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, inparticle 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 (α i) 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 HO₂ 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 Cor and Cin."

"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."

Reference

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