Manuscript #: acp-2021-1002

Response to Anonymous Referee #RC1:

In this manuscript, the authors incorporated their sophisticated SOA model (UNIPAR) with an air quality model (CAMx) and simulated SOA concentrations from different formation pathways and different precursors. Observed concentration of organic matter (OM) is better reproduced by the UNIPAR mode than by a conventional two product model (SOAP). By applying the UNIPAR model, the SOA formation from gas-particle partitioning, in-particle oligomerization, and aqueous-phase reactions are separately calculated, and their contributions have been quantified.

This manuscript is well written and includes useful information about the numerical modeling of SOA formation processes in the ambient air. However, I have several concerns as below. I recommend this manuscript for publication after the following concerns are adequately addressed.

Response: We thank the reviewer for the valuable comments on this manuscript. To response to the comments from the reviewer, the explanation and discussions are added in the revised manuscript. A line-by-line response for each comment are listed below.

1 Comments on Methodology

I am afraid that methodology (model and emissions) is not comprehensively described, or adequate references are cited.

- You wrote in L109 that "The mathematical equations used to construct the stoichiometric coefficient array are reported in Section S1" and four parameters (A, B, C, and D) for different precursors and conditions (NO_x level and aging status) are given in Table 3. However, I could not find the information how did you consider dependence on NO_x (high/low) and aging degree (fresh/aged) for the calculation of stoichiometric coefficients in the ambient conditions.

Response: The information for the impact of the NO_x level and aerosol aging has been added to the section S1 in the supporting information and reads now.

"The mass-based stoichiometric coefficient (αi) of each lumping species i can be calculated based the variables listed in Table S3. Both the stoichiometric coefficient array derived from the fresh compositions and that from the aged compositions are determined as a function of NO_x levels using the mathematical equations. To simulate age-dependent SOA formation, the stoichiometric coefficient array is reconstructed over time by a weighted average of fresh and aged stoichiometric coefficients based on the normalized concentration of oxidized organic radicals and HO_2 with a hydrocarbon concentration. The detailed information of the calculation of age-dependent stoichiometric coefficient of lumping species was discussed in the previous study (Zhou et al., 2019)."

- You set six categories for oxidation products: non-reactive (P), slow (S), medium (M), fast (F), very fast (VF), and multifunctional alcohols (MA). Products with these categories are always produced or did you consider any condition dependence?

Response: The value of the stoichiometric coefficient associated with volatility and reactivity in aerosol phase depends on the precursors, the oxidation status (aging), and the NO_x levels. It is not necessary that all stoichiometric coefficient arrays are filled. For example, the quantity of MA is high in isoprene products but very little or none in the products from other precursors.

- Thermodynamic parameters of oxidation products (vapor pressure and vaporization enthalpy) are not explicitly shown.

Response: Prior to the establishment of the physicochemical parameters (vapor pressure, enthalpy of vaporization) of the UNIPAR lumping species, the physicochemical parameters of all explicit products were individually calculated and classified into vapor pressure groups which is paired with enthalpy of the vaporization. In UNIPAR, the stoichiometric coefficients associated with volatility was not semi-empirically determined but determined by considering the properties of explicit products.

- Information of emission amounts is not shown. As you estimated the contributions of SOA precursors, total emissions or their distributions are important information. I have two more concerns about emissions:

Response: As seen in Section 2.3.2, the emission of air pollutants was prepared by using SMOKE from emission inventories originating from various sources (i.e., point sources, area sources, biogenic sources (MEGAN), automobiles non-mobile sources, etc.).

- You wrote in L255 that "During the wet period, HC emissions increased". It appears from Figures S5 and S6 that daytime temperature is higher during the dry periods than wet, and thus, I speculate that BVOC emissions are higher during the dry period. Quantitative information and reasons for the increase of HC emissions should be given.

Response: Thank the reviewer for this question. The sentence has been revised as follows.

"During the wet period, the concentration of anthropogenic HC increased".

As the reviewer mentioned, the variation of the flux of the biogenic hydrocarbons depends on the geological factors and the metrological conditions mainly influenced by temperature and sunlight intensity. The SOA model simulation result shows the gradual increase of biogenic SOA mass from early May to early June due to seasonal change under the metrological conditions. There are some variations in the biogenic SOA production due to the daily change in the biogenic hydrocarbon emission associated with differences in temperature and sunlight intensity. However, the variation of the biogenic SOA mass is relatively small compared to that of anthropogenic origin SOA during the simulated period as seen in Figure 7.

- L308: "isoprene SOA is negligible at all sites due to low isoprene emissions". Information of isoprene emissions (preferably with terpene and aromatics) is required.

Response: In South Korea, biogenic hydrocarbons mainly originate from coniferous tree, which is dominated by the pine trees. Therefore, the monoterpene flux in South Korea are relatively significant (Lee et al., 2017). An estimation of isoprene and monoterpenes emissions in the global scale based on 30-years Megan simulation showed that the relative significance of isoprene contribution to the biogenic hydrocarbons is little. In order to response to the reviewer, we added sentence to the revised manuscript (last paragraph in Section 3.3).

"An estimation of biogenic hydrocarbon emissions in the global scale, simulated by Sindelarova et al. (2014) by using Megan for 30 years, showed that the relative significance of isoprene emission is little in South Korea."

2 Comments on precursors' contributions:

You wrote in L308 as "Isoprene SOA is negligible at all sites", and concentrations of isoprene SOA was small over the domain as shown in Figure 8 (g) and (h). However, previous observational and simulation studies have indicated that isoprene SOA has important contributions in East Asia in May-June (e.g., Hu et al., Zhu et al., and Ding et al.). I recommend the authors to discuss the differences of your estimate with previous studies.

Hu et al. (2017) doi:10.5194/acp-17-77-2017

Zhu et al. (2018) doi: 10.1016/j.apr.2017.09.001

Ding et al. (2016) doi: 10.1038/srep20411

Response: The emission of isoprene is spatial sensitive. Due to the difference in tree species and climates (temperature, humidity, sunlight, and precipitation), the isoprene emission in Northeast Asia is much lower than that in Southeast Asia. The modeling results in the previous studies also reported (Ding et al., 2016; Hu et al., 2017; Zhu et al., 2018) that the isoprene emission was significantly lower in Northeast China than that in South China. South Korea has a clear four seasons with cold winter and hot summer. In addition, the emission of isoprene in May during the KORUS-AQ campaign is relatively lower than that in the summer seasons (July-September).

3. Comments on OMH and OMP

You wrote in L324-326 that "Under the dry period (Fig. 3), the predicted SOA mass by the UNIPAR model is dominated by gas-particle partitioning onto organic phase and oligomerization in organic aerosol. During the wet period, SOA production forms mainly through gas-aqueous partitioning and aqueous reactions."

I could not get how did you separate contributions of oligomer SOA and SOA from aqueous-phase reactions (I guess both are categorized OMH). Quantitative information of the contributions of the three pathways is helpful to readers.

Response: In the UNIPAR model, the SOA formation is processed by the two mechanisms: OMP from multiphase partitioning and OMH from oligomerization in both organic and salted aqueous solution. To clarify the SOA formation in the UNIPAR model, the following sentence is added to the revised manuscript and reads now (item 4 and 7 in section 2.2).

"The SOA mass formed from the partitioning process (OMP) is attributed to C_{or} and C_{in}."

"The SOA mass in the UNIPAR model is attributed to OMP and OMH."

4 Comment on OM and OC

It is not clear whether you showed organic mass (OM) or organic carbon (OC) in Figures 3-5. I guess OM concentration is calculated by your simulation model, whereas OC concentration is measured by carbon analyzers. Conversion factor from OC to OM (or vice versa) should be explicitly noted.

Response: We thank the reviewer for this comment. Both observation data and simulation results are organic matter (OM). The figure captions in Figures 3 and 4 are corrected in the revised manuscript and reads now.

"Figure 3: Time profiles of OM concentration ($\mu g/m^3$) averaged over eight hours for the observation data and the CAMx simulation results at the (a) Bulkwang, (b) Olympic Park, (c) Daejeon, and (d) Gwangju supersites."

"Figure 4: Time profiles of hourly averaged OM concentrations ($\mu g/m^3$) for the observation data and the CAMx simulation results at the (a) Bulkwang, (b) Olympic Park, (c) Daejeon, and (d) Gwangju supersites."

Specific comments:

L51: References for the following sentence is necessary: "In particular, the current model applied to regional scales suffers from a substantial negative bias under high humidity conditions."

Response: Reference has been added to the second paragraph in introduction of the revised manuscript and reads now.

"In particular, the current model applied to regional scales suffers from a substantial negative bias under high humidity conditions (Heald et al., 2011; Pye et al., 2017; Li et al., 2020)."

-L104: eight aromatics?

Response: Total 10 aromatics were included in the UNIPAR simulation: benzene, toluene, ethylbenzene, propylbenzene, o-xylene, m-xylene, p-xylene, 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, and 1,3,5-trimethylbenzene. The description in the item 2 of the Section 2.2 has been updated and reads now.

"The UNIPAR model of this study includes 151 lumping species, of which 50 originate from ten aromatics (benzene, toluene, ethylbenzene, propylbenzene, o-xylene, m-xylene, p-xylene, 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, and 1,3,5-trimethylbenzene);"

L214: VCPs sourced from "residential, commercial, and industrial sectors"?

Response: The sentence has been revised in the second paragraph of Section 3.1 and reads now.

"The SOA simulation needs to be updated to include sesquiterpenes, intermediate VOCs, and volatile chemical species sourced from residential, commercial, and industrial sectors (Mcdonald et al., 2018)."

L300: "OMH attributes to 50% of aromatic SOA": it appears OMH contribution is smaller than 50% in Fig. 7 (during the wet period).

Response: The sentence has been updated in the third paragraph of the Section 3.3 and reads now.

"In Fig. 7a-7d (UNIPAR), OMH attributes to 22% to 48% of aromatic SOA, showing the importance of heterogeneous reactions of aromatic products to form SOA during the KORUS-AQ campaign."

L342: 53% of total anthropogenic VOC emissions in LA?

Response: The sentence has been updated in the third paragraph in Section 4 and reads now.

"In addition, the recent study by Mcdonald et al. (2018) showed that volatile chemical products (>53% of total anthropogenic VOC emissions in Los Angeles, USA) originating from consumer and industrial products, which are currently unaccounted for in models, can significantly contribute to SOA burden in the urban atmosphere."

Response to Anonymous Referee #RC2:

"Yu et al., describe the impact of implementing a state-of-science module for the formation of secondary organic aerosol from traditional as well as "novel" pathways including multi-phase processes involving particles. They evaluate their model against ground observations taken during a recent field campaign over South Korea for the duration of 1 month.

The manuscript is well written and presents the main findings in a concise and understandable fashion. Conclusions are sound presented in a balanced manner, mostly considering the state of the science in the field at this time. My main points are (1) the need to also focus on the remainder of the lifecycle of organic matter in the atmosphere, (2) to make better use of the wealth of data generated during KORUS-AQ to evaluate the model, and (3) a broader evaluation of the model performance. I would recommend major revisions."

Response: We appreciate the reviewer for the time and effort on this study. Additional discussion about the aerosol lifecycle and the model evaluation using the field data are added in the revised manuscript. A line-by-line response to the reviewer's comment is listed as below.

Main points:

(1) Organic aerosol lifecycle

Concentrations of OA in the atmosphere are determined by its sources (emission, secondary production) as well as its sinks. The authors claim to do better firstly because their model represents more of the physics and chemistry that probably takes place in the atmosphere, and secondly because it evaluates better against observations. I concur with the former, but find the latter needs to be discussed (further) in the manuscript. A lot of work has shown that OA can photolysis, age, and deposit in ways most models do not consider, thereby changing its properties and lifetime. Why is being closer to observations now "better" with UNIPAR, maybe you are just compensating model deficiencies in other areas?

Response: We agree with the reviewer that there are uncertainties associated with aerosol aging in the model of this study. In the CAMx-UNIPAR simulation, the deposition of the OA originated SOA was treated based on the one size bin for the fine particulate matter. The UNIAPR model include the aging of gas products but needs an aerosol aging process due to OA aging in the future. The discussion about aerosol deposition can be found in the section 2.1. The discussion about OA aging and aerosol deposition has been added to the section 4 Atmospheric implications and uncertainties and reads now.

"In addition, the deposition of SOA was estimated with the one particle size bin. The different particle size can have different sink fluxes causing the uncertainty in the lifetime of OM. The UNIPAR model is capable of predicting aging of gas products but currently has no feature for OM aging."

(2) KORUS-AQ campaign data

KORUS-AQ was also a large aircraft campaign, a treasure trove of observations is readily available (including OA data!) from several aircraft platforms. It would be almost negligent to not use this data to evaluate a 3D m model simulation and instead focus only on three ground stations. There is so much more to learn about OA model performance when looking "up in the sky"!

Response: Thank the reviewer for the important comments. In the future, we will utilize the aircraft data. Prior to the compare the simulation with aircraft data, the evaluation of the aircraft data based on emission sources need to be performed.

(3) Model performance evaluation

The authors have provided quite some data to look at overall model performance, but I suggest to complete this in the following areas: how well is NOx represented, what is the performance for temperature and humidity, and how well does the model represent the main SOA precursor levels (aromatics, terpenes and isoprene)? Again, see point 2, there is a wealth of data available!

Response: The NO_x observations during the KOROS-AQ were not available in many sites. We can find NO_x observation data only at the Olympic Supersite of the selected four sites of this study (Figure S4). The sites chosen in this study are at best in the availability of various data and timeline.

The temperature and humidity were produced from the WRF model and they accorded well with the filed measurements (R^2 = 0.9999 for temperature and R^2=0.9688 for relative humidity). This information can be found in the figure caption in Figures S5 and S6 and reads now:

"The temperature inputs from the WRF simulation accords well with the filed measurements (R^2 = 0.9999)."

"The RH for the CAMx meteorological inputs from the WRF simulation accords well with the filed measurements (R^2 = 0.9688)."

Specific comments:

15 ff "explicit" gas-phase chemistry?

Response: Corrected and reads now.

"The UNIfied Partitioning-Aerosol phase Reaction (UNIPAR) model utilizes the explicit gas mechanism to better predict SOA formation from multiphase reactions of hydrocarbons."

37 why italic for "via"?

Response: Word "via" was not italicized now.

37 HC abbreviation, first mention, explain!

Response: This has been corrected in the 2nd paragraph in introduction.

48: The fact that SOA precursors can undergo multi-phase chemistry involving a liquid-phase implies they are hygroscopic, which leads to important questions regarding their fate in the atmosphere. E.g., is deposition accounted for correctly (see, e.g., Knote et al., 2015)? Also, given that at least during daytime, we are in a photochemically active environment, what about photolysis losses of oxidized volatile organic compounds (OVOCs) (e.g., Hodzic et al, 2015)?

Knote, C., Hodzic, A., and Jimenez, J. L.: The effect of dry and wet deposition of condensable vapors on secondary organic aerosols concentrations over the continental US, Atmos. Chem. Phys., 15, 1–18, https://doi.org/10.5194/acp-15-1-2015, 2015.

Hodzic, A., Kasibhatla, P. S., Jo, D. S., Cappa, C. D., Jimenez, J. L., Madronich, S., and Park, R. J.: Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime, Atmos. Chem. Phys., 16, 7917–7941, https://doi.org/10.5194/acp-16-7917-2016, 2016.

Response: In the regional scale model, the produced SOA sinks via the dry and the wet deposition. The UNIPAR model considers the dynamic oxidation of gas products under varying NO_x levels. In addition, the lumping species also sink to aqueous droplets (i.e., cloud) in the model and lose via the dry deposition. We agree with the reviewer in that SOA can be decomposed due to photolysis under the sunlight. The discussion about OA aging and the aerosol deposition has been added to the section 4, Atmospheric implications and uncertainties, and reads now.

"In addition, the deposition of SOA was estimated with the one size bin for the fine particulate matter. The different particle size can have different sink flux causing the uncertainty in the aerosol lifecycle of OM. The UNIPAR model is capable of the prediction of gas products aging but currently has no feature for OM aging."

49: citations are for box models, better suited in relation to this study are examples for the regional and global scale, e.g. Budisulistiorini et al., 2017 (IEPOX), Knote et al., 2015 (GLYOXAL) and Stadler et al., 2018 (IEPOX), Myriokefalitakis et al., 2008 (GLYOXAL), respectively

Sri Hapsari Budisulistiorini, Athanasios Nenes, Annmarie G. Carlton, Jason D. Surratt, V. Faye McNeill, and Havala O. T. Pye Environmental Science & Technology 2017 51 (9), 5026-5034 DOI: 10.1021/acs.est.6b05750

Knote, C., Hodzic, A., Jimenez, J. L., Volkamer, R., Orlando, J. J., Baidar, S., Brioude, J., Fast, J., Gentner, D. R., Goldstein, A. H., Hayes, P. L., Knighton, W. B., Oetjen, H., Setyan, A., Stark, H., Thalman, R., Tyndall, G., Washenfelder, R., Waxman, E., and Zhang, Q.: Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aerosol in a 3-D model, Atmos. Chem. Phys., 14, 6213–6239, https://doi.org/10.5194/acp-14-6213-2014, 2014.

Stadtler, S., Kühn, T., Schröder, S., Taraborrelli, D., Schultz, M. G., and Kokkola, H.: Isoprene-derived secondary organic aerosol in the global aerosol–chemistry–climate model ECHAM6.3.0–HAM2.3–MOZ1.0, Geosci. Model Dev., 11, 3235–3260, https://doi.org/10.5194/gmd-11-3235-2018, 2018.

Myriokefalitakis, S., Vrekoussis, M., Tsigaridis, K., Wittrock, F., Richter, A., Brühl, C., Volkamer, R., Burrows, J. P., and Kanakidou, M.: The influence of natural and anthropogenic secondary sources on the glyoxal global distribution, Atmos. Chem. Phys., 8, 4965–4981, https://doi.org/10.5194/acp-8-4965-2008, 2008.

Response: In order to respond to the reviewer, several references have been added to the revised manuscript and reads now (the 2nd paragraph in introduction).

"Several chemical transport models account for the aqueous reactions of few explicit products (i.e., glyoxal and IEPOX (epoxy diols form isoprene products)) that potentially may significantly impact the SOA formation (Ervens et al., 2011; Sumner et al., 2014; Budisulistiorini et al., 2017; Knote et al., 2014)."

51: citation to prove this claim?

Response: The references have been added to the revised manuscript and reads now (the 2nd paragraph in introduction).

"In particular, the current model applied to regional scales suffers from a substantial negative bias under high humidity conditions (Heald et al., 2011; Pye et al., 2017; Li et al., 2020)."

52: which "conventional model", not true in this broad claim form!

Response: The sentence has been modified in the revised manuscript and reads now (the 2nd paragraph in the introduction).

"The SOA model, such as the partitioning-base two product model, has no feature for SOA formation via aqueous phase reactions of different oxygenated products formed from various HCs."

56: all these citations are the reference for UNIPAR, or is there a single one that serves as reference? It needs to be made clear where UNIPAR is scientifically published.

Response: The sentence has been modified in the revised manuscript and reads now.

"The UNIfied Partitioning-Aerosol phase Reaction (UNIPAR) model was developed by Im et al. (2014) to predict SOA mass based on multiphase reactions of toluene and 1,3,5-trimethylbenzene, was developed. In the UNIPAR model, the products predicted using explicit gas mechanisms are lumped based on volatility and emerging chemistry in the aerosol phase. This UNIPAR model has been extended to various SOA originating from isoprene, terpenes, aromatics, and gasoline and demonstrated through the extensive photochemical outdoor smog chamber data (Beardsley and Jang, 2016; Cao and Jang, 2010; Zhou et al., 2019; Yu et al., 2021; Han and Jang, 2022)."

59: what is "arrayed" supposed to mean?

Response: Word "arrayed" has been changed to "estimated" (the 3rd paragraph in introduction).

"The model parameters linked to the thermodynamic properties and aerosol chemistry are also estimated according to the lumped species characteristics."

62: CAMx needs to be introduced (regional scale model...) and cited!

Response: This sentence has been modified in the revised manuscript and reads now (the 4th paragraph in introduction).

"In this study, the UNIPAR model was incorporated with the CAMx model (comprehensive air quality model with extensions, v7.10) (Environ, 2020) to predict the SOA formation in the regional scale during the Korean-United States Air Quality (KORUS-AQ) campaign that took place between 10 May, 2016 and 10 June, 2016."

75: SOAP is quite outdated - there should be more recent developments for CAMx that would better show the effect of UNIPAR over the _current_ state of science. See e.g. Jiang et al., 2021, for references.

Jiang, J., El Haddad, I., Aksoyoglu, S., Stefenelli, G., Bertrand, A., Marchand, N., Canonaco, F., Petit, J.-E., Favez, O., Gilardoni, S., Baltensperger, U., and Prévôt, A. S. H.: Influence of biomass burning vapor wall loss correction on modeling organic aerosols in Europe by CAMx v6.50, Geosci. Model Dev., 14, 1681–1697, https://doi.org/10.5194/gmd-14-1681-2021, 2021.

Response: Thank you for the suggestion. In the future, we will implement the updated modules and parameters. In this study, our focus is the demonstration of the importance of aqueous phase reactions of organic species to form SOA by suing the UNIPAR model.

75: Also, how do comparable model systems fare during KORUS-AQ? There is a good overview by Park et a., 2021, on multi-model results that should provide insights into how the model used here fares compared to others.

Rokjin J. Park, Yujin J. Oak, Louisa K. Emmons, Cheol-Hee Kim, Gabriele G. Pfister, Gregory R. Carmichael, Pablo E. Saide, Seog-Yeon Cho, Soontae Kim, Jung-Hun Woo, James H. Crawford, Benjamin Gaubert, Hyo-Jung Lee, Shin-Young Park, Yu-Jin Jo, Meng Gao, Beiming Tang, Charles O. Stanier, Sung Soo Shin, Hyeon Yeong Park, Changhan Bae, Eunhye Kim; Multi-model intercomparisons of air quality simulations for the KORUS-AQ campaign. Elementa: Science of the Anthropocene 21 January 2021; 9 (1): 00139. doi: https://doi.org/10.1525/elementa.2021.00139

Response: The focus of our study is to demonstrate the importance of multiphase partitioning of organic species and their aqueous reactions (151 lumping species) by using the UNIPAR model. In particular, the UNIPAR can estimate the activity coefficient of lumping species on aqueous phase allowing the impact of aerosol water mass on SOA formation. In addition, the simulation in this study was compared to the ground-based observations during KORUS-AQ campagin.

In order to respond to the reviewer, we discussed the recent model simulation of the organic aerosol in the regional scale in the second paragraph of introduction and in the last paragraph of Section 3.1

"Park et al. (2021) extensively evaluated the prediction of the organic aerosol produced during the KORUS-AQ campaign by using different air quality models, which were varying in chemistry mechanisms, aerosol thermodynamics, the types of SOA precursors, and the SOA schemes. In their study, the SOA formation was simulated with the SOAP, the 4 bin-base VBS or the 5-bin-base VBS modules. The predicted organic aerosol masses were, however, underestimated compared to observation data (HR-ToF-AMS) suggesting the limitation of the current SOA modules."

"For organic matter, the average Normalized Mean Bias (NMB, %) between model predictions and observations at the four monitoring sites are -50% for CAMx-SOAP and -39% for CMAx-UNIPAR. A similar level of the NMB ($\approx$ 46%) was reported in the previous simulation for the same campaign (Park et al., 2021)"

141 ff: are organic acids considered when calculating aerosol acidity? How good is your aerosol water content, as it is crucial for acidity calculations?

Response: The aerosol acidity and the aerosol water content both were estimated by using the ISORROPIA inorganic thermodynamic model. In general, many inorganic thermodynamic models use the ZSR relation to estimate water activity of the system that is directly related to predict aerosol water content (Stokes and Robinson, 1966; Zdanovskii, 1948). It is known that the estimation of water prediction is relatively accurate and similar between models. However, the calculation of the activity coefficient of the proton in the highly concentrated salted system are uncertain due to the lack of database and it is various between models as discussed in the previous studies (Jang et al., 2020; Pye et al., 2020). During the KORUS-AQ campaign, inorganic acids were significantly titrated, and aerosol acidity was near neutral. Thus, the aerosol water mass mainly influenced aqueous phase reactions of organics and their partitioning to aqueous phase.

142: typo "ISORRIPIA"

Response: This has been corrected.

"In order to process SOA formation in the inorganic aqueous phase, the inorganic composition and aerosol acidity are predicted by using the inorganic thermodynamic model, ISORROPIA (Fountoukis and Nenes, 2007), and then incorporated into the UNIPAR model. For the ISORROPIA model, mutual deliquescence relative humidity (MDRH) is predicted."

155: "MOZART", all caps

Response: This has been corrected.

"The boundary conditions were converted from the MOZART-4 global simulation results (https://www.acom.ucar.edu/wrf-chem/mozart.shtml) (Emmons et al., 2010)."

194: I would expect at least a short model evaluation for the main drivers of OA formation: meteorology (temperature, humidity, radiation), oxidants (O3, NOx) and precursors (aromatics, terpenes, isoprene). See also main concerns.

Response: Please find the response to the 3rd main comment above.

210ff: how well does your model capture the precursors you actually included? Measurements of aromatics, terpenes and isoprene should be available!

Response: As seen in Section 2.3.2, the emission of air pollutants was determined by using SMOKE from emission inventories originating from various sources (i.e., point sources, area sources, biogenic sources (MEGAN), automobiles non-mobile sources, etc). During the KORUS-AQ campaign, only few precursors were monitored (i.e., toluene). The predicted toluene was on average 94% of observed toluene.

356ff: "Furthermore, the UNIPAR model integrated with regional models enables better prediction of future SOA burdens under different scenarios of air pollutant emissions." This statement is too broad to be supported by the analysis shown here - why are you better equipped represent future scenarios better? Because you seem to compare better to 3 ground stations in one geographical corner of the world for 1 month in one year? Because you represent processes better? Address!

Response: The sentence has been removed.

Figure S5: do model and measurements coincide (i.e., the model is perfect), or might there be a difference in modelled vs. measured temperature, leading to differences in the thermodynamic environment that should be discussed? Figure S6: same question as for S6!

Response: Temperature and humidity in the model were obtained from the WRF simulation results and they accorded well with the measurements (R^2=0.999 for temperature and R^2=0.969 for relative humidity). This information can be found in the figure captions in Figures S5 and S6 in the revised manuscript.

References:

Beardsley, L. R. and Jang, M.: Simulating the SOA formation of isoprene from partitioning and aerosol phase reactions in the presence of inorganics, Atmospheric Chemistry and Physics, 16, 5993-6009, 10.5194/acp-16-5993-2016, 2016.

Budisulistiorini, S. H., Nenes, A., Carlton, A. G., Surratt, J. D., McNeill, V. F., and Pye, H. O. T.: Simulating Aqueous-Phase Isoprene-Epoxydiol (IEPOX) Secondary Organic Aerosol Production During the 2013 Southern Oxidant and Aerosol Study (SOAS), Environ Sci Technol, 51, 5026-5034, 10.1021/acs.est.6b05750, 2017.

Cao, G. and Jang, M.: An SOA Model for Toluene Oxidation in the Presence of Inorganic Aerosols, Environ. Sci. Technol., 44, 727-733, 2010.

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

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