8: In this study, the ALW_{org} is independently calculated by mechanistically allowing water molecules to partition into the organic phase with UNIFAC calculated activity. In such a case, Eq 3 can be used to provide an independent estimation of κ_{org}. Linear regression analysis can be performed using the calculated κ_{org} against the model calculated O/C ratios, as shown in Figure 4. We have removed this sentence to avoid confusion.

Comment 9: *Page 9, lines 212-216: Add a reference to tables S1 and S2. Furthermore, why the values of OM:OC in tables S1/S2 are different than the values provided by your reference (Pye et al., 2017)?*

Response 9: We used OM:OC ratio in Pankow et al. (2015). There were mistakes in the original Table S1 and S2 and have been corrected in the revised supplemental materials. The text has also been revised in L228-229 as following:

"**The OM:OC ratio of each SOA component follows Pankow et al. (2015) as shown in Table S1-S2.**"

Comment 10: *Page 9 lines 217-218: Can you add the total size of the model domain?*

Response 10: The text has been revised in L231-233 as follows:

"**The simulation domain has a horizontal resolution 36 km × 36 km (100 × 100 grids) and a vertical structure of 18 layers up to 21 km, which covers eastern China as shown in Figure S1.**"

Comment 11: *Page 9 lines 233: (1) What boundary conditions are used? Please make a comment on how these can affect the simulation results. (2) I would recommend adding spatial maps of primary organic aerosol emissions and SOA precursor emissions and summarizing in a Table the domain average emission rates of POA and each SOA precursors.*

Response 11: (1) We used a predefined boundary profile in CMAQ that represents a clean continental condition. The northern and western boundaries, as well as areas to the further north and west, are mostly remote areas with much lower emissions. The mountains in the north and west part of the domain also limit the influence of emissions enter from the boundaries to the central part of the domain. The influence of marine air from the south and east boundaries is also small, as local emissions dominate the concentrations.

(2) A figure and a table showing emissions of POA and SOA precursors were added to the revised supplemental materials as Figure S2 and Table S4.

Comment 12: *Pages 9-10 lines 234-238: (1) This paragraph needs to be expanded and written in a separate section. In this section the authors should describe in more detail the following: i) Basecase simulation. Please explain how the default CMAQ is simulating POA and SOA and how different is this modelling configuration with the one the authors are testing, ii) Sensitivity simulations. Please explain in much more detail the sensitivity simulations conducted in this work. (2) In addition the authors say that they have conducted three sensitivity scenarios named S1, S2 and S3. In their manuscript they only show results from S3 and they never discuss the results of S1 and S2.*

Response 12: (1) Details of how default CMAQ simulates SOA formation through equilibrium partitioning of lumped semi-volatile products into the organic phase (which include both POA and SOA) have been described by Carlton et al. (2010) and Hu et al. (2017) so we don't think it is necessary to repeat it here. We expanded each simulation scenario in details in the revised text in L249-259:

"**Four scenarios are investigated in this study. The base case (BS) applies the default secondary organic aerosol module of CMAQ v5.0.1. In this case, no water partitioning into OPM is considered. Lumped semi-volatile products from the oxidation of various precursors partition into a single organic phase, which is considered as an ideal mixture of POA and SOA with γ_{org}=1. The water case (S1) includes water partitioning into OPM, which is again considered as an ideal solution (γ_{org}=1 and γ_{H2O}=1). The UNIFAC case (S2) considers the interaction between organic constituents with UNIFAC calculated activity coefficients ($\gamma_{org}\neq$1) but does not allow water partitioning into OPM. The combined case (S3) allows both water partitioning and interactions between all constituents (including water and organics) using UNIFAC calculated activity coefficients ($\gamma_{org}\neq$1 and $\gamma_{H2O}\neq$1).** "

(2) The impacts of S1 and S2 were discussed in Section 4 of the original manuscript Page 16 L405-419. We also included a description of the outlines of the results and discussions in the original manuscript Page 6 L140-148. The text has been revised in L259-262 to make it clear:
"**The results of BS and S3 are used to examine the overall impacts of water partitioning into OPM and polarity of organics on SOA and ALWorg, as shown in Section 3.1-3.4. The separate influences of those two processes on SOA from S1 and S2 are discussed in Section 3.5.**"

Comment 13: *Page 10 line 240: The section "Model evaluation" is extremely problematic and raises questions on the validity of the modelling results given that the model evaluation is insufficient. More specifically: 1) Given that the CMAQ default configuration has been modified to consider the importance of water and organic compound polarity on SOA formation, an accurate evaluation of the updated model performance is needed. 2) You should compare the model results for organic compounds during both July and January 2013. Currently, the evaluation includes a comparison with OC observations only during January over only three locations of the relatively large model domain. Furthermore, please mention in the text what factor have you used to convert the modeled OA to OC. 3) The total PM2.5 measurements have been used to evaluate the model performance during July without explaining the rationale of this choice since the focus of this study is solely the organic fraction of the aerosols. I suggest removing the PM2.5 evaluation or at least moving it to the supplement. 4) Can you include more OC/OA observations over other areas of their domain in your evaluation? 5) It is also important to compare the simulated*

POA and SOA against observations (e.g., from AMS). Furthermore, it would be helpful to show how the model performance against SOA measurement changes between the BC and the S1, S2, S3 cases.

Response 13: (1) For the updates in CMAQ except for water partition into OPM and non-ideality of the organic-water mixture, previous studies have extensively examined the model performances and will not be further discussed in detail in this work. The text has been revised in L149-161 to reflect this:

"**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_2 and SO_2 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.**"

(2) 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 $PM_{2.5}$ in July of 2013 available for model evaluation. Thus, we opt to use the most relevant data to provide a very limited assessment of the capability of the model in predicting SOA. The factors for OA to OC conversion follow the OM:OC ratio listed in Table S1 and S2 for SOA. POC is directly predicted by the model. The text has been revised in L291-292 to make this clear:

"**The factors used to convert SOA to OC (SOC) are listed in Table S1-S2. OC from POA (POC) is directly predicted by the model.**"

(3) Even though limited OC/OA measurements are available to us during this period, the base case model is later applied by another research group to model wintertime SOA formation in east China (Liu et al., 2020). The predicted OC and SOA agree well with observations (Figure 2 of Liu et al. 2020), and the model performance statistics for OC and SOA are similar to those of $PM_{2.5}$. We agree with the reviewer that $PM_{2.5}$ is not an ideal indicator to evaluate the capability of the model in predicting SOA, however, as a significant fraction of $PM_{2.5}$ in July is secondary, this still provides an indirect assessment of the model prediction of oxidation capacity of the atmosphere, which is import for

SOA formation. Additional modeling studies are needed to evaluate the performance of the model in summer.
We have explained this in the revised text (L327-329) as following:
"**Due to the lack of observed OC and OA in July of 2013, as an alternative, model performances are evaluated by comparing predicted and observed $PM_{2.5}$ at ground sites (Figure S1) as shown in Figure S3.**"

(4) We do not have more OC and OA data for the simulating episode of 2013.

(5) We did not have SOA observations in this episode. We compared the modeled SOA/POA ratio with AMS observations from other literature, finding a significant underestimation in the current model. This bias might be due to missing SOA from combustion (intermediate volatile organic compounds, IVOCs) and not treating POA as semi-volatile. We added a discussion about the model bias in the revised manuscript in 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 0.83 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."

Since SOA is underestimated, no significant differences in BS and S3 are observed. Case S1 and S2 are designed for the sensitivity test of model results to water partitioning and non-ideality of condensed organics separately. Therefore, we did not evaluate model performances from S1 and S2.

Comment 14: *Page 10 lines 251-253: The authors state here that the impacts of water-cocondensation and polarity of organic condensed species on SOA* *formation are not significant during winter. This highlights the need to evaluate their model results during July where they have found significant changes with the basecase simulation. Furthermore, the results from the three sensitivity simulations should be evaluated individually.*

Response 14: Unfortunately, we have no observations of OC, OA or (and) SOA of July 2013. We tried to evaluate model performances by comparing the predicted and observed $PM_{2.5}$ as an alternative. This has been explained in the revised manuscript in L327-329:

"**Due to the lack of observed OC and OA in July of 2013, as an alternative, model performances are evaluated by comparing predicted and observed PM2.5 at ground sites (Figure S1) as shown in Figure S3.**"

Since SOA is underestimated, no significant differences in BS and S3 are observed. Case S1 and S2 are designed for the sensitivity test of model results to water partitioning and non-ideality of condensed organics separately. Therefore, we did not evaluate model performances from S1 and S2.

Comment 15: *Page 10 lines 254-256: These are indeed possible factors. Can the authors comment, based on their analysis, which of these two possible factors is more important and try to be more specific? A comparison with AMS observations would be helpful here.*

Response 15: We did a sensitivity test by simulating the same episode with the current CMAQv5.3.1 in which POA was treated as semi-volatile and aging in the gas phase. Also, a missing source of SOA from intermediate VOCs (IVOCs) oxidation and aging of IVOCs and VOCs oxidation products (pcSOA) was added in CMAQv5.3.1. The results showed that the modeled SOA/POA has been improved from 0.12 of case S3 to 0.83 (Figure R1), more close to AMS observations. However, there were still some peak values underestimated by the model, which might be due to the uncertainties of POA emissions. We have revised the text (L309-326), as mentioned in Response 13 (5).

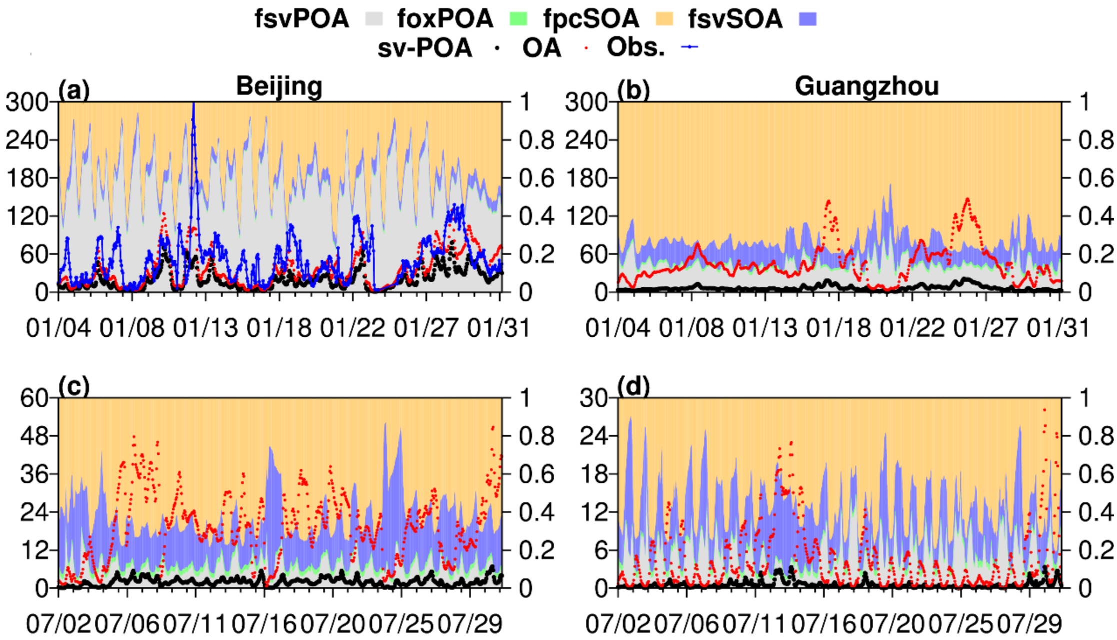

Figure R1. Modeled concentration of semi-volatile POA (sv-POA), OA and fraction of each organic aerosol component fsvPOA (sv-POA), foxPOA (oxidation of sv-POA), fpcSOA (pc-SOA) and fsvSOA (traditional SOA) in Beijing (a, c) and Guangzhou (b, d). Observations of OA in January 2013 at Beijing (Obs.) are also included in (a). The left axis is the concentration ($\mu g\ m^{-3}$) and the right axis is the fraction of OA components.

Comment 16: *Page 11 line 263: The aging of POA, under specific conditions can enhance the SOA formation, especially over polluted areas. Can the authors comment how this important omission of their model configuration can affect their result? Once again, a comparison against POA and SOA from AMS observations will be helpful to identify the limitations of their model due to the treatment of POA as non-volatile and non-reactive.*

Response 16: We did a sensitivity test by CMAQv5.3.1 that includes SOA from POA aging, IVOCs oxidation, and aging of IVOCs and VOCs oxidation products. The modeled SOA/POA is improved greatly in Beijing with no significant improvement in OA compared to the results of case S3. A significant improvement of SOA was observed from the contribution from IVOCs oxidation, and the aging of IVOCs and VOCs oxidation products (pc-SOA). Discussions about this have been included in the revised text in L309-326, as mentioned in Response 15.

Also, we 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.

Comment 17: *Page 11 line 264: Please provide two spatial maps of the fraction SOA/Total OA during January and July 2013 so as to show the contribution of POA and SOA to total OA during each simulation period.*
Response 17: We have added a figure of SOA/OA ratio by case BS and S3 (as shown below) in the revised supplemental materials (Figure S13).

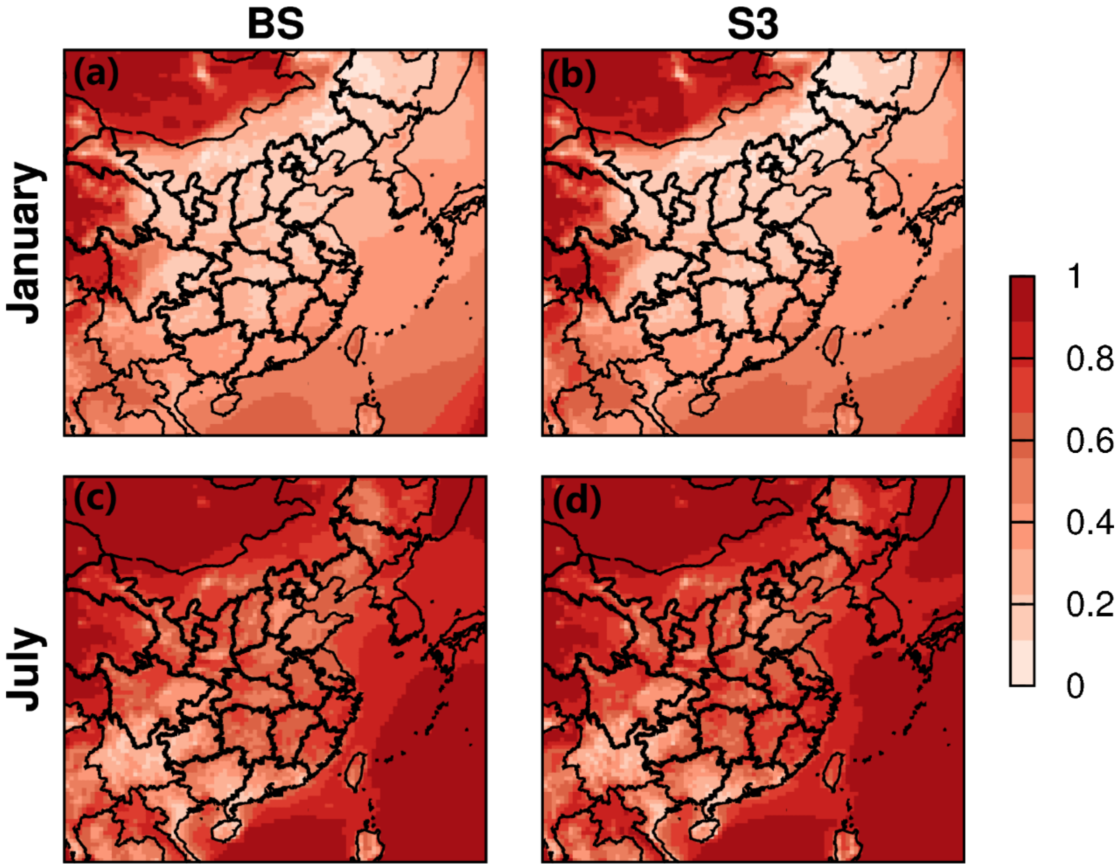

Figure R2. Averaged SOA/OA ratio from case BS and S3 during January and July of 2013.

Comment 18: *Page 13 line 317: You need a zonal map to show how the water partition changes with altitude and not the total column.*
Response 18: Most of the SOA and ALW_{org} retain in the lower levels of the troposphere. The information on altitude variation may not be very useful. Thus, no changes were made regarding this comment. We have also deleted this sentence in the revised manuscript to avoid confusion.

Comment 19: *Page 13, lines 322-336: It is not clear how you calculate the κ_{org} in your model. This is very important for this section.*
Response 19: We explained this in the revised text in L214-216:

“**Since ALW_{org} in this study is calculated mechanistically using the partitioning theory, κ_{org} can be estimated by rearranging Eq3:**

$$\kappa_{org} = \frac{ALW_{org}}{\rho_w V_{org}} \times \frac{1 - a_w}{a_w} \quad \textbf{(Eq4)}$$

”

Comment 20: *Figure 1: The quality of the figure is poor. It is extremely difficult to see all the plotting data and the changes due to the use of different scenarios (especially in figure 1b).*

Response 20: We removed the results of BS since they are very similar to those of S3. Now the figure has been revised as follows:

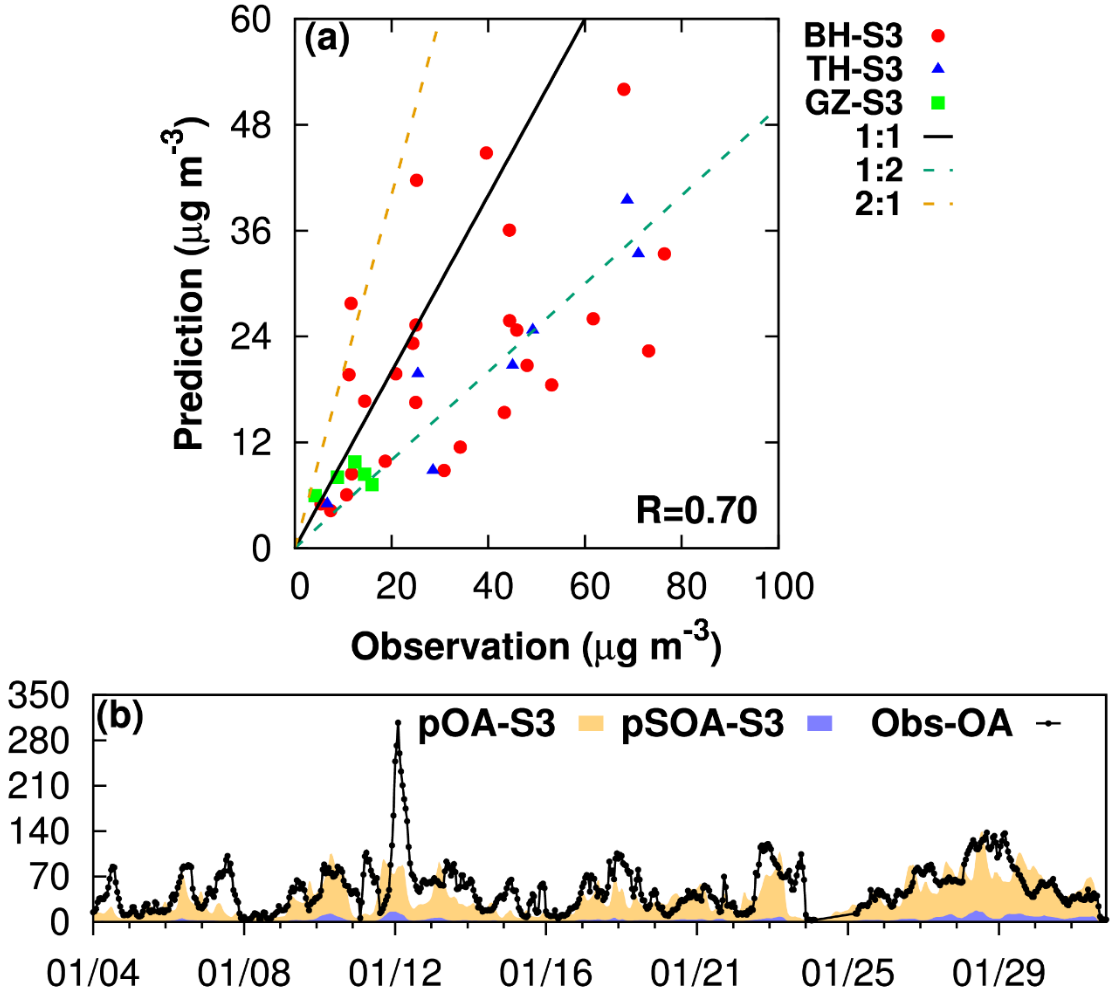

Figure 1. Comparison of (a) observed and modeled organic carbon concentration at University of Beihang (BH), Tsinghua University (TH) and Guangzhou (GZ); (b) observed organic aerosol (Obs-OA) at Beijing and predictions of total OA (pOA) and SOA (pSOA), unit is μg m-3. Locations of monitoring sites are shown in Figure S1.

Comment 21: *Figure 2: I found the use of daily maximum concentration in the* *“difference” maps misleading. Given that you have the monthly average SOA from the basecase simulation, I would prefer to see the absolute (and relative) change of the monthly average SOA due to the use of S3 as well, and not the daily maximum. Furthermore, please add (a), (b), etc. to each subplot of the figure and add this information to the figure caption (apply this change in the rest of the figures as well).*

Response 21: The monthly-averaged daily maximum differences have been replaced by the monthly-averaged differences for all the corresponding figures to reflect the general impacts on SOA and ALW_{org}. Each panel of the figure is labeled in sequence.

Comment 22: *Figure 4: All the fitted correlations listed here suggest that compounds with very low or zero O:C have negative hygroscopicity. Can you comment on this limitation and include a discussion in the text?*

Response 22: The relatively low values of hygroscopicity for low O:C ratio might be due to the linear regression. We also did an exponential fitting for the two variables so that the hygroscopicity falls in the range of (0,1) and is positively correlated with O:C ratio. The text and figure 4 have been revised accordingly.

In L402-407:

“**In both seasons, κ_{org} approaches zero and negative values as O:C decreases, which might be due to the linear regression of κ_{org} and O:C. To avoid this, an exponential fitting of the two variables is performed so that κ_{org} falls in the range of (0,1) and is positively correlated with O:C. In this case, the fitted correlations are $\kappa_{org}=1-\exp(-(O{:}C/1.88)^{2.29})$ and $\kappa_{org}=1-\exp(-(O{:}C/1.06)^{4.50})$ for January and July of 2013, respectively.**”

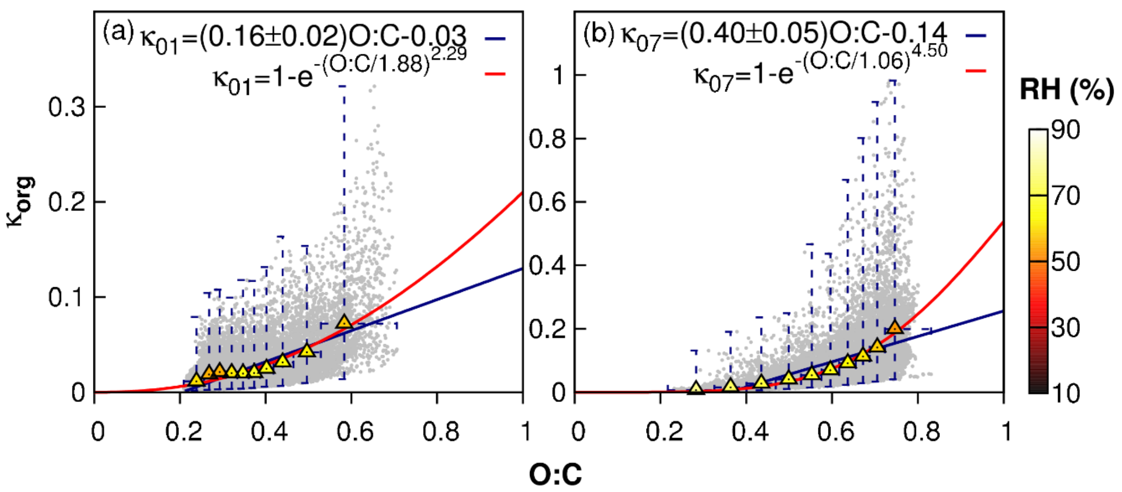

Figure 4. The correlation of hygroscopicity of organic aerosol (κ_{org}) and O:C ratio at 9 representative cities including Shenyang (SS), Beijing (BJ), Jinan (JN), Zhengzhou (ZZ), Xi’an (XA), Nanjing (NJ), Shanghai (SH), Chengdu (CD), and

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

Comment 23: *Table S3. Please explain in the table what the fraction in the last column stands for. How have you estimated the molecular weight and fraction of the POA from unknown compounds?*
Response 23: The last column is the molar fraction of each POA surrogate. We have clarified this in the first row of this column in the table. The molecular weight and molar fraction of unknown compounds of POA have been listed in the original table already, which are 390 and 0.3, respectively.

Minor Comments
Comment 24: *The language and structure of the sentences can be substantially improved in many parts of the manuscript. Just a few examples are listed here, but I suggest revising thoroughly the wording in the whole text.*
Response 24: We thank the reviewer for pointing this out. The whole text, as well as figures and tables, have been revised carefully.

Comment 25: *Page 5, lines109-11: Please rephrase. The sentence sounds wrong.*
Response 25: The text has been revised in L109-111 as follows:
"**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 26: *Page 5, lines117-118: Please rephrase.*
Response 26: The sentence has been revised in L116-117 as follows:
"**The total water content is the summation of water associated with each solute at the same water activity.**"

Comment 27: *Page 6 line 144: OC and OA abbreviations have not been used before in the main text.*
Response 27: OA abbreviation has been explained in a previous part of the revised manuscript in L120-122:
"**Pye et al. (2017) found that the modeled organic aerosol (OA) improved significantly but biased high at nighttime when ALW_{org} is included in the calculation.**"

Thus, this sentence has been revised in L139-141 as follows:

“**The model performance was evaluated against observed meteorological parameters (temperature and relative humidity, RH) as well as $PM_{2.5}$, organic carbon (OC), and OA at ground monitoring sites.**”

Comment 28: *Page 8, line 204: Please correct the “can estimated” to “can be estimated”.*
Response 28: The text has been revised as instructed.

Comment 29: *Page 8 line 206: I would use the word “correlate” instead of “dependent”*
Response 29: The text has been revised in L219-229 as follows:

“**In many studies, κ_{org} is assumed to increase linearly with the oxidation state of OA, expressed as the O:C ratio (Massoli et al., 2010;Duplissy et al., 2011;Lambe et al., 2011).**”

Comment 30: *Page 9 line 229: Change “in” with “on”*
Response 30: The text has been revised as instructed.

Comment 31: *Page 10 line 254: Change the sentence to: “In Beijing and Guangzhou, these impacts are not significant during winter”*
Response 31: The sentence has been revised in L298-300 as:
“**No significant differences in OC are observed in S3 compared to BS (not shown), likely due to the biased-low SOA predicted in the current model so that limits the impact of ALW_{org} on SOA formation.**”

Comment 32: *Page 11 line 274-275: Please rephrase.*
Response 32: The sentence has been revised as follows:
“**The criteria of MFB and MFE followed recommendations by Boylan and Russell (2006).**”

Comment 33: *Page 11 line 282: Which two areas? You have mentioned several areas in the previous sentence.*
Response 33: The sentence has been revised as follows:
“**Monthly-averaged SOA concentrations in the above areas are up to 25 and 15-20 μg m^{-3}, respectively.**”

Comment 34: *Page 13 line 317: “column water”. Please rephrase*
Response 34: The text has been revised in L378 as follows:
“**Based on the column concentrations of ALW_{org} and ALW_{org}/SOA ratio (Figure S8),**”

References

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Impacts of water partitioning and polarity of organic compounds on secondary organic aerosol over Eastern China

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Abstract

Secondary organic aerosol (SOA) is an important component of fine particular matter ($PM_{2.5}$~~) in China~~). Most air quality models use an equilibrium partitioning method along with ~~estimated~~the saturation vapor pressure (SVP) of semi-volatile organic compounds (SVOCs) to predict SOA formation. However, ~~this method ignores~~ the models typically assume that the organic particulate matter (OPM) is an ideal mixture and ignore the partitioning of water vapor to ~~the organic aerosols and the organic phase non-ideality, both of which affect the partitioning of SVOCs.~~OPM. In this study, the Community Multi-scale Air Quality model (~~CMAQv5.0.1) was used~~ CMAQ) is updated to investigate the ~~above impacts~~ impacts of water vapor partitioning and non-ideality of the organic-water mixture on SOA formation during winter (January) and summer (July) of 2013 over eastern China. The ~~organic aerosol module was~~ updated ~~by incorporating water~~model treats the partitioning of water vapor molecules into OPM and uses the UNIFAC model to estimate the activity coefficients of species in the organic ~~particulate matter (OPM) and considering non-ideality of organic~~-water mixture. The modified model can generally capture the observed surface organic carbon (OC~~), the total organic aerosol ()~~ with a correlation coefficient R of 0.7, and the surface OA~~) and diurnal variation of $PM_{2.5}$ at ground sites.~~ with the mean fractional bias (MFB) and mean fractional error (MFE) of -0.28 and 0.54, respectively. SOA concentration shows significant seasonal and spatial variations, with high ~~concentration levels~~concentrations in the North China Plain (NCP), Central China and Sichuan basin (SCB) ~~areas~~regions during winter (up to 25 μg m^{-3}) and in the Yangtze River Delta (YRD) during summer (up to ~~12~~16 μg m^{-3}). ~~When water partitioning is included in~~ In winter, SOA ~~concentrations increase~~ decreases slightly in the updated model, with the monthly-averaged ~~daily maximum~~ relative ~~difference~~change of 10-20% in the highly concentrated areas, mainly due to organic-water interactions. The monthly-averaged concentration of SOA increases greatly in summer, by 20-50% at the surface and ~~10-~~30~~% for~~-60% in the whole column~~, mostly~~. The increase of SOA is mainly due to the increase in biogenic SOA in inland areas and anthropogenic SOA. The increase in SOA is more significant in summer, by 20-90% at the surface and 30-70% for the whole column. The increase of SOA over the land is mostly due to biogenic SOA while the increase of SOA over the in coastal regions is related with that of anthropogenic origin. Further analysis of two representative cities, Jinan and Nanjing, shows that changes of SOA are favored under hot and humid conditions. The increases in SOA cause a 12% elevation in theareas. As a result, the averaged aerosol optical depth (AOD) is increased by up to 10% and 15% enhancement in the cooling effectseffect of aerosol radiative forcing (ARF) is enhanced by up to 15% over YRD in summer. The aerosol liquid water content associated with OPM (ALW_{org}) at the surface is relatively high over the landin inland areas in winter and over the ocean in summer, with the monthly-averaged daily maximumconcentration of 2-90.5-3.0 and 5-127 μg m^{-3}, respectively. By using the The hygroscopicity parameter (κ) of OA based on the κ-Köhler theory, we calculated the hygroscopicity of OA with is determined using the modeled ALW_{org}, finding that the. The correlation of κ with O:: C ratio varies significantly across different cities and seasons. Water Analysis of two representative cities, Jinan (in NCP) and Nanjing (in YRD), shows that the impacts of water partitioning and non-ideality of the organic-water mixture on SOA are sensitive to temperature, relative humidity (RH), and the SVP of SVOCs. The two processes exhibit opposite impacts on SOA in eastern China. Water uptake increases SOA by up to 80% in the organic phase, while including non-unity activity coefficients decreases SOA by up to 50%. Our results indicate that both water partitioning into OPM only promotes SOA formation, while non-ideality of organic-water mixture only leads to decreases in SOA in most regions of eastern China. Water partitioning into OPMand the activity coefficients of the condensed organics should be considered in air quality models in simulating SOA formation from gas-particle partitioning, especially in hot and humid environments.

Keywords: SOA, non-ideality, water partitioning, hygroscopicity

1 Introduction

Secondary organic aerosol (SOA) is formed via a complex interaction of volatile organic compounds (VOCs) with oxidants and primary particles emitted from anthropogenic and biogenic sources in the atmosphere. As an important component of fine particular matter ($PM_{2.5}$), SOA can cause severe air pollution in urban and suburban areas (Huang et al., 2014) and exhibit adverse health effects ~~(Polichetti et al., 2009;Feng et al., 2016;Xing et al., 2016;Atkinson et al., 2014).~~(Atkinson et al., 2014). SOA also plays an important role in new particle formation and particle growth ~~(Man et al., 2015;Zhang et al., 2011;Wiedensohler et al., 2009;Yue et al., 2011;Liu et al., 2014;Ehn et al., 2014;Huang et al., 2019;Jokinen et al., 2015) and further contributes~~that further contribute to the enhancement of cloud condensation nuclei (CCN) ~~(Yue et al., 2011;Wiedensohler et al., 2009;Liu et al., 2014;Jokinen et al., 2015)~~(Wiedensohler et al., 2009; Ehn et al., 2014). This will, in turn, impact the atmospheric aerosol burden, precipitation and water circulation, solar radiation budget, and climate ~~(Rosenfeld et al., 2008;Spracklen et al., 2011;Quaas et al., 2008;Ramanathan et al., 2001;Hatzianastassiou et al., 2007;Hegerl et al., 2015). However, the mechanisms of these~~(Ramanathan et al., 2001). However, the extents of those influences are not well understood so far, due to the high uncertainties associated with the formation and physical and chemical properties of SOA ~~(Shrivastava et al., 2017)~~(Shrivastava et al., 2017). Large gaps still exist in SOA mass ~~loadings~~loading and properties between ~~model estimates~~models and ~~laboratory and field measurements~~observations ~~(Gentner et al., 2017;Ervens et al., 2011;Hayes et al., 2015)~~(Gentner et al., 2017; Ervens et al., 2011; Hayes et al., 2015). Therefore, it is crucial to explore and resolve this issue to improve our knowledge of the roles of SOA in the environment, human health, and climate.

Gas-particle partitioning of semi-volatile and low-volatile organic compounds (SVOCs and LVOCs) generated from VOC oxidation is an important pathway of SOA formation. In most current chemical transport models (CTMs), this process is treated as an equilibrium partitioning process that depends on the mass concentration of the organic particulate matter (OPM), ambient temperature~~,~~ (T), the mean molecular weight of OPM, and the volatility of~~ purer~~ condensed organics ~~(Pankow, 1994)~~(Pankow, 1994). The ~~volatilities~~formation of condensed organic products ~~from a certain precursor VOC are either~~is commonly represented by ~~that of several~~ lumped ~~surrogates based on~~surrogate SVOCs in a 2-product model with volatilities and SVOC yields fitted to chamber experiments~~ (2 product model)~~ ~~(Odum et al., 1996)~~(Odum et al., 1996)~~ or fitted into different bins of a fixed volatility range (usually 0.01-10^5 μg m^{-3}) (volatility basis set model, VBS model) (Donahue et al., 2006). Although the above~~. To better represent the volatility of primary organic aerosol (POA) and the multi-generation oxidation of SVOCs to a wider range, Donahue et al. (2006) proposed the volatility basis set (VBS) model in which the mass yields of SVOCs are fitted to a fixed number of volatility bins (usually 0.01-10^5 μg m^{-3}). The VBS model has been adopted by several CTMs (such as WRF-Chem, GEOS-Chem, etc.).

Although the models can capture the general trend of SOA evolution and mass concentration to some extent ~~(Slowik et al., 2010;Li et al., 2017a;Baek et al., 2011;Bergström et al., 2012;Woody et al., 2016;Heald et al., 2006)~~(Li et al., 2017a; Bergström et al., 2012; Woody et al., 2016)~~, both of them neglected~~ , two key factors ~~that~~currently neglected in models may lead to biases: 1) the molecular structures and interactions of functional groups (-OH, -C=O, -COOH, etc.) of condensed organics (non-ideality); 2) partitioning of water vapor, ~~an~~the most abundant atmospheric constituent besides O_2 and N_2, to OPM. The non-ideality alters the volatility of condensed organics, and thus their contributions to the total SOA mass loading (Cappa et al., 2008). Water partitioning into OPM can reduce the partial pressure of organics ~~and lead~~due to~~ increase in SOA mass, which is called the~~ Raoult's Law effect ~~(Prisle et al., 2010)~~(Prisle et al., 2010)~~. This impact~~ and lead to increases in SOA mass. The amount of aerosol liquid water associated with organics (ALW_{org}) may vary for different ~~SOA~~ precursors ~~(Healy et al., 2009;Prisle et al., 2010)~~(Healy et al., 2009; Prisle et al., 2010). The above two aspects will not only affect the chemical composition of SOA but also the inorganic portion ~~(Ansari and Pandis, 2000;Meyer et al., 2009) and optical properties (Liu and Wang, 2010;Denjean et al., 2015)~~(Ansari and Pandis, 2000) and optical properties (Denjean et al., 2015) of aerosols.

Laboratory and field studies have ~~confirmed the fact that~~observed water absorbed by SOA ~~(quantified as hygroscopicity, κ)~~ from a variety of precursor VOCs ~~(Lambe et al., 2011;Zhao et al., 2016b;~~(Lambe et al., 2011; Zhao et al., 2016b; Asa-Awuku et al., 2010~~;Varutbangkul et al., 2006)~~; Varutbangkul et al., 2006). The hygroscopicity of SOA ~~is highly~~, quantitatively described by the hygroscopicity parameter, κ, is correlated with the oxygen-to-carbon ratio (O:C) and increases with more oxidized SOA during photochemical aging ~~(Poulain et al., 2010;Wang et al., 2014;Lambe et al., 2011;Tritscher et al., 2011a;Zhao et al., 2016b;Massoli et al., 2010;Tritscher et al., 2011b;Duplissy et al., 2011)~~(Lambe et al., 2011;Zhao et al., 2016b). The OPM-associated water ~~partitioning~~ can be estimated using the κ-Köhler theory under the Zdanovskii-Stokes-Robinson (ZSR) assumption of no interactions between any constitutes in aerosols ~~(Petters and Kreidenweis, 2007)~~(Petters and Kreidenweis, 2007). The total water content is the ~~summarization~~summation of water associated with each ~~constitute~~solute at the same ~~RH.~~water activity. Guo et al. ~~(2015)~~(2015) found that this simplified method, along with the ISORROPIA model which is used to predict aerosol liquid water (ALW) associated with the inorganic portion of aerosols, ~~reproduced~~could reproduce the observed total ALW in the ambient environment. Pye et al. ~~(2017)~~(2017) ~~applied this approach along with a parameterization of overall κ based on O:C ratio and a simplified method to estimate activity coefficients of organics and~~ found that the modeled organic aerosol (OA ~~and ALW are~~) improved ~~during daytime~~significantly but ~~still~~ biased ~~low~~high at nighttime~~.~~ ~~Shortcomings still exist in the above method for water associated organics (~~ when ALW_{org}~~)~~

is included in the calculation. However, as ~~interactions between~~ the interaction among organic species and between organics and water in the organic-water mixture ~~are not considered, which~~ has been shown to play an important role in SOA formation and water partitioning to OPM ~~(Kim et al., 2019)~~(Kim et al., 2019)~~. A representation of water partitioning along with SVOCs with consideration of water-organic and organic-organic interactions in CTMs showed significant influences in SOA and ALW in the eastern U.S. where biogenic SOA dominated in OA and the internal mixing assumed for the aerosol (Pankow et al., 2015;Jathar et al., 2016).~~

~~China has been suffering from severe $PM_{2.5}$ pollution especially in the eastern region with fast urbanization and economic development~~ , the ALW_{org} estimated by the κ-Köhler theory and its impact on SOA might not be accurate. 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 ~~(Guo et al., 2014;Fu and Chen, 2017;Yang et al., 2016)~~(Pankow et al., 2015; Jathar et al., 2016)~~. The secondary portion has been proved to be dominated in $PM_{2.5}$ and organic aerosol increases during haze events (Huang et al., 2014;Sun et al., 2019). In addition,~~.

China has been suffering from severe $PM_{2.5}$ pollution especially in the eastern region with fast urbanization and economic development (Fu and Chen, 2017). SOA is a very important component of $PM_{2.5}$ in China that contributes about 20-50% (Li et al., 2017b). The fraction of SOA in OA increases during haze events (Huang et al., 2014; Sun et al., 2019). Previous modeling studies ~~indicated~~in China indicate that SOA was underpredicted ~~in this region (Wang et al., 2011;Lin et al., 2016;Jiang et al., 2012)~~(Lin et al., 2016; Jiang et al., 2012) and the impacts of the non-ideality and water-OPM partitioning on modeled SOA have not been evaluated.

In this study, regional simulations of SOA during January and July of 2013 over eastern China ~~under several scenarios~~ were conducted to investigate the seasonal variation of SOA due to water partitioning into OPM. ~~Model performances were firstly~~The model performance was evaluated against observed meteorological parameters (temperature and relative humidity, RH) as well as $PM_{2.5}$, organic carbon (OC~~,~~), and OA at ground monitoring sites. ~~Then, the~~The regional and seasonal impacts on SOA, ALW_{org}, and ~~water content~~properties of aerosols were quantified. ~~Factors related to the impacts on SOA,~~Lastly, influences of the results by several factors including ~~sources of precursors, chemical compositions and~~ meteorological ~~conditions were further~~ parameters, estimations of saturation vapor pressures (SVP) of condensed organics, and the individual impacts of ALW_{org} and non-ideality of the organic-water mixture on SOA prediction were analyzed. ~~Lastly, the impacts on aerosol optical properties and hygroscopicity were investigated.~~

2 Methodology

2.1 Model description

The Community Multi-scale Air Quality model (CMAQ v5.0.1) coupled with a modified SAPRC-11 was used in this study. 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)~~ products~~ from different groups of major precursors ~~(Ying et al., 2015); Secondly, heterogeneous formation of secondary nitrate and sulfate from NO_2 and SO_2 reaction on particle surface (Ying et al., 2014), and~~(Ying et al., 2015). Secondly, SOA from isoprene epoxydiols (IEPOX), methacrylic acid epoxide (MAE) and dicarbonyls through surface-controlled irreversible reactive uptake were added ~~(Li et al., 2015;Pankow et al., 2015)~~(Hu et al., 2017; Li et al., 2015; Liu et al., 2020; Ying et al., 2015)~~ were added; Thirdly~~. Thirdly, the heterogeneous formation of secondary nitrate and sulfate from NO_2 and SO_2 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)~~(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.

~~Two types of SOA were considered in the current model, "semi-volatile" (SV) portion that formed via equilibrium absorption-partitioning of SVOCs, and "non-volatile" (NV) portion that formed via direct oxidation of aromatics at low-NO_x, isoprene oxidation under acidic conditions, reactive uptake of dicarbonyls, IEPOX and MAE, and oligomers. The SV-SOA module mostly based on that of Pankow et al.~~ The SOA module mostly follows Pankow et al. ~~(2015)~~(2015) ~~with several updates in the treatment of primary organic aerosol (POA) by including it in the non-ideality calculation of the organic-water mixture. The mass distribution of SVOCs between the gas-phase and particle-phase follows the equation:~~

. Two types of SOA as traditionally treated in CMAQ were considered, "semi-volatile" (SV) portion that formed via equilibrium absorption-partitioning of SVOCs, and "non-volatile" (NV) portion that includes the oligomers and SOA formed via direct oxidation of aromatics at low-NO_x. 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. POA was treated as non-volatile and non-reactive. The mass distribution of SVOCs between the gas-phase and particle-phase follows the equation:

$$K_{p,i} = \frac{F_i}{M \cdot A_i}$$ (~~Eq 1~~Eq1)

where $K_{p,i}$ ~~(m^3~~ ($m^3 \mu g^{-1}$) is the gas~~/~~-particle partitioning constant for compound i, F_i(μg m^{-3}) is the concentration of species i in the particle~~-~~-phase, A_i~~(~~ (μg m^{-3}) is the concentration of species i in the gas~~-~~-phase, and M (μg m^{-3}) is the total mass concentration of the absorbing organic phase~~.~~ (i.e. OPM). The gas~~/~~-particle partitioning constant $K_{p,i}$ is dependent on the chemical composition of ~~the absorbing organic phase.~~OPM. Pankow et al. ~~(1994)~~(1994) derived $K_{p,i}$ for SVOCs partitioning into an absorbing organic phase as:

$$K_{p,i} = \frac{RT}{10^6 \overline{MW} \xi_i p^o_{L,i}} \quad \text{(~~Eq 2~~Eq2)}$$

where $p^o_{L,i}$(atm) is the ~~saturation vapor pressure~~SVP of the pure compound i at temperature T(K), ξ_i is the activity coefficient of species i in the absorbing organic phase, $\overline{MW}$(g mol^{-1}) is the average molecular weight of ~~the absorbing organic phase~~OPM, R (8.314 J mol^{-1} K^{-1}) is the gas constant, and 10^6 is used to convert the ~~unit~~units to $m^3 \mu g^{-1}$.

There are 12 lumped SVOCs generated by oxidation of alkanes, alkenes, and aromatics ~~oxidized~~ under different NO_x conditions ~~(~~and 8 NV organic products as listed in Table S1~~).~~ and Table S2. More details about the lumped precursors such as formation conditions (“high” and “low” NO_x), lumping species and method, and yields from parent VOCs can be found in Carlton et al. (2010) and summarized in SI. Activity coefficients of SVOCs were calculated based on the composition of ~~absorbing organic phase~~OPM using the ~~UNIversal Functional Activity Coefficient (~~UNIFAC~~) method (Fredenslund et al., 1975),~~ method, with assigned carbon number (n_c), functional groups and energy interaction parameters to both SV and NV compounds ~~(Pankow et al., 2015)~~(Pankow et al., 2015). The UNIFAC model is one of the commonly used models that activity coefficients of condensed organics and their interactions with water can be estimated. This method has been adopted to investigate the impacts of non-ideality and water~~-OPM~~ partitioning into OPM on SOA for different precursors in box models ~~(Seinfeld et al., 2001;Bowman and Melton, 2004)~~(Seinfeld et al., 2001; Bowman and Melton, 2004) and CTMs ~~(Jathar et al.,~~

~~2016;Pankow et al., 2015;Kim et al., 2019)~~(Pankow et al., 2015; Kim et al., 2019). ~~The primary organic aerosols (~~In the current model, POA~~)~~ was assumed to have a bulk composition of ten categories of surrogate species (Table S3), as used by Li et al. ~~(2015)~~(2015). POA is also involved in the calculation of activity coefficients for the ~~organics in the condensed phase.~~organic-water mixture. Detailed information about the surrogate species including ~~the~~their structures and properties can be found in Li et al. ~~(2015)~~(2015) and references therein.

In addition to organic compounds, water partitioning into OPM is enabled according to Eq 1 and Eq 2. In such a case, the absorbing phase in Eq 1 includes both organic aerosols and water ~~partitioning into~~associated with OPM. As water ~~considered~~condenses in the absorbing organic phase, it will further alter the molar fraction of each composition, the activity coefficient of SVOCs and the SV-SOA mass concentrations as a result. In the current model, we assumed no interactions between the inorganic and organic phases.

~~As the water partitioning into OPM is highly correlated with the hygroscopicity of aerosols (κ), their correlation can be expressed by the κ-Köhler theory with Kelvin effect neglected (Peter et al., 2006):~~

2.2 Estimating the hygroscopicity of OA

Based on the κ-Köhler theory with linearly additive hygroscopic behavior of each component of the mixed particle, ALW_{org} is related to the hygroscopicity parameter for the organic mixture (κ_{org}) by Eq3 (Petters and Kreidenweis, 2007):

$$ALW_{org} = V_{org} = \rho_w V_{org} \kappa_{org} \frac{a_w}{1 - a_w} \quad \text{(Eq3)}$$

where ρ_w is the density of water (assumed to be 1 g cm^{-1}), V_{org} is the volume concentration of ~~organic~~organics, and a_w is the water activity (assumed to be the same as RH). ~~Taken~~Since ALW_{org}

in this study is calculated mechanistically using the partitioning theory, κ_{org} can be estimated by rearranging Eq3:

$$\kappa_{org} = \frac{ALW_{org}}{\rho_w V_{org}} \times \frac{1-a_w}{a_w} \quad \text{(Eq4)}$$

V_{org} can be estimated from the modeled mass concentration of OA, assuming the density of organic aerosolOA to be 1.2 g cm^{-3} (Li et al., 2019), the hygroscopicity of the total OA can estimated. This simplified method can be used to estimate OPM associated water .

In many studies, κ_{org} is assumed to increase linearly with the oxidation state of OA, expressed as the O:C ratio (Guo et al., 2015;Li et al., 2019)(Massoli et al., 2010;Duplissy et al., 2011;Lambe et al., 2011). In addition, the hygroscopicity of organic aerosol is dependent on the degree of oxygenation, showing a positive linear relationship with the O:C ratio (Massoli et al., 2010;Duplissy et al., 2011;Lambe et al., 2011;Hong et al., 2018;Li et al., 2019). We therefore estimated the correlation of κ and O:C ratio at 9 representative cities during January and July. The correlation of κ_{org} and O:C ratio at 9 representative cities was evaluated during January and July of 2013, with the reduced major axis regression method (Ayers, 2001). The O:C ratio of the total OA was calculated as followingusing Eq5:

$$O{:}C = \sum_{i=1}^{n} f_i\,(O{:}C)_i \quad \text{(Eq4Eq5)}$$

where f_i and $(O{:}C)_i$ are the molar fraction and O:C ratio of organic aerosol component i. For POA, a fixed molar fraction and composition has beenwere assumed following Li et al. (2015)(2015). For SOA, the O:C ratio was estimated by their OM:OC ratio (Simon and Bhave, 2012):

. For SOA, the O:C ratio was calculated by using their organic matter to organic carbon ratio (OM:OC) following Simon and Bhave (2012):

$$\text{O:C} = \frac{12}{15} = \frac{12}{15}(\text{OM:OC}) - \frac{14}{15} \quad \text{(Eq5Eq6)}$$

OM:OC ratio of each SOA component follows Pye et al. (2017).

The OM:OC ratio of each SOA component follows Pankow et al. (2015) as shown in Table S1-S2.

2.3 Model application

The simulation domain has a horizontal resolution of 36 km × 36 km (100 × 100 grids) and a vertical structure of 18 layers up to 21 km, which covers eastern China as shown in Figure S1. Anthropogenic emissions were generated from the Multi-resolution Emission Inventory for China (MEIC) ~~(Zhang et al., 2009;Li et al., 2014;Zheng et al., 2014;Liu et al., 2015)~~(Zhang et al., 2009) v1.0 with a 0.25° × 0.25° resolution (http://www.meicmodel.org) for China, and the Regional Emission inventory in Asia version 2 (REAS2) (Kurokawa et al., 2013) with a 0.25° × 0.25° resolution (http://www.nies.go.jp/REAS/) for the rest of the domain. Biogenic emissions were generated by the Model for Emissions of Gases and Aerosols from Nature (MEGAN) v2.1, with the leaf area index (LAI) from the 8-~~—~~day Moderate Resolution Imaging Spectroradiometer (MODIS) LAI product (MOD15A2) and the plant function types (PFTs) from the Global Community Land Model (CLM 3.0). Open biomass burning emissions were generated from the Fire INvnetory from NCAR (FINN) ~~(Wiedinmyer et al., 2011)~~(Wiedinmyer et al., 2011). Dust and sea salt emissions were generated ~~in line~~online during CMAQ simulations. The total emissions of major SOA precursors and their spatial distributions are shown in Table S4 and Figure S2. Meteorological fields were generated using the Weather Research and Forecasting (WRF) model v3.6.1 with initial and boundary conditions from the NCEP FNL Operational Model Global Tropospheric Analyses dataset. More details about the model application can be found in Hu et al. (2016).

Four scenarios are investigated in this study. The base case (BS) ~~that applied~~applies the default secondary organic aerosol module of CMAQ~~; the water~~ v5.0.1. In this case ~~(S1) that only~~, no water partitioning into OPM ~~was~~is considered~~; the UNIFAC case (S2) that effects~~. Lumped semi-volatile products from the oxidation of ~~molecular structure of the primary and secondary~~various precursors partition into a single organic ~~species were included;~~phase, which is considered as an ideal mixture of POA and SOA with γ_{org}=1. The water case (S1) includes water partitioning into OPM, which is again considered as an ideal solution (γ_{org}=1 and the$\gamma_{H2o} = 1$). The UNIFAC case (S2) considers the interaction between organic constituents with UNIFAC calculated activity coefficients (γ_{org}≠1) but does not allow water partitioning into OPM. The combined case (S3) thatallows both water partitioning and interactions between all constituents (including water and organics) using UNIFAC calculated activity coefficients (γ_{org} ≠1 and $\gamma_{H2o} \neq 1$). The results of BS and S3 are used to examine the overall impacts of water partitioning into OPM and polarity of organics on SOA and ALW_{org}, as shown in Section 3.1-3.4. The separate influences of those two processes on SOA from S1 and S2 were combined together. are discussed in Section 3.5.

3 Results and discussion

3.1 Model evaluation

TemperatureThe meteorological inputs and relative humidity (emissions have been used in several previous publications. Model performance on meteorological parameters (temperature and RH)), gaseous species and gas and aerosol concentrations have been extensively evaluated (Hu et al., 2016; Hu et al., 2017; Qiao et al., 2018; Shi et al., 2017). A summary of the model performance related to this study is provided below. Observed meteorological data were obtained from the National Climatic Data Center (ftp://ftp.ncdc.noaa.gov/pub/data/noaa). Observations of OC at two urban locations, Beijing (Cao et al., 2014; Wang et al., 2015) and Guangzhou (Lai et al., 2016) and OA in Beijing (Sun et al., 2014) during January of 2013 as well as surface $PM_{2.5}$ at several monitoring sites during July of 2013 from China National Environmental Monitoring Center (http://113.108.142.147:20035/emcpublish/) were used to evaluate model estimates of aerosols. Details of measurement methodology and uncertainties of observations are listed in the corresponding references.

Temperature and RH are the two meteorological factors that affect SOA formation. Table 1 shows the comparison of lists model statistics of mean observation (OBS), mean prediction (PRE), mean bias (MB), gross error (GE) and correlation coefficient (R) based on WRF predictions and observations at monitoring sites located 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) 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 WRFthe 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. Model estimates of daily organic carbon (OC) from the BS case wereS3 are compared with measurements at monitoring sites in Beijing and Guangzhou during the winterin January of 2013 (Figure 1(a)). Overall, 1a). The factors used to convert SOA to OC (SOC) are listed in Table S1-S2. OC from POA (POC) is directly predicted by the model. Generally, the ratio between modeled and observed OC concentration falls in the range of 1:2 to 2:1, with a correlation coefficient R of 0.70. The model tends to underestimate OC, especially in Beijing on highly polluted days (by -37~48%). No significant improvements to modeled OC were observed in S3. The impacts of water co-condensation and polarity of organic condensed species on SOA exhibit strong seasonal and spatial features, which are further discussed in Section 4. The impacts in Beijing and Guangzhou are not significant during winter. an R-value of 0.7. The model tends to underestimate OC on high concentration days. Overall, the mean fractional bias (MFB) and mean fractional error (MFE) of OC are -0.20 and 0.27, within the criteria (MFB$\leq \pm$0.6; MFE$\leq$0.75) suggested by EPA (2007). The bias in OC might be due to under-estimatedunderestimated POA emissions and under-predictedunderpredicted SOA in CMAQ from missing precursors (Hu et al., 2017;Zhao et al., 2016a); Zhao et al., 2016a).

~~The model estimate of OA was further investigated. As~~ . No significant differences in OC are observed in S3 compared to BS (not shown~~ in~~), likely due to the biased-low SOA predicted in the current model that limits the impact of ALW_{org} on SOA formation.

The underestimate of SOA can be seen from Figure ~~1(b),~~1b as well. CMAQ can well capture the observed diurnal variation of OA ~~at~~in Beijing during wintertime, except for the underestimates of peak values. ~~A better agreement~~ The correlation coefficient of modeled to observed OA is 0.55. We find a 25% underestimate of OA on average. Better agreement between the model and ~~the~~ observations is ~~observed~~shown on non-polluted days (daily-averaged concentration less than 75 μg m^{-3}). ~~The monthly-averaged mean fractional bias (MFB) and mean fractional error (MFE) are -0.13 and 0.27, respectively. POA is~~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 \pm$0.6; MFE$\leq$0.75) suggested by EPA (2007). 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~~, accounting for 88% due to aging of POA not treated in the current model. The fraction of SOA is small, resulting in little impacts on SOA by~~ 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 0.83 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 ~~insignificant improvements~~non-ideality of the ~~modeled OA~~organic-water mixture on SOA will be evaluated in ~~S3.~~ a future study.

Due to the lack of observed OC and OA in July of 2013, as an alternative, model performances are evaluated by comparing predicted and observed $PM_{2.5}$ at ground sites (Figure ~~S2 shows the comparison of modeled and observed $PM_{2.5}$ at monitoring sites~~ S1) as shown in Figure ~~S1 (a) during July of 2013.~~S3. Generally, ~~our~~the model can well reproduce the diurnal variation of $PM_{2.5}$ in most regions. Predicted $PM_{2.5}$ on ~~high concentration~~highly concentrated days ~~are~~is biased low~~ compared to observations~~, especially in the North ~~Central~~China Plain (NCP). ~~The~~ NCP ~~region~~ has the highest $PM_{2.5}$ ranging from 60 μg m^{-3} to 300 μg m^{-3}~~ compared to other regions.~~. The bias in modeled $PM_{2.5}$ is significant in cities in ~~the~~ Northwest. This might be due to missing dust emissions in the current inventory (Hu et al., 2016). To further evaluate the model performance, ~~statistics of~~ averaged MFB and MFE ~~were~~of modeled $PM_{2.5}$ are plotted against ~~observed $PM_{2.5}$ concentration at all monitoring sites (~~observations of each site as shown in Figure ~~S3).~~S4. The criteria of MFB and ~~goal~~MFE followed recommendations ~~of~~by Boylan and Russell ~~(2006)~~(2006). Our model ~~performed~~performs well ~~as~~since most of the predictions meet the criteria and a large fraction (>58%) meet the goal. The averaged MFB and MFE across all the sites are -0.28 and 0.39~~ respectively~~, indicating slightly underestimate of $PM_{2.5}$ by the model.

3.2 Impacts ~~of water partitioning~~ on SOA and ALW_{org}

~~Distribution~~The spatial distribution of SOA ~~varied~~varies greatly in the two seasons. In winter, SOA is relatively high in eastern SCB and ~~in~~ the ~~contiguous areas~~central and eastern provinces of Shandong, Henan, Anhui, and Hubei ~~provinces~~ (Figure 2 and Figure ~~S4~~S5). Monthly-averaged SOA concentrations in the above ~~two~~ areas are up to 25 and 15-20 μg m^{-3}, respectively. The major precursors of SOA Anthropogenic emissions are originated from anthropogenic the major sources of SOA (Figure S6), such as dicarbonyl products of aromaticsfrom the oxidation, xylenes of xylene and toluene (Figure S5). (Hu et al., 2017). In summer, surface SOA is high in NENortheast, NCP, and YRD regions. Shanghai, Jiangsu province and coastal areas of Yellow Sea show the . The highest SOA of ~9-12 μg m^{-3} occurs in Shanghai and Jiangsu provinces as well as the coastal area of the Yellow Sea, with the value of ~9-16 μg m^{-3} at the surface and ~20-25 mg m^{-2} asin the column total (col-SOA) inof the atmosphere below 21 km (Figure S4S5). Different from winter SOA, a significant fraction of summer SOA is originated from biogenic emissions in Shanghai and Jiangsu province (Figure S5S7). Anthropogenic SOA is high highly concentrated in July inthe coastal areas of the Yellow Sea and Bohai Bay.

Combined effects of water partitioning into OPM and non-ideality on SOA formation (S3) also exhibit strong seasonal variation. In winter, the increase of SOA is relatively small,slightly decreased by ~1-4.5 μg m^{-3} (10-20%) on average at the surface (Figure 2) and less than ~51 mg m^{-2} (10-30%) as for 20%) in the column concentration (Figure S4). The influences on SOA also differ in different altitudes. For example, the maximum increment at the surface is observed in Shandong province in NCP (Figure 2), while SOA at higher levels of the atmosphere is more significant in South China (Figure S4). The increase in SOA is mostly attributed to anthropogenic sources in winter (Figure S5 and S7).) over high SOA regions where anthropogenic sources dominate. We show later that the decrease of SOA is mainly due to the large activity coefficients of SVOCs which decrease $K_{p,i}$. In summer, higher temperature and relative humidity (RH) promote water partitioning and SOA formation as well as water partitioning into OPM. At the surface,so that SOA increases by 3-9 μg m^{-3} (40-50%) in coastal areas and 2-9 μg m^{-3} (20-90%) over the land, which are dominated by anthropogenic and biogenic origin, respectively (Figure S6). For col-SOA, in addition to coastal areas, more significant increase is observed in YRD, most of Henan province, and the contiguous areas of Hubei, Hunan, and Jiangxi province (Figure S4) by about 30-70%.apparently over the entire domain, with the highest enhancement of 2-4 μg m^{-3} (20-50%) at the surface (Figure 2) and 4-6 mg m^{-2} (30-60%) in the column (Figure S5) over YRD and the coastal area of Yellow Sea. Anthropogenic SOA dominates the total change in winter as shown in Figure S6. In summer, the increase of SOA is attributed to biogenic sources in inland areas and anthropogenic sources over the ocean (Figure S7).

Regional distribution of water partitioning into OPM ALW$_{org}$ is similar to the changeschange of SOA. as shown in Figure 3 shows the regional distribution of monthly . In winter, a maximum averaged daily maximum ALW$_{org}$. We see up to 9concentration of 3.0 μg m^{-3} for ALW$_{org}$ at surface occurs in Shandong in winter where great increment inthe high SOA appears as well. In region, where significant changes of SOA also occur. In other areas, the averaged concentration of ALW$_{org}$ is about 2-60.5-1.5 μg m^{-3}. TheOverall, the average ratio of ALW$_{org}$ to SOA is about 0.1-0.53 in winter. In summer, water partitioning into OPM mostly involvesoccurs in the east coastal areasarea at the surface where a significant increase of anthropogenic SOA (such as those from toluene and xylenes) is observed. This might be due to the high polarity of anthropogenic SVOCs (having more -COOH groups) that absorb more water. In the coastal areas, ALW$_{org}$ is about 5-12 μg m^{-3}, with a ratio to SOAarea, the averaged concentration of 0.3-0.6. ALW$_{org}$ over the land is about 25-7 μg m^{-3}, with the ALW$_{org}$ /SOA ratio of 0.5-1.0. Over the land, the averaged concentration of ALW$_{org}$ is about 1-3 μg m^{-3} (ALW$_{org}$/SOA ratio of 0.12-0.45) in most areas, whichNortheast and East China. Water partitioning is mostly associated with the increase of BSOA such as biogenic SOA originated from isoprene and monoterpenes oxidation that produces SVOCs with abundant OH group in SVOCs. The highest.

Based on the column concentrations of ALW$_{org}$ is 16 μg m^{-3} near Shanghai (and ALW$_{org}$/SOA ratio of 0.57). Water partitioning also varies at different altitudes (Figure S9). In (Figure S8), in winter, more column water partitions into OPM (col-ALW$_{org}$) in Chongqing, Hunan, Guanxi, Guangdong and Guizhou province, with theALW$_{org}$ must have occurred in the south and southwest regions at higher levels where a significant increase of col-SOA occurs (Figure S5). The averaged col-ALW_{org}/col-SOA ratio ~~of~~ in the high SOA area is 0.~~2~~1-0.3. In summer, the ALW_{org} must be high at higher altitudes over the central regions in China. The maximum col-ALW_{org} is ~~predicted~~about 7 mg m^{-2} over~~ the land, especially in~~ YRD, with the col-ALW_{org}/col-SOA ratio of ~~0.1~~ about 0.3~~ over eastern China~~.

~~Figure 4 shows~~3.3 Impacts on aerosol properties

Since ALW_{org} is determined in S3, the ~~correlation~~values of κ_{org} ~~with~~can be estimated from the modeled ALW_{org}, OA and RH using Eq 4. 9 representative cities were selected to investigate the relationship of κ_{org} vs. O:C ~~ratio.~~and its seasonal variation as shown in Figure S9 and S10. The results of all the cities in winter and summer are merged and analyzed as shown in Figure 4. Pairs of κ_{org} and O:C data are grouped into 10 O:C bins and the averaged κ_{org} in each bin are then calculated. Overall, the estimated O:C ratio is within the range of 0.2-0.~~6. In summer, the oxidation state of OA shows different degrees of enhancement compared to winter at most of the cities except Guangzhou, due to increased contribution of SOA to total OA. The averaged κ_{org} of OA in each O:C bin falls in the range of 0.001-0.1~~8. The averaged κ_{org} in each O:C bin is less than 0.1 in winter, with the highest ~~κ_{org} (~0.3) at Beijing~~value in Guangzhou. As more ALW_{org} is formed in summer~~.~~, the averaged κ_{org} also increases greatly with the highest value of 0.35 in Beijing. The linear correlation between κ_{org} and O:C shows significant spatial and seasonal variations. For example, the slope of κ_{org}-O:C is ~~much~~70-90% smaller in winter ~~(45-74% less)~~ than in summer in the Northern cities such as Shenyang, Beijing, Zhengzhou, and Xi'an~~, while~~. However, in Guangzhou, the slope ~~of κ_{org}-O:C in winter is much~~is 83% higher ~~(47-104% more)~~in winter than in summer~~ in the Southern cities, such as Nanjing,~~. In Chengdu~~ and Guangzhou. In Jinan and Shanghai~~, the slope is quite similar in both seasons. ~~The fitted correlations are very different from previous studies with a relatively higher~~Overall, the slope of κ_{org}~~-O:C from~~ vs. O:C in the 9 cities is 0.16 in winter and 0.40 in summer. Most of the fitted linear correlations of the individual city fall outside of the range of 0.18 to 0.37 suggested in previous studies ~~(Duplissy et al., 2011;Lambe et al., 2011;Massoli et al., 2010;Chang et al., 2010)~~(Duplissy et al., 2011;Lambe et al., 2011;Massoli et al., 2010;Chang et al., 2010), indicating that the hygroscopicity of organic aerosols ~~with chemical complexity~~ cannot be simply represented by a single parameter such as the O:C ratio ~~(Rickards et al., 2013)~~(Rickards et al., 2013)~~.~~

~~**3.3 Impacts on solar radiation**~~

. In both seasons, κ_{org} approaches zero and negative values as O:C decreases, which might be due to the linear regression of κ_{org} and O:C. To avoid this, an exponential fitting of the two variables is performed so that κ_{org} falls in the range of (0,1) and is positively correlated with O:C. In this case, the fitted correlations are κ_{org}=1-exp(-(O:C/1.88)$^{2.29}$) and κ_{org}=1-exp(-(O:C/1.06)$^{4.50}$) for January and July of 2013, respectively.

The impacts on aerosol optical depth (AOD) and aerosol radiative forcing (ARF) ~~were~~are further investigated. Figure 5 shows the monthly-averaged AOD at 550 nm in January and July of 2013. It ~~was~~is calculated ~~as~~by summarizing the ~~accumulation~~product of model estimated extinction coefficient of fine particles (~~EXT_i~~$b_{ext,i}$) multiplied by the thickness (HL_i) ~~of~~in each layer:

$$\mathrm{AOD} = \cancel{\sum_{i=1}^{N} EXT_i} \sum_{i=1}^{N} b_{ext,i} \times HL_i \qquad \text{(Eq6)}$$

~~Where~~where N is the number of layers. There are two methods to estimate the aerosol extinction coefficient in ~~CMAQ~~CMAQv5.0.1. One is ~~using~~based on the Mie theory ~~(EXT_m~~and the predicted aerosol component concentrations ($b_{ext,m}$), and the other is based on ~~extinction values~~correlation from the IMPROVE monitoring network that considers the impacts of hygroscopicity of different aerosol components (~~EXT_r)~~$b_{ext,r}$) ~~(Malm et al., 1994)~~(Malm et al., 1994). AOD calculated with the two types of extinction coefficient are denoted as AOD_m and AOD_r, respectively.

In Figure 5, a clear pattern of high AOD_r in SCB and NCP and low AOD_r in west China is observed in both winter and summer ~~is observed~~, consistent with previous studies

(He et al., 2019;He et al., 2016;Luo et al., 2014)(He et al., 2019; He et al., 2016; Luo et al., 2014). An identifiedidentical trend in AOD$_m$ is observed as shown in Figure S10S11. The monthly-averaged AOD$_r$ ranges from 1.10 to 3.52 in January and from 0.43 to 0.89 in July. AOD$_m$ is lower than AOD$_r$, falling in 0.7-2.2 in January and 0.3-0.6 in July. The model significantly overestimates AOD in January but agrees better with observations from MODIS in thewhere AOD is high regions in July (Figure S11). BiasesS12). The bias in the predicted AOD might be partially due to the empirical equation applied in the calculation of AOD in CMAQ (Wang et al., 2009;Liu et al., 2010)(Wang et al., 2009; Liu et al., 2010), and partially due to the uncertainties of fine AOD overland from MODIS data (Wang et al., 2009;Levy et al., 2010)(Wang et al., 2009; Levy et al., 2010). With water partitioning into OPM, changes in SOA mass concentration and chemical composition lead toThe increase of AOD, which due to ALW$_{org}$ shows a strong spatial and seasonal pattern. In winter, there isare no significant increasechanges in AOD$_r$ across the whole domain, due to insignificant changes of SOA. In summer, AOD$_r$ increases significantly in YRD and the adjacent area of Hubei, Hunan, and Jiangxi provinceareas by up to 1210%.

ARF represents the changeschange in the radiative flux at the top of the atmosphere due to aerosols. The off-lineAn offline version of the Shortwave Radiative Transfer Model For GCMs (RRTMG_SW) iswas used to calculate the direct radiative effect of aerosols on shortwave radiation (Iacono et al., 2008)(Iacono et al., 2008). Generally, fine aerosols exhibit cooling effects on the shortwave radiation in both winter and summer over the entire domain as shown in Figure 6. This impact is much stronger in the areas where AOD is high (Figure 5). The ARF at top of atmosphere (TOA) is highest in Shandong in winter and in the coastal areasarea near Jiangsu province in summer, which are about -125 W m^{-2} and -96 W m^{-2}, respectively. In winter, no significant changes of ARF are observed in the high regions of eastern China (Figure 66b). This is likely attributed to an insignificant contribution of SOA to PM$_{2.5}$ in winter compared to other components with cooling effects, such as sulfate. In summer, SOA is an important component of PM$_{2.5}$ (20-60%), and the effects of water partitioning on shortwave radiation ~~is~~are relatively stronger. An enhancement of up to 15% in the cooling effects of ARF occurs near the YRD region where AOD significantly changes as well.

~~4 Discussion~~

3.4 Sensitivity to T, RH, and SVP

Meteorological conditions and SOA precursors affect the impacts of water partitioning on SOA. ~~Figure 7 shows the effects of different factors on the daily maximum change of SOA in Jinan and Nanjing, two representative cities in winter and summer, respectively. As shown in Figure 7(a), the daily maximum elevation of SOA occurs when RH is greater than 70% in both cities. This is consistent with the previous study in the Southeast U.S. during summer (Pankow et al., 2015). A clear correlation of the changes in SOA with SOA concentration in Nanjing (R=0.84) during summer can be observed. However, this correlation is relatively weak in Jinan (R=0.44) during winter. There is no strong correlation between changes in SOA and temperature as shown in Figure 7(b), likely due to the daily variation of SOA mass and composition.~~ To better illustrate the dependency of SOA on temperature, RH, and ~~relative humidity~~SVP of SVOCs, an offline calculation of SOA formation was performed at two representative cities (Jinan in NCP during winter and Nanjing in YRD during summer) when the daily maximum SOA increases occurred. We ~~assumed~~assume temperature (T) and water vapor mixing ratio (QV) to be within the range of $\bar{X} \pm \sigma$, where $\bar{X}$ and σ are the mean and standard deviation calculated based on WRF ~~prediction~~predictions at each location. We ~~chose~~choose 10 evenly distributed values for T and QV within the range of $\bar{X} \pm \sigma$. The temperature dependence parameter of ~~saturation vapor pressure~~SVP (ΔH~~) was~~/R) is also scaled by 0.2, 0,8, 1.4, and 2.0 separately for all the SVOCs. As shown in Figure ~~8~~7, SOA ~~indicates~~exhibits a negative correlation with ~~temperature~~T and a positive correlation with ~~RH~~QV in both cities. SOA is more sensitive to ~~RH~~QV under ~~cool~~cold conditions (~~JN~~Jinan) and to temperature under hot conditions (~~NJ). An interesting finding is that significant increases in SOA~~ Nanjing). When the temperature is fixed, the sensitivity of SOA to $\Delta H/R$ is different in the two cities ~~occur during different time periods of the day. Water partitioning tends to affect SOA in the afternoon and evening in Jinan, which mostly happens in the early morning and at noon in Nanjing. The different timing is likely~~ . We find more changes in SOA across $\Delta H/R$ in Jinan. This is attributed to ~~a substantial increase in SOA precursors in the two cities. In Jinan, the most contributing SVOCs are originated~~the temperature correction factor (ζ_{corr}) of K_p in CMAQ as defined below:

$$\zeta_{corr} = \frac{K_{p,T_{ref}}}{K_{p,T}} = \frac{T_{ref}}{T} \exp\left[\frac{\Delta H}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right] \qquad \text{(Eq8)}$$

where T_{ref} is the reference temperature (298K) and T is temperature. According to Figure 7, the range of T is 265-274K in Jinan and 300-307K in Nanjing. The deviation of temperature from ~~toluene and xylenes oxidation, as well as oligomers formed by their oxidation products in OPM. Possible emission sources include transportation, petroleum refining, manufacturing, painting, etc. SOA increase in Nanjing is mostly associated with biogenic sources including isoprene and monoterpenes.~~the reference value (298K) is greater in Jinan than in Nanjing. Therefore, the unit change of $\Delta H/R$ causes greater variations of ζ_{corr} and thus K_p in Jinan. As a result, SOA is more sensitive to $\Delta H/R$. The impacts of SVP estimation on SOA are more significant in winter than in summer.

3.5 Separate impacts of water partitioning/polarity of condensed organics

Impacts of water partitioning into OPM and non-ideality of the organic-water mixture on SOA are in opposite directions. Water partitioning ~~alone~~ increases SOA by ~10-20~~-60~~% in winter and ~~~20-100~~30-80% in summer in most areas of the domain (Figure ~~S12~~8). This is because ~~that~~ the molecular weight of water is quite small and will reduce the molar averaged weight of OPM ($\overline{MW}$) in ~~Eq 2~~Eq2 ~~(Pankow et al., 2015)~~(Pankow et al., 2015). The reduced $\overline{MW}$ further increases ~~$K_{p,i}$ promoting the~~K_p and promotes mass transfer of SVOCs from the gas~~-~~-phase to ~~the~~ OPM. On the other hand, by considering non-ideality of the organic-water mixture, activity coefficients of SVOCs are usually greater than 1.0 in this study, leading to a decrease in ~~$K_{p,i}$~~K_p. As a result, the total SOA concentration is reduced by ~~up to~~ ~10-20% in winter and ~~~30~~10-50% in summer ~~in the high regions~~ (Figure ~~S13~~9). Overall, the final impacts are the combined consequences of the two ~~"~~processes~~"~~. In winter, the increase of SOA caused by water partitioning is offset by the decrease of SOA due to the polarity of SVOCs in most areas of the domain, resulting in ~~no significant changes.~~slight decreases of SOA. In summer, ~~effects~~the effect of water partitioning ~~overcome~~overcomes that of SVOC polarity so ~~as~~ the total SOA loading increases. This further leads to an enhanced attenuation of shortwave solar radiation and cooling of the atmosphere.

~~5 Conclusion~~

4 Conclusions

The WRF/CMAQ model was used to investigate the impacts of water partitioning into OPM and non-ideality of the organic-water mixture on SOA formation over eastern China during January and July of 2013. SOA is greatly enhanced in summer especially in YRD and over the Yellow Sea by up to ~~90~~50% and ~~70~~60% at the surface and in the whole column, respectively. No significant impacts on SOA are observed in winter. This might be due to the underestimation of SOA in the current model. ALW_{org} is highly correlated with the ~~changes~~change of SOA, with the ratio of ALW_{org} to SOA of 0.1-0.~~5~~3 and 0.~~2-~~1-~~.~~0~~.6~~ at the surface where significant changes of SOA occur in winter and summer, respectively. By using the modeled ALW_{org}, correlations between κ_{org} and O:C ~~were~~are examined in 9 representative cities, showing significant spatial and seasonal variations. The increases in SOA lead to ~~12% elevation of AOD and 15%~~ an enhancement in the averaged AOD and the cooling effects of ~~ARF~~aerosols, by up to 10% and 15% respectively in summer. The model predicted SOA is sensitive to temperature and QV in both seasons, with higher sensitivity to QV during winter and temperature during summer. Estimation of SVP also affects modeled SOA, especially in a cold environment. The effects of water partitioning into OPM and non-ideality of the organic-water mixture on SOA ~~were also examined~~

~~separately~~are the opposite. Since the activity coefficients of SVOCs are mostly greater than 1.0 during the simulated episode, SOA ~~concentrations decrease~~ concentration decreases when the non-ideality effect is considered. ~~Daily~~Averaged SOA concentration decreases by up to ~~~10~~20% in winter and ~~~30~~50% in summer~~ in the high regions.~~. Water partitioning alone increases SOA by ~~~~10-20~~-60~~% in winter and ~~~20-100~~30-80% in summer. It should be noticed that the results shown in this study are the lower limit as the current model tends to underestimate SOA. It is crucial to consider both effects in simulating SOA formation under hot and humid conditions in CTMs.

Data availability

Data used in this manuscript can be provided upon request by e-mail to the corresponding authors, Qi Ying (qying@civil.tamu.edu~~),~~) and Jianlin Hu (jianlinhu@nuist.edu.cn).

Competing interests

The authors declare that they have no conflict of interest.

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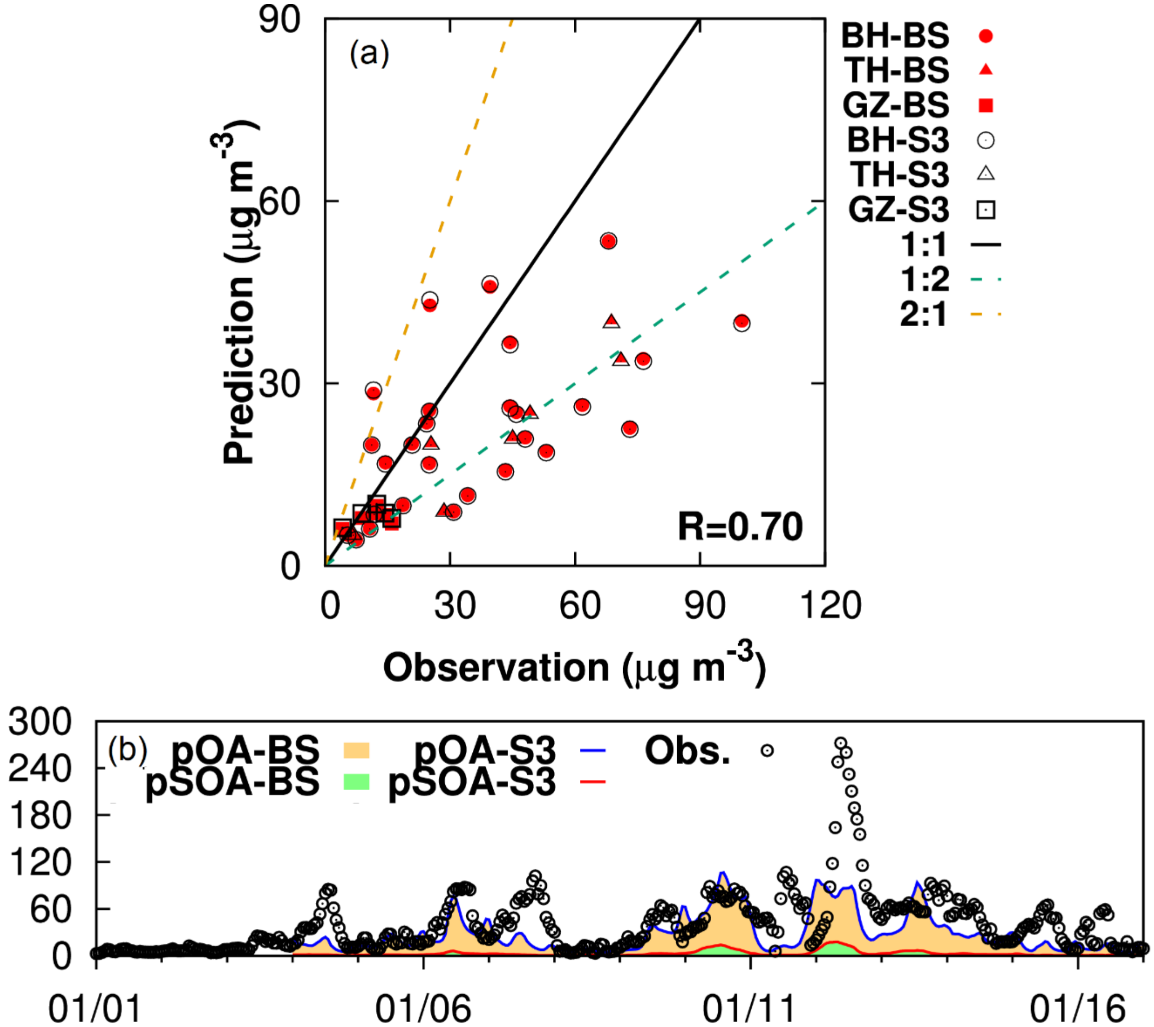
Table 1. Statistical analysis of modeled temperature (K) and relative humidity (%) of January and July of 2013 at the monitoring sites in different geographical areas as shown in Figure S1. and listed cities in Table S5.

		Northeast		NCP		Northwest		YRD		Central		Sichuan BasinSCB		PRD		Southwest		Benchmark
		Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	
Temperature (K)	ObsOBS	256.72	296.42	264.4263.9	297.94	268.1266.9	294293.0	277.87	303.62	276.1275.7	301.81	276.91	296.6295.7	289.4290.3	301.1	282.12	294.6295.1	
	PredPRE	251.6	296.53	261.32	298.98 268.5	295267.0	293.5	278.54	302.80	276.32	301.50	274273.5	294.0293.1	288.9289.4	300.20	278.48	291.18	
	MB	-5.14.6	-0.1	-3.12.7	1.34	0.481	1.0.5	0.7	-0.81.2	0.25	-0.31	-2.56	-2.6	-0.85	-1.0	-3.74	-3.54	≤±0.5
	GE 6.3 4.4	5.15	5.1.8 6.1	5.4.0	2.3	3.6	43.2	4.52.0	4.42.3	2.6.7	52.4	3.5.1	4.2.7	6.1.9	1.8	4.41	3.5	≤2.0
	NumR	101010.9	112980.9	150720.9	168200.8	114750.8	128300.8	68350.9	76200.8	212100.9	238090.8	135730.8	151920.9	70170.9	77150.6	120880.9	135900.8	
Relative Humidity (%)	ObsOBS	77.95	80.43	74.95	72.41	61.060.2	70.869.1	79.3	69.571.3	76.93	71.28	6968.5	74.24	7677.0	80.86	69.670.3	78.777.9	
	PredPRE	85.30	73.42	78.46	58.157.5	48.749.2	54.2	5675.1	73.676.9 73.0	64.5	70.30	57.87	78.4	75.23	84.58	78.075.8	87.085.1	
	MB	7.45	-7.01	3.54.1	-14.36	-12.311.0	- 15.014.6	-4.2	- 5.6 3.4 - 12.4 - 0.9	-11.68	4.2-1.8	-010.8	4.0	3-1.7	8.4.2	8.25.5	7.2	
	GE	12.411.3	11.3	14.6	16.6	19.28	19.1	11.0	11.6	16.28	2211.7 21.3	22.68	1711.6	17.89.5 20.8	17.9.2 25.5 17.2	15.04	13.810.4	20.6 16.2
	NumR	101010.5	112980.8	150720.4	168200.8	114750.3	128300.7	68350.6	76200.6	212100.6	238090.6	135730.2	151920.6	70170.6	77150.5	120880.5	135900.7	

*OBS is mean observation, PRE is mean prediction, MB is mean bias, GE is gross error, R is the correlation coefficient of predictions vs. observations. MB=$\sum_{i=1}^{N}(C_p - C_o)/N$; GE=$\sum_{i=1}^{N}\left|C_p - C_o\right|/N$, where C_p and C_o are the prediction and observation data, N is the total number of data.

**Northeast is Northeast China, NCP is North China Plain, Northwest is Northwest China, YRD is Yangtze River Delta, Central is Central China, SCB is Sichuan Basin, PRD is Pearl River Delta, Southwest is Southwest China.

***Benchmarks of MB and GE are for the MM5 model of 4-12 km horizontal resolution by Emery et al. (2001)



(a)
Prediction (μg m-3)
Observation (μg m-3)
0
30
60
90
120
BH-BS
TH-BS
GZ-BS
BH-S3
TH-S3
GZ-S3
1:1
1:2
2:1
R=0.70
(b)
pOA-BS
pSOA-BS
pOA-S3
pSOA-S3
Obs.
300
240
180
120
60
0
01/01
01/06
01/11
01/16

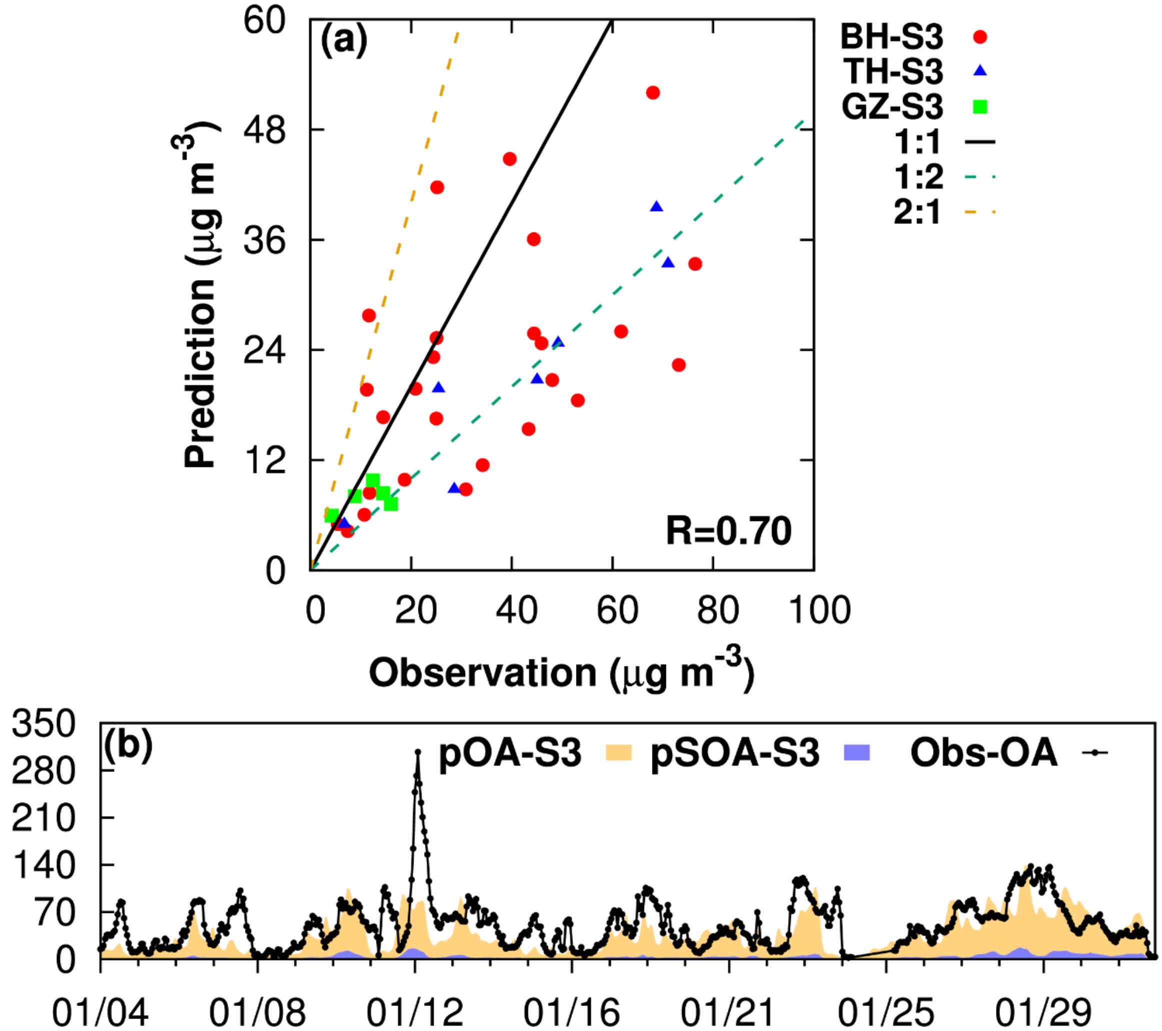
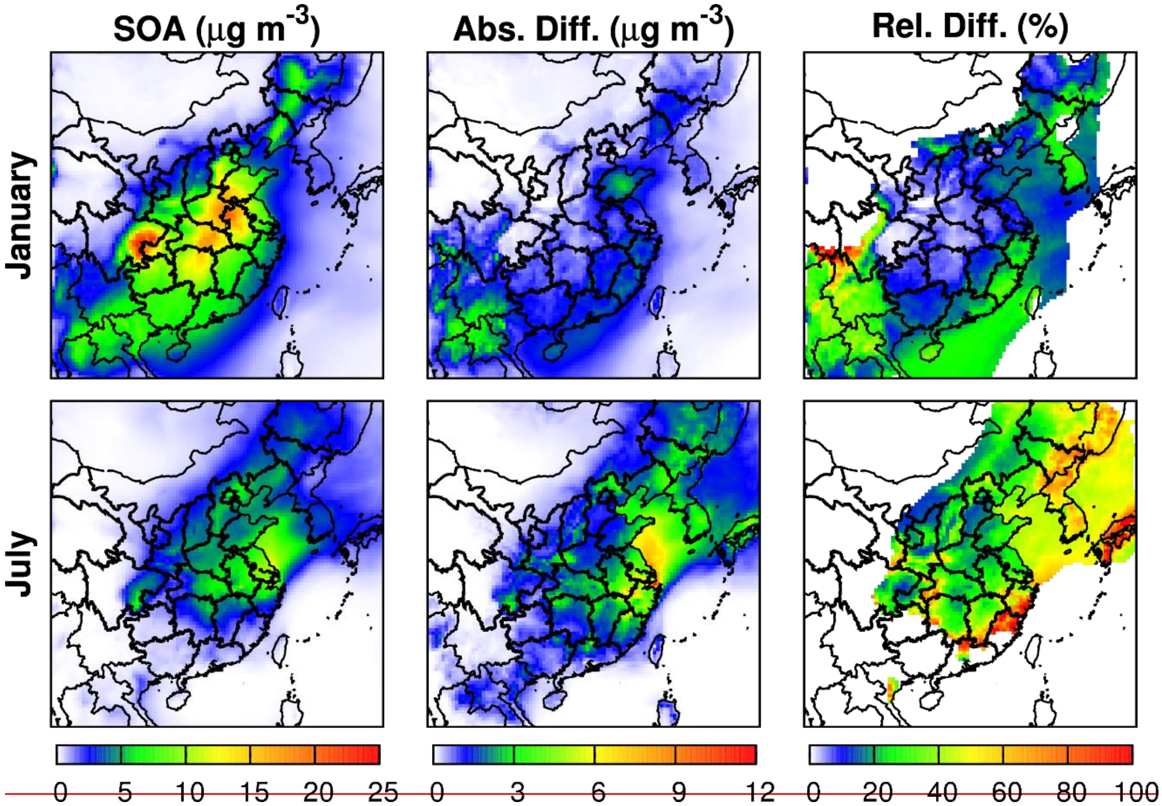


Figure 1. Comparison of (a) observed and modeled organic carbon concentration at University of Beihang (BH), Tsinghua University (TH) and Guangzhou (GZ); (b) observed organic aerosol (Obs.) -OA) at Beijing and predictions of total OA (pOA) and SOA (pSOA), unit is μg m^{-3}. Locations of monitoring sites are shown in Figure S1.



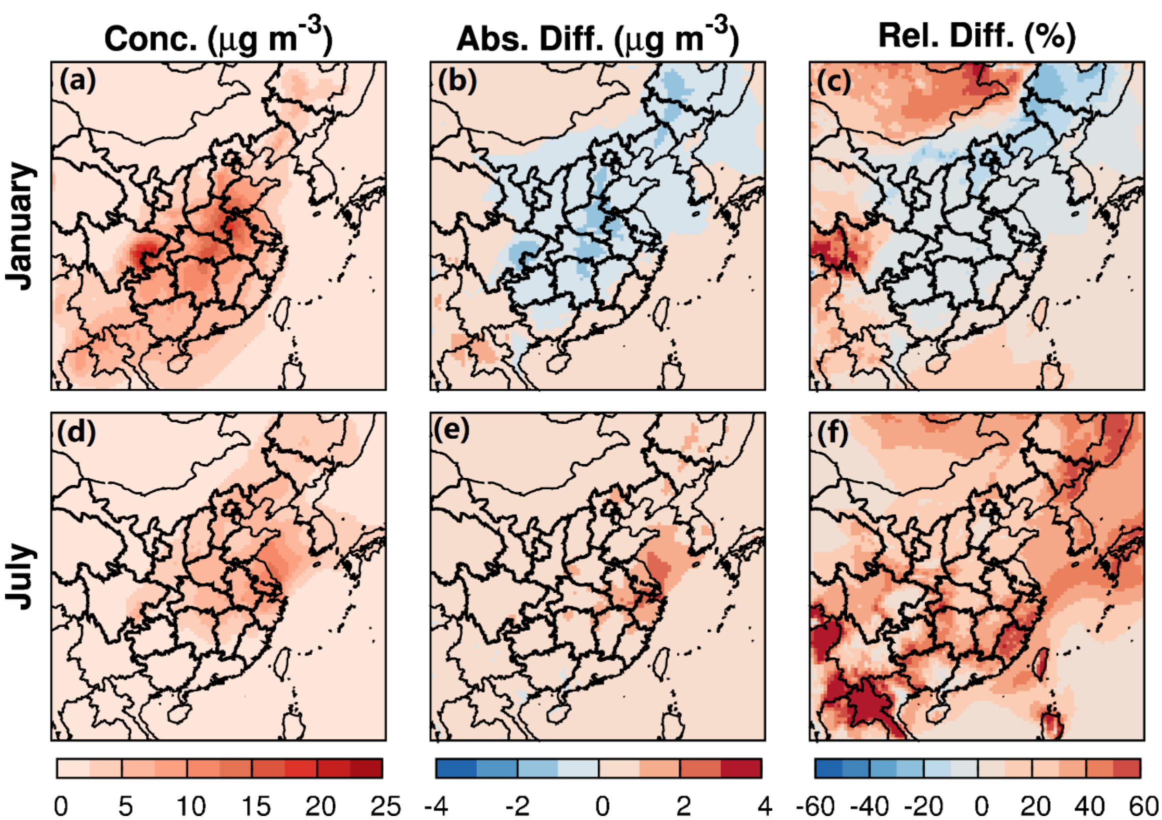
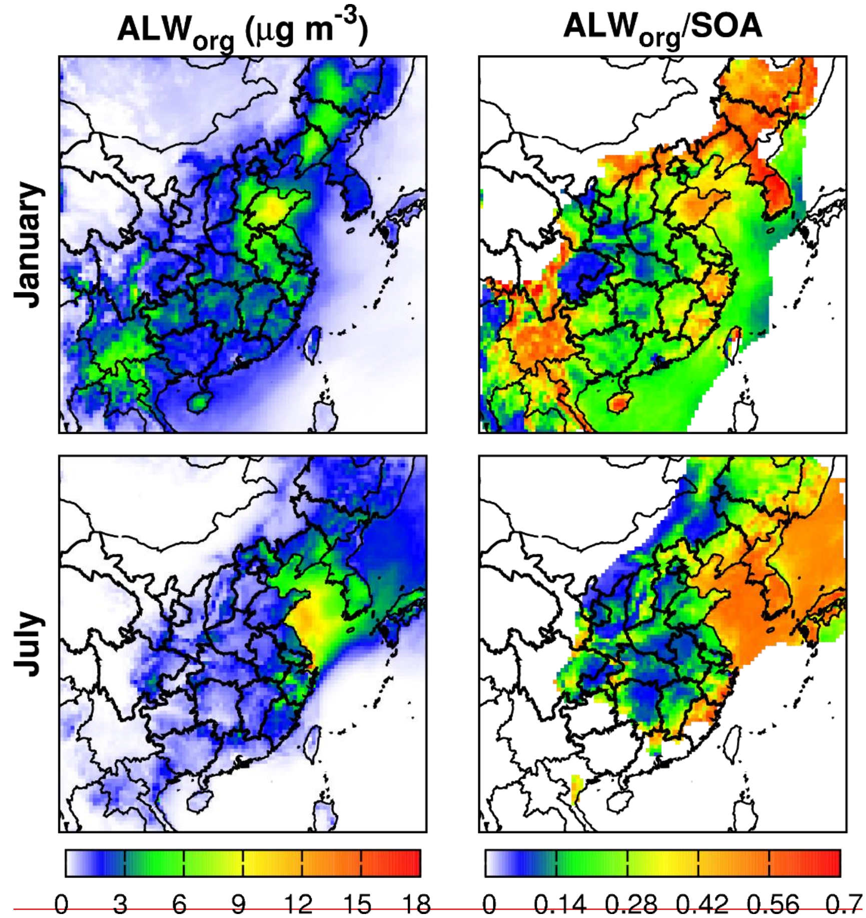


Figure 2. Monthly-averaged ~~total~~ SOA ~~in BS~~from S3 and ~~monthly averaged daily~~

maximum changes of SOA due to water partitioning into OPM and non-ideality of the organic-water mixture. "Abs. Diff." represents absolute differences (S3-BS); "Rel. Diff." represents relative differences ((S3-BS)/BS, %). Relative differences are shown in areas with monthly-averaged SOA concentration greater than 1 μg m^{-3}.


ALW$_{org}$ (μg m^{-3})
ALW$_{org}$/SOA
January
July
0 3 6 9 12 15 18
0 0.14 0.28 0.42 0.56 0.7

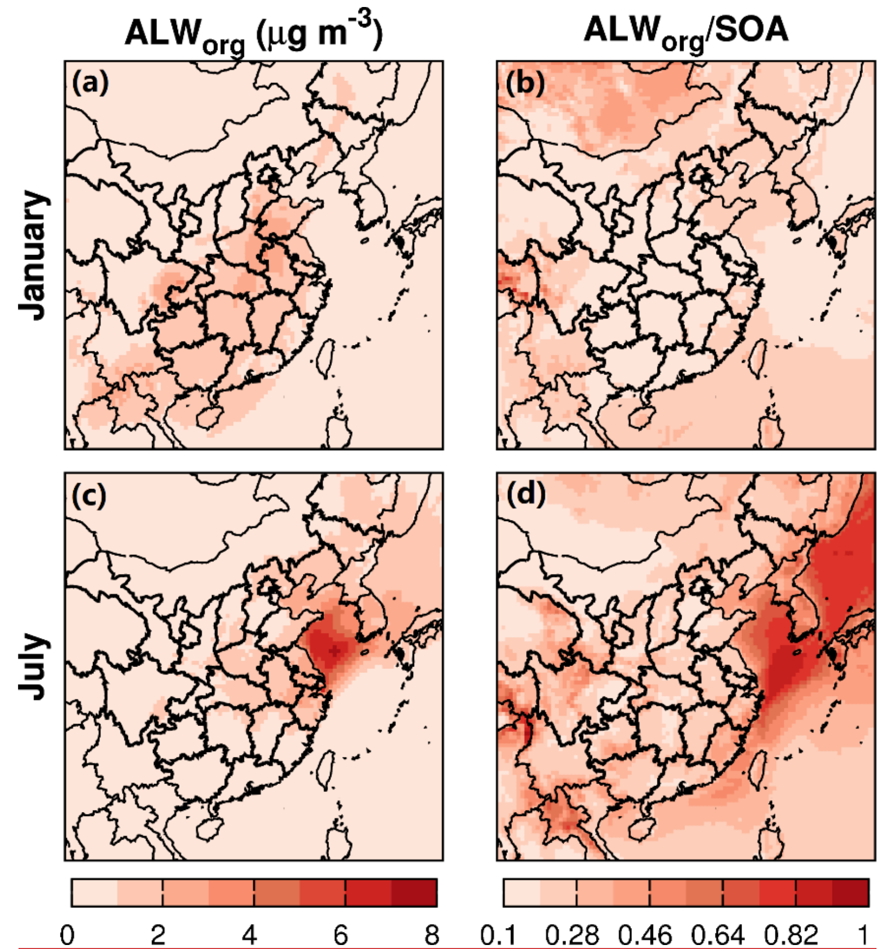
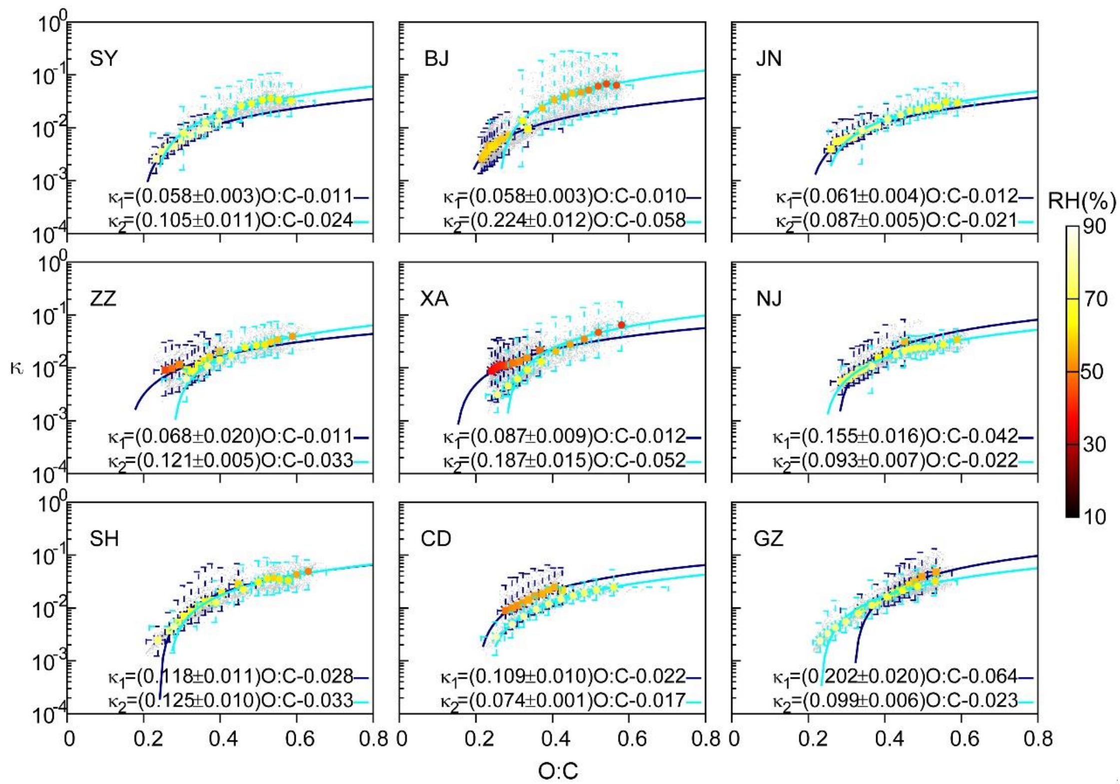


Figure 3. Monthly-averaged daily maximum water partitioning into the organic-phase (ALW_{org}, μg m^{-3}) and the ratio to SOA (ALW_{org}/SOA) during January and July of 2013. AWL_{org}/SOA is shown in areas with monthly-averaged SOA concentration greater than 1 μg m^{-3}.



SY
κ1=(0.058±0.003)O:C-0.011
κ2=(0.105±0.011)O:C-0.024
BJ
κ1=(0.058±0.003)O:C-0.010
κ2=(0.224±0.012)O:C-0.058
JN
κ1=(0.061±0.004)O:C-0.012
κ2=(0.087±0.005)O:C-0.021
ZZ
κ1=(0.068±0.020)O:C-0.011
κ2=(0.121±0.005)O:C-0.033
XA
κ1=(0.087±0.009)O:C-0.012
κ2=(0.187±0.015)O:C-0.052
NJ
κ1=(0.155±0.016)O:C-0.042
κ2=(0.093±0.007)O:C-0.022
SH
κ1=(0.118±0.011)O:C-0.028
κ2=(0.125±0.010)O:C-0.033
CD
κ1=(0.109±0.010)O:C-0.022
κ2=(0.074±0.001)O:C-0.017
GZ
κ1=(0.202±0.020)O:C-0.064
κ2=(0.099±0.006)O:C-0.023
RH(%)
κ
O:C

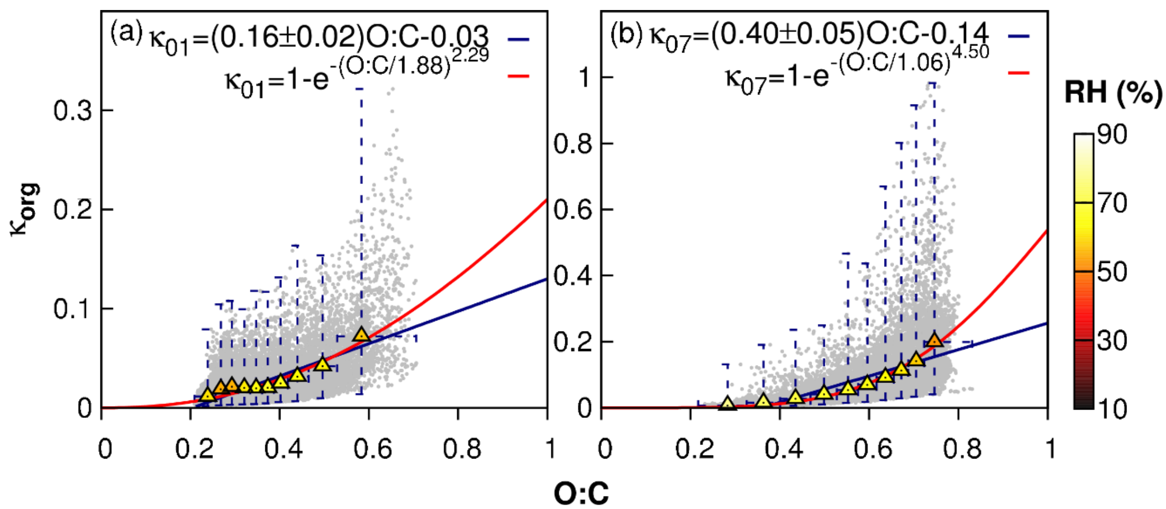
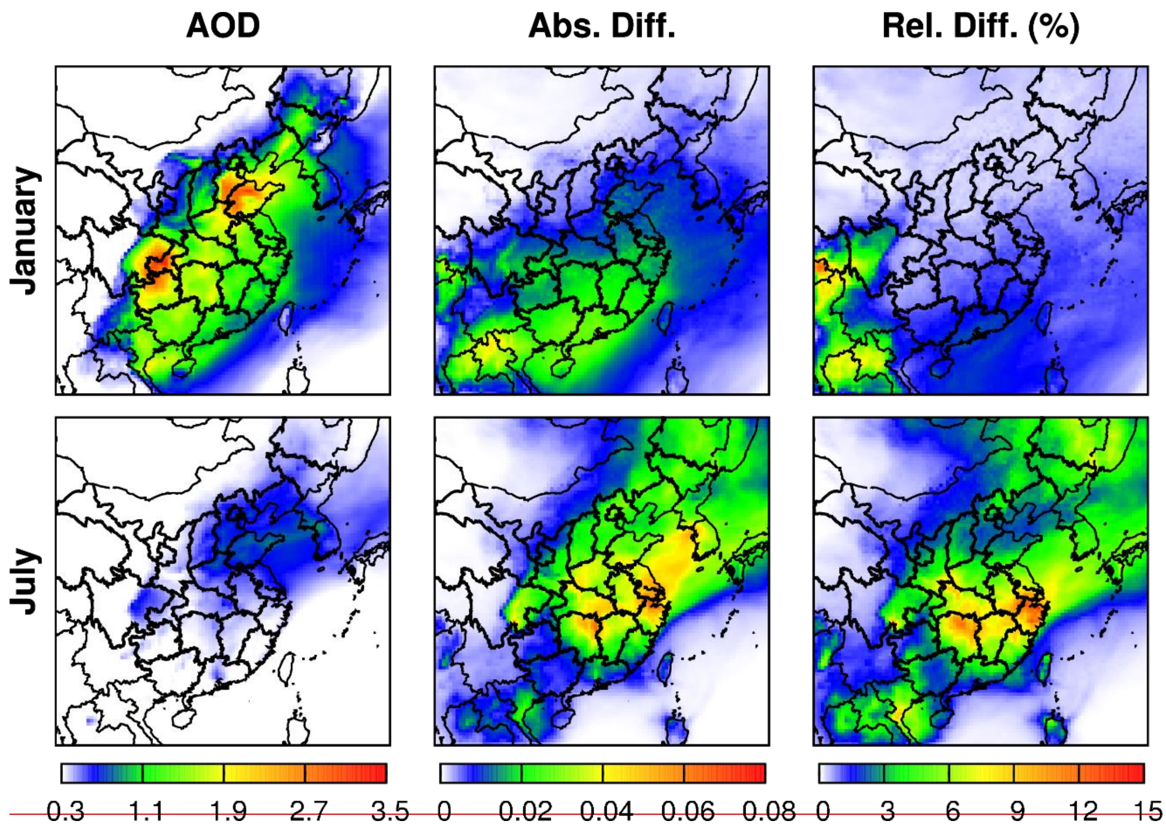


Figure 4~~.~~. The correlation of hygroscopicity of organic aerosol (~~κ)~~κ_{org}) and O:C ratio ~~at~~in 9 representative cities including Shenyang (SS), Beijing (BJ), Jinan (JN), Zhengzhou (ZZ), Xi'an (XA), Nanjing (NJ), Shanghai (SH), Chengdu (CD), and Guangzhou (GZ~~). Gray dots on the background represent all the data~~) in January (a) and July~~, which~~ (b) of 2013. O:C ratios are categorized into ~~several O:C~~10 bins. In each bin, the ranges of ~~κ~~O:C and ~~O:C ratio~~κ_{org} are represented by ~~dashed~~ bars~~ colored for January (navy) and July (light blue), with the mean value~~. The mean values of O:C and κ_{org} are represented by triangles colored by the averaged RH of each bin. ~~The mean κ~~The relationship between κ_{org} and O:C ~~ratio are~~is fitted by a linear function with reduced major axis regression~~ –~~ (blue lines) and an exponential function (red lines), respectively. κ_{01} and κ_{07} represent the fitted correlation for January and July, respectively.



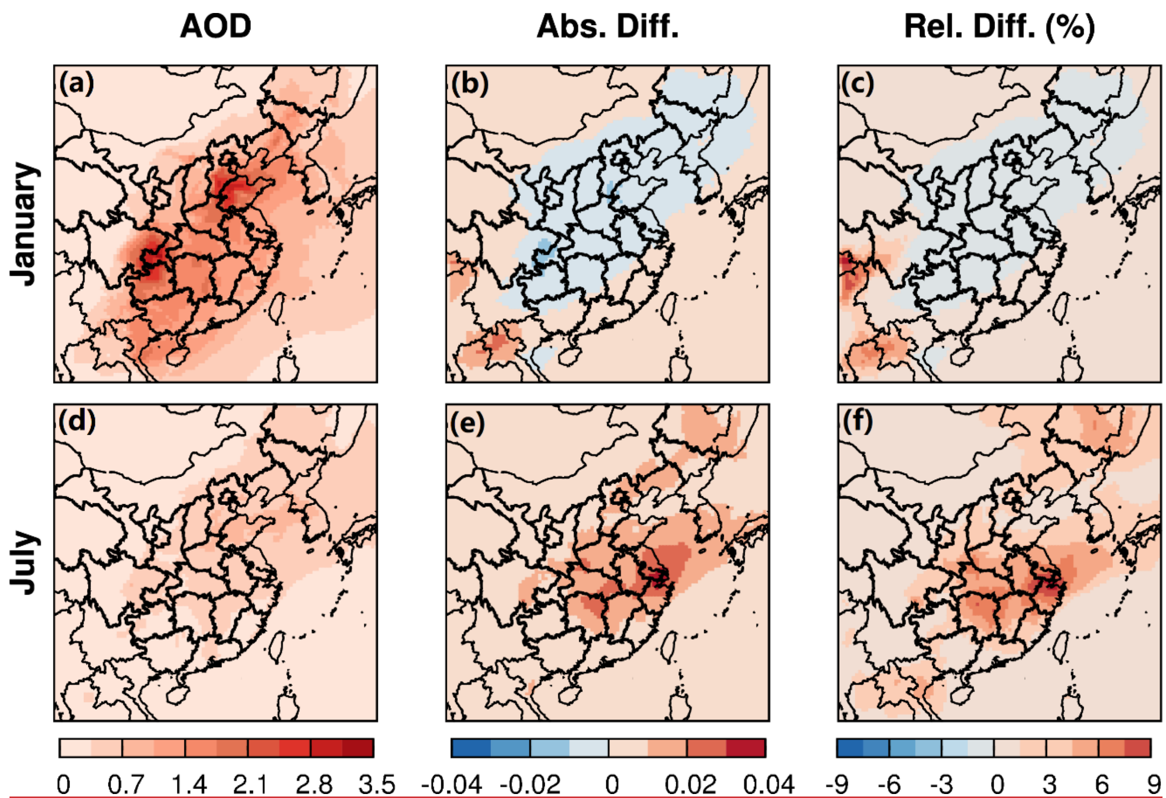
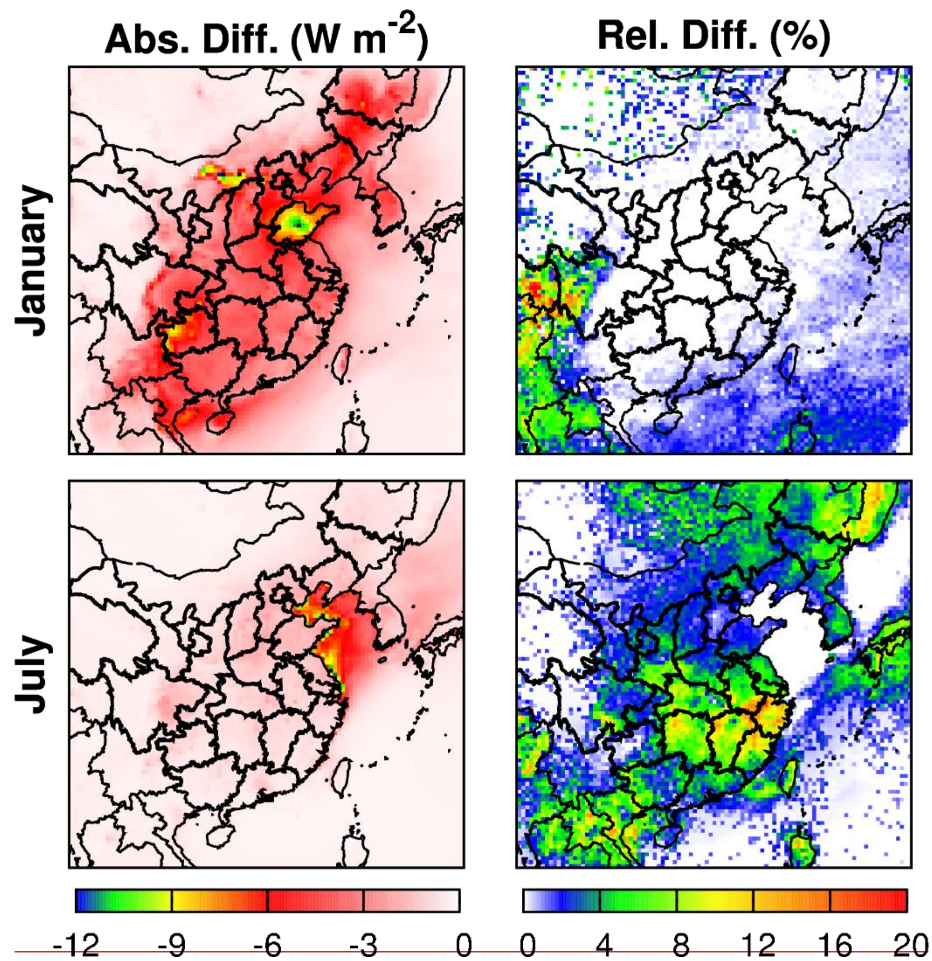


Figure 5. Monthly-averaged AOD_r at 550 nm and ~~the monthly-averaged daily maximum~~ changes of AOD_r due to water partitioning into OPM and ~~the~~ non-ideality of the organic-

water mixture. “Abs. Diff.” represents absolute differences (S3-BS); “Rel. Diff.” represents relative differences ((S3-BS)/BS, %).



Abs. Diff. (W m^{-2})
Rel. Diff. (%)
January
July
-12 -9 -6 -3 0
0 4 8 12 16 20

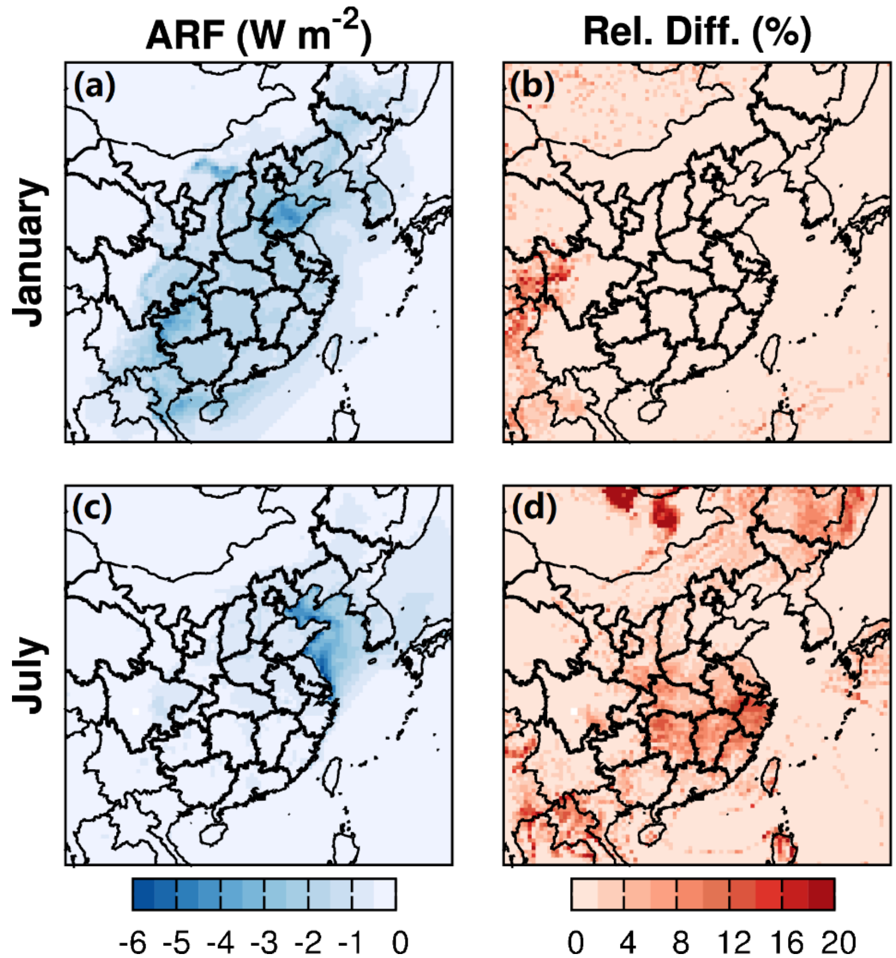
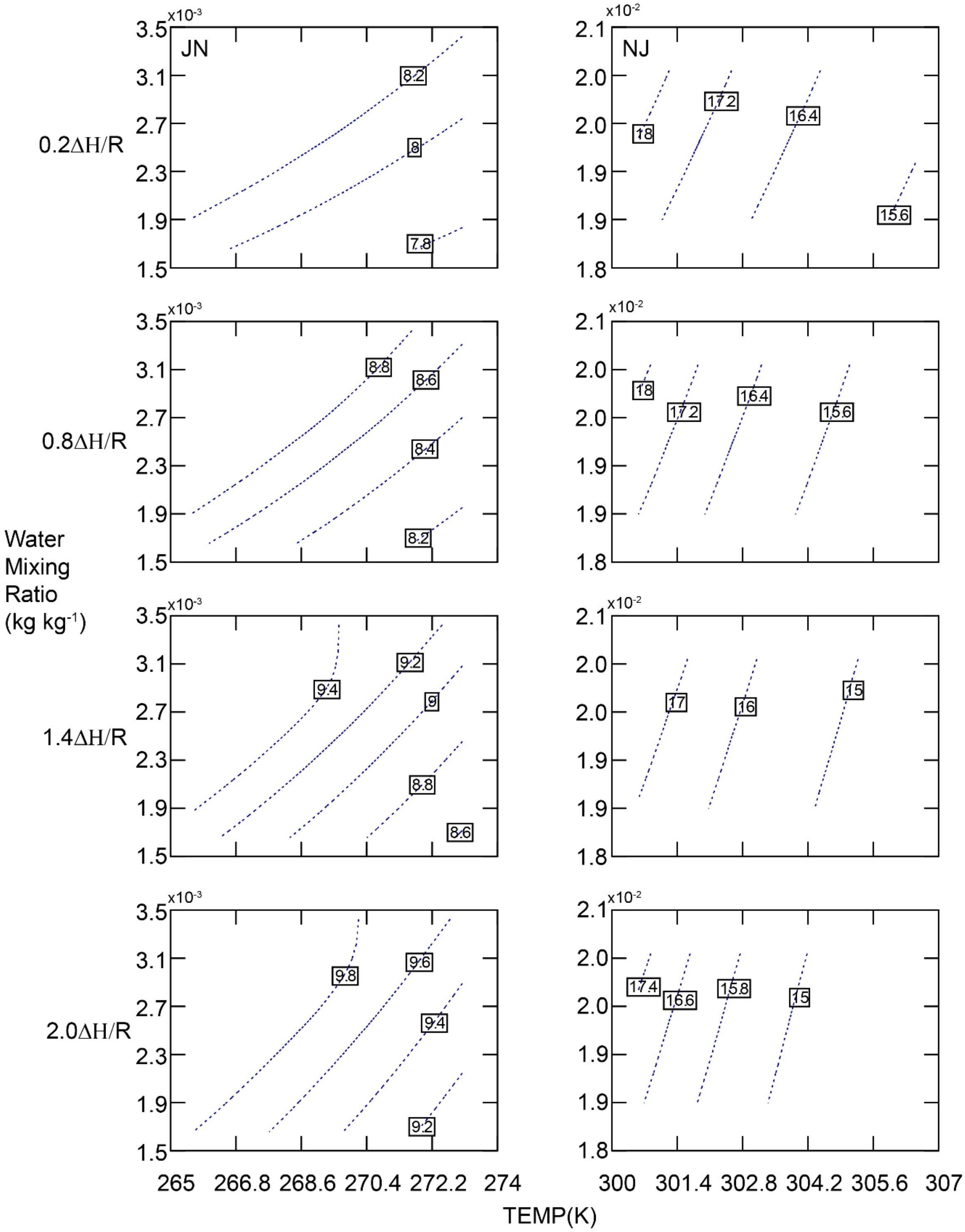


Figure 6. Monthly-averaged daily maximum variation of shortwave direct aerosol radiative forcing at the top of the atmosphere from S3 and the relative changes due to water partitioning into OPM and non-ideality of the organic-water mixture during January and July of 2013. "Rel.Abs. Diff." represents absolute differences (S3-BS); "Rel. Diff." represents relative differences ((S3-BS)/BS, %).



JN
NJ
0.2ΔH/R
0.8ΔH/R
1.4ΔH/R
2.0ΔH/R
Water Mixing Ratio (kg kg-1)
x10-3
x10-2
3.5
3.1
2.7
2.3
1.9
1.5
2.1
2.0
1.9
1.8
8.2
8
7.8
18
17.2
16.4
15.6
8.8
8.6
8.4
9.4
9.2
9
17
16
15
9.8
9.6
17.4
16.6
15.8
265
266.8
268.6
270.4
272.2
274
300
301.4
302.8
304.2
305.6
307
TEMP(K)

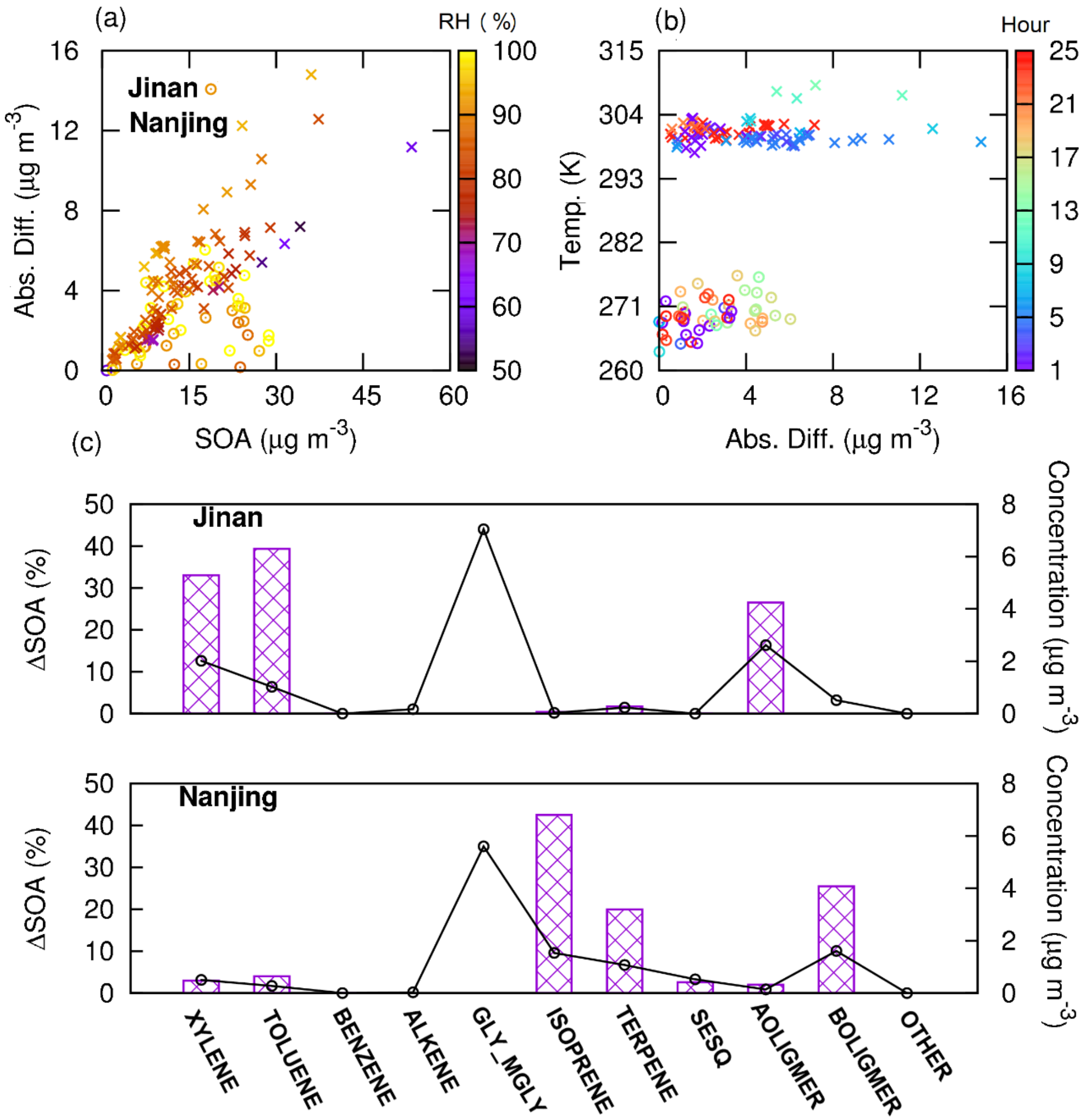
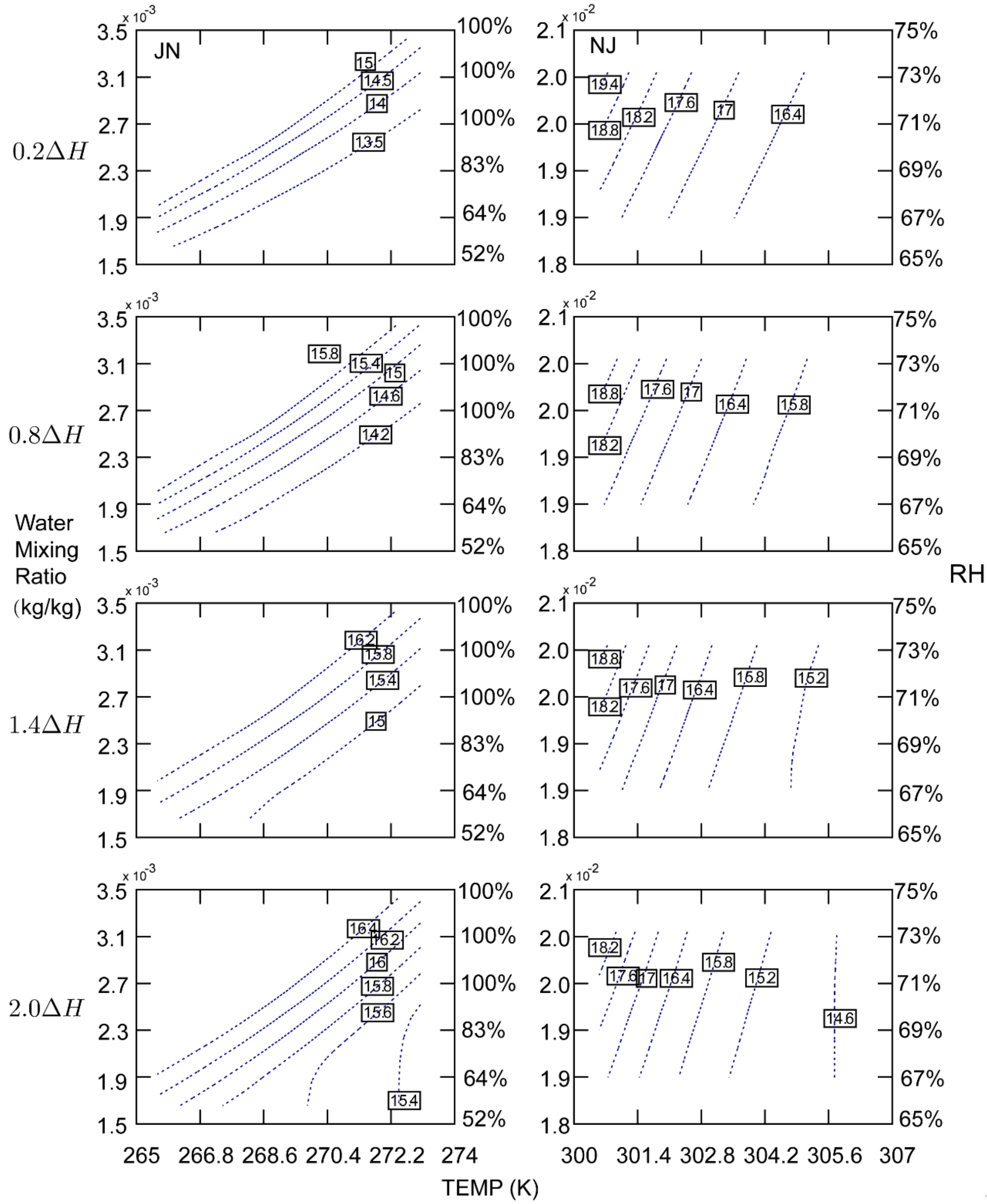


Figure 7. ~~Correlation of water partitioning on SOA with (a) RH (b) temperature at Jinan in winter and Nanjing in summer, and the contribution from each SOA component to the total SOA increase. In plot (a) and (b),~~The sensitivity~~"Abs. Diff."~~ ~~represents the daily maximum change of SOA that is calculated as S3-BS. Color box represents RH in (a) and the hour in the day in (b) when daily maximum change of SOA occurred. In (c), the left axis represents contribution of each SOA component to the daily maximum SOA change due to water partitioning, and the right axis represents the concentration of each SOA component.~~



~~Figure 8. Sensitivity~~ of SOA formation to temperature (TEMP), ~~relative humidity (RH)~~water mixing ratio, and the temperature dependence parameter of SVP ($\Delta H/R$) at Jinan (JN, first column) and Nanjing (NJ, second column).

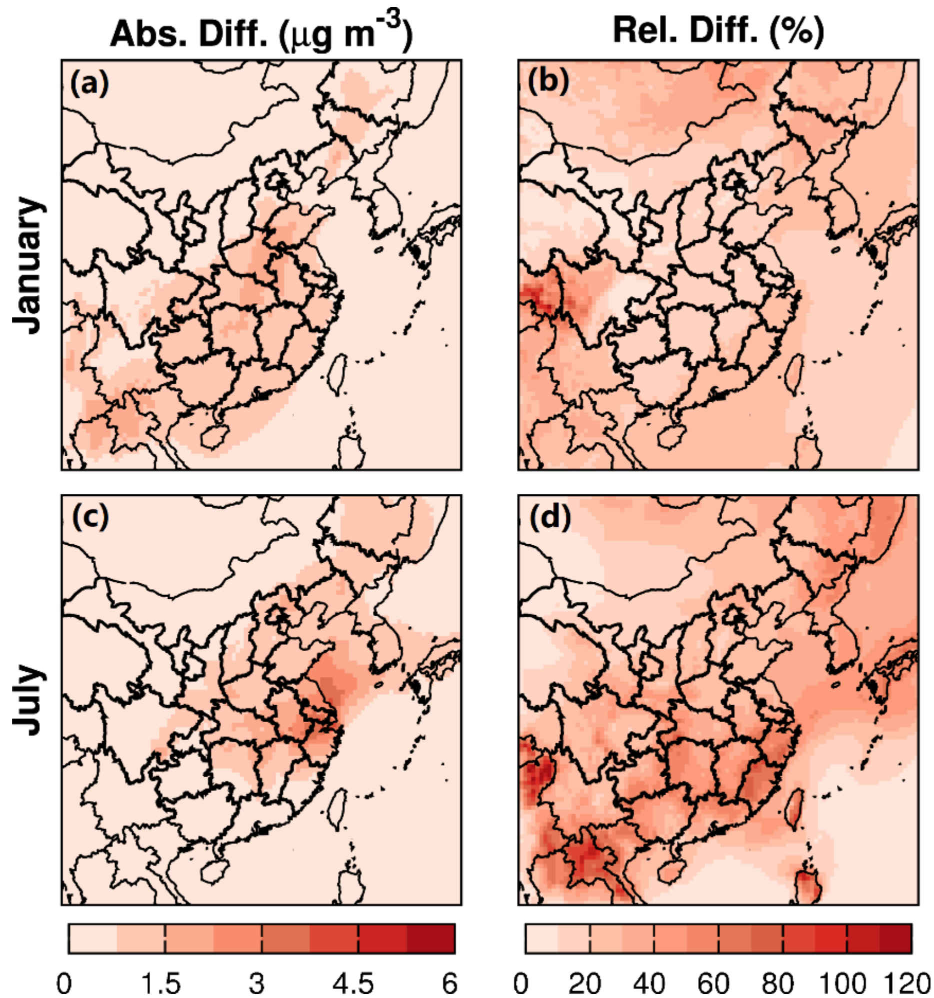


Figure 8. Monthly-averaged impacts of water partitioning into OPM on SOA. "Abs. Diff." represents absolute differences (S3-S2); "Rel. Diff." represents relative differences ((S3-S2)/S2, %).

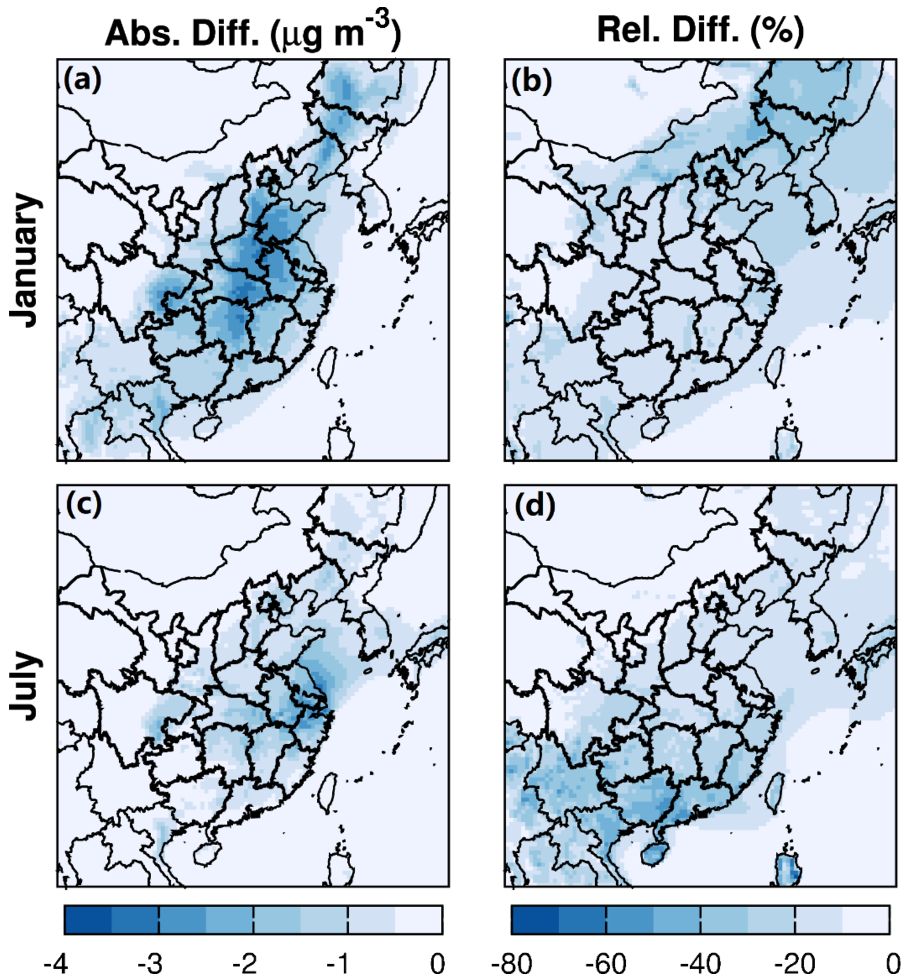


Figure 9. Monthly-averaged impacts of non-ideality of the organics-water mixture on SOA. "Abs. ~~The relative humidity is showing on the right side of y-axis.~~
Diff." represents absolute differences (S3-S1); "Rel. Diff." represents relative differences ((S3-S1)/S1, %).