Atmospheric Chemistry and Physics Discussions

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Comment on acp-2021-1002

Anonymous Referee #2

Referee comment on "Secondary organic aerosol formation via multiphase reaction of hydrocarbons in urban atmospheres using CAMx integrated with the UNIPAR model" by Zechen Yu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1002-RC2, 2022

Review of Yu et al., "Secondary Organic Aerosol Formation via Multiphase Reaction of Hydrocarbons in Urban Atmospheres Using the CAMx Model Integrated with the UNIPAR model"

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.

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 photolyse, 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?

(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"!

(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!

Specific comments:

15ff "explicit" gas-phase chemistry?
37 why italic for "via"?
37 HC abbrevation, first mention, explain!
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 OVOCs (e.g., Hodzic et al, 2015)?
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
51: citation to prove this claim?
52: which "conventional model", not true in this broad claim form!
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.
59: what is "arrayed" supposed to mean?
62: CAMx needs to be introduced (regional scale model...) and cited!
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.
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.
141 ff: are organic acids considered when calculating aerosol acidity? How good is your aerosol water content, as it is crucial for acidity calculations...
142: typo "ISORRIPIA"
155: "MOZART", all caps
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.
210ff: how well does your model capture the precursors you actually included? Measurements of aromatics, terpenes and isoprene should be available!
356ff: 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!
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 S5!

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