Response to comments on "Development and implementation of SOMA: A Secondary Organic Module for Aerosol integration in high-resolution air quality simulations"

We would like to thank the reviewers for evaluating our manuscript and for providing their constructive comments. Below you can find point-by-point answers to the RC1 reviewer's comments. All references are provided at the end.

RC1: 'Comment on egusphere-2025-193', Anonymous Referee #1, 29 Jun 2025

Ioannidis et al. develop parameterizations for SOA mass yields using GECKO and laboratory data and combine those parameterizations with a CFD model to examine the spatial and source patterns of SOA in a targeted urban area. SOA is a complex pollutant with a multitude of sources and pathways. It contributes to PM1 significantly and there is a need to develop approaches that can simulate SOA formation in atmospheric models. Hence, the paper's need is justified. However, the paper's motivation (described in the introduction) is adequate but not strong. I also found the methods inadequate and confusing. The use of CFD seems novel but I contend that the use case to study SOA is not a good fit. Overall, the paper is weak, and I do not recommend publication of this paper in ACP. The comments below outline my most important objections, and I would strongly recommend the authors consider those as they think of novel applications for their CFD modeling.

Major comments:

Comment 1:

1. Even for highly reactive VOCs, the timescales for SOA production at ambient concentrations of OH and O3 are on the order of hours (even longer for species like toluene that has an e-folding lifetime of over 1.5 days). The motivation for using CFD in 'street canyons' to estimate SOA production – which is more regional - isn't quite strong. I remain highly skeptical of the primary finding that the SOA in a domain this small (1.8 km) is dominated by SOA from local sources within the domain. I suspect that CFD at this spatial scale would be much more useful in tracking the spatiotemporal evolution of primary particles and gases where transport and dilution are much more relevant than chemistry. In my opinion, to get at airshed level burdens of SOA, 0D box models (e.g., Hayes et al., ACP, 2015) and high-resolution chemical transport models (e.g., Pennington et al., ACP, 2021) are likely better tools to model the formation, evolution, and properties of SOA.

Response:

We appreciate the reviewer's concern regarding the spatial scale of SOA formation processes. We agree that SOA chemistry, especially for less reactive species like toluene, extends over larger spatial and temporal scales. However, the objective of our work is not to capture the full regional SOA burden, but to explore spatial heterogeneity and source attribution in urban hot-spots using a hybrid high-resolution approach. CFD is used not to simulate chemical

transformations directly, but to produce high-resolution VOC concentration fields and wind-dependent recirculation patterns that feed into SOMA.

To address the reviewer's concern more explicitly, we have revised the Introduction and Discussion sections to better position the purpose and limitations of using CFD in this context. In L94-96 we revised the manuscript to state: "While the full formation of SOA can span hours to days, initial oxidation steps begin within the first hours after VOC emission. In street canyon environments, turbulent recirculation can extend the residence time of reactive VOCs, enabling early-stage SOA formation within the local domain." Also, in L104-106 we explain further: "In this study, CFD is not used to explicitly simulate complex atmospheric chemistry, but rather to resolve realistic concentration and flow fields that feed into SOMA, which calculates SOA formation using corrected yield expressions."

Comment 2:

2. Figure 1: Details about the initial VOC, OH/O3 concentration, OA mass loading are all missing. Why are results shared in the methods section?

Response:
We thank the reviewer for this observation. Figure 1 displays the GECKO-A modelled SOA yields over 240 hours for four selected VOCs (toluene, limonene, isoprene, and α-pinene) under fixed conditions. These are not simulation results from our model but rather yield outputs from GECKO-A used as input data in SOMA. As such, we placed the figure in the Methods section to document the origin of the input data.

As stated in L133–135: "The SOA yields given by GECKO-A correspond to constant conditions of temperature (T) at 25°C, ozone (O_3) concentration of 40 ppb and relative humidity RH at 70% (Camredon et al., 2007)."

The initial mass mass loading is 1 pptvC, and this is now explicitly mentioned in the caption of Figure 1 in L136 for clarity:

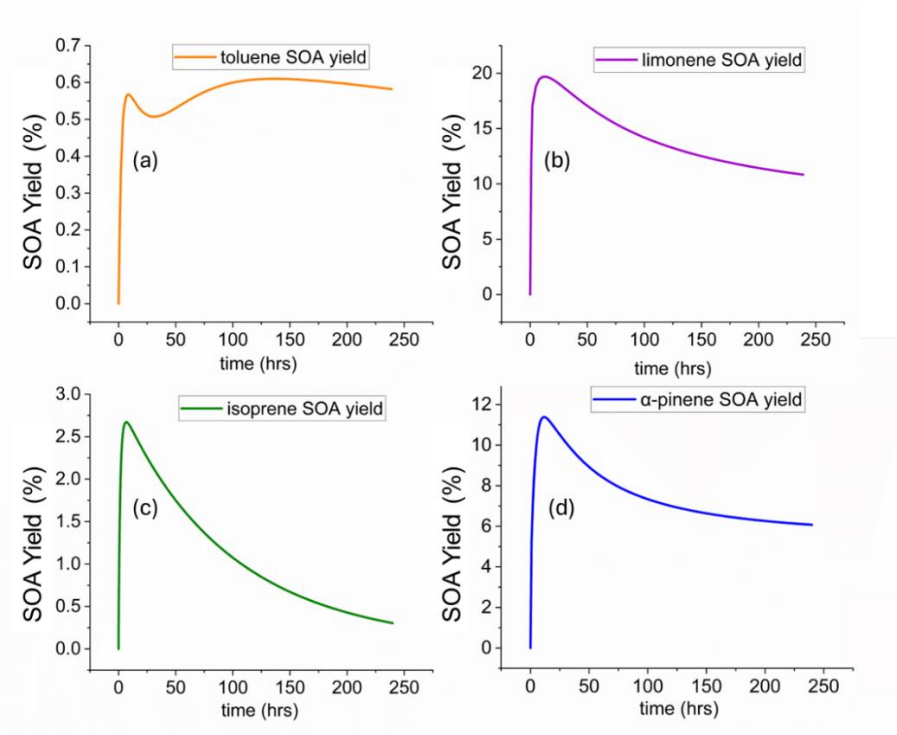

Figure 1. Time series of SOA yields from GECKO-A for urban conditions (NO_x=200 ppb), for toluene (a), limonene (b), isoprene (c) and α-pinene (d) for an initial organic mass loading of 1 pptvC.

Comment 3:

3. Sections 3.2-3.4: I don't understand the rationale for these methods. I see several glaring problems. First, toluene SOA has been widely studied and there 10s (if not 100s) of studies that have documented SOA mass yields. While there isn't an expectation to include every published study on toluene SOA, what is expected is a rationale for why these (i.e., Deng and Chen) were picked and how they are representative of the broader consensus about toluene SOA mass yields. Second, it is unclear why GECKO wasn't directly run for the same experimental conditions for initial VOC, NOx, RH, and T. Depending on the model-measurement performance, a case could have been made for parameterizing SOA_exp/SOA_org to differences in O3. Regardless, I would still be skeptical of this parametrization as no attempt was made to mechanistically explain why the model underestimates the measurements so substantially. Third, SOA is expected to be a strong function of the OA/SOA mass loading (depending on whether an organic seed was used to aid SOA condensation) and hence any model-measurement difference in yield is likely to also be a function of the SOA mass concentration (which in itself is the primary output that is being used to compute the yield). This non-linearity is probably the most difficult to resolve.

Response:

We thank the reviewer for raising these important points. The selection of the (Deng et al., 2017) and (Chen et al., 2022) studies was based solely on data availability: these are experiments we found that reported the required input parameters (VOC concentration, RH, temperature, NO_x, ozone, OH, duration, and final SOA mass), which were necessary for our calibration analysis. Our goal was not to provide a comprehensive representation of toluene

SOA literature but to build a consistent dataset for regression-based correction. To highlight this we state in the manuscript in L294-299: "To validate the SOA model and to provide reference for calibration, we gathered experimental information on the four mentioned VOCs in section 2.1. The selected experimental studies were limited to those that provided all required variables: VOC concentration, reaction time, NO_x, ozone, RH, temperature, OH concentration, and final SOA mass. This was necessary because our modelling approach with SOMA uses these specific environmental and chemical parameters as direct inputs to simulate SOA formation. Without complete documentation of these conditions, the experiments would not be suitable for generating accurate model inputs or for evaluating model performance."

We acknowledge the reviewer's question on why GECKO-A was not directly run under the same experimental conditions. In the revised manuscript in L289-292 to explain: "The fitting curve allows us to calculate the SOA yield of any given time-period, for any NO_x level. This interpolated fitting curve allows us to estimate SOA yield values at any NO_x level without rerunning the GECKO-A model, which is necessary since we do not have access to the GECKO-A code but only to its published yield output library."

Regarding SOA loading effects, we focused on experiments without organic seed aerosol to avoid complications related to condensation sinks and OA background levels. In L304-307 we state: "Both studies were also selected because they did not include seed aerosol, in order to avoid uncertainties related to partitioning onto pre-existing OA. The data collected for every compound serve as reference for validation and as a base for experimental fitting based on the GECKO-A constant conditions to correct the SOA yields used by SOMA."

Comment 4:

4. Section 4.1: I do not agree with how the model was setup for background/boundary values of toluene. I don't see how a uniform background toluene assumption is justifiable given that the concentrations inside the modeled domain vary spatially. The modeled domain is identical to the regions surrounding it so if toluene varies inside the domain, that fact should also hold for regions immediately surrounding the domain.

Response:

We thank the reviewer for raising this important point. We agree that assuming a spatially uniform background can oversimplify real-world variability, especially in a heterogeneous urban environment. In response, we have revised Section 4.2.2 to clarify the rationale and limitations of our assumption. Specifically, we added the following text:

In L428-433: "Certain VOCs can remain in the air for hours to days, allowing them to recirculate in urban areas and contribute to SOA production. This contribution often originates from neighboring areas and can accumulate over various time scales. To estimate the contribution of SOA originating from neighboring areas to the study domain, we assume that traffic patterns are consistent throughout the broader urban region due to similar driving conditions of urban environments. This assumption was supported by the fact that the modelled domain lies centrally within Augsburg and is surrounded by areas with comparable land use, emission sources, and road networks."

L472-474 we also state: “We assumed that similar emissions and meteorological conditions in the broader urban setting lead to comparable VOC and SOA levels in the surrounding areas, justifying their use as boundary input.”

These additions aim to make the reasoning behind our boundary setup transparent and acknowledge its potential impact on model results. We hope this adequately addresses the reviewer’s concern.

Comment 5:

5. The strong suit of this work is the CFD modeling and what can be learned from it. The development of methods to estimate SOA mass yields are clunky at best. This work’s Achilles heel is GECKO, which is great at gaining a fundamental understanding of multi-generational SOA chemistry and not a good fit for predicting SOA mass yields. Why not use published VBS parameterizations with NOx dependence to get at SOA mass yields directly? They may not be perfect but would work better than GECKO and allow the authors to focus on the CFD insights.

Response:

We appreciate the reviewer’s point regarding the potential utility of VBS parameterizations. Our choice to use GECKO-A was driven by the desire for consistency across experimental conditions (NO_x, RH, T), and because it provides chemically-resolved, compound-specific SOA yields that serve as a transparent input to SOMA. To support our approach we explain in the introduction section in L61-69: “SOA formation models are commonly used to simulate secondary organic aerosol production. One widely used approach is the Volatility Basis Set (VBS) model, which simulates the partitioning of semi-volatile and intermediate-volatility organic compounds (S/IVOCs) between the gas and aerosol phases (Sasidharan et al., 2023). While VBS frameworks are robust and widely adopted for regional-scale chemical transport models, they typically require the definition of volatility bins and yield parameterizations that must be fitted to experimental data, and they do not include explicit chemical reaction pathways. In principle, VBS approaches can account for environmental dependencies such as NO_x levels, temperature, or relative humidity by adjusting the bin yields, but this is often done indirectly and can require significant fitting effort. In contrast, our approach uses SOA yields generated by the GECKO-A model, which includes detailed multi-generational oxidation chemistry and accounts for precursor-specific reaction mechanisms.”

To acknowledge the limitation of our approach but enhance our stand we state in L69-72: “We acknowledge that GECKO-A is not optimized for direct SOA mass prediction under atmospheric conditions; however, its mechanistic representation provides a chemically consistent foundation from which SOA yields can be generated.”

We appreciate this valuable suggestion and will explore using VBS-based formulations in future iterations of our modeling approach.

Minor comments:

Comment 7:

Line 32: I haven’t seen ‘OHs’ written in plural.

Response:

We corrected that inconsistency in the manuscript in L33-35: "OH react with VOCs, initiating chemical transformations that reduce VOC volatility and promote their condensation onto existing particles or formation of new ones (Hallquist et al., 2009)."

Comment 8:

Line 111: Provide references for the NOx levels used to generate GECKO output. Same goes for the choice of RH and T.

Response:

The GECKO-A output library's details are available online here: https://www2.acom.ucar.edu/modeling/gecko/details

In L130-135 we modified the manuscript to include a reference of the early development of GECKO: "The GECKO-A model produces SOA yields for five distinct NOx concentration levels, corresponding to remote (0.002 ppb), remote continental (0.0025 ppb), continental (5 ppb), polluted continental (20 ppb) and urban (200 ppb) environments (Camredon et al., 2007). Users can select a specific VOC and a NO_x pollution scenario, and then visualize how SOA yield changes over time, with temporal resolution ranging from seconds to hours. The SOA yields given by GECKO-A correspond to constant conditions of temperature (T) at 25°C, ozone (O_3) concentration of 40 ppb and relative humidity RH at 70% (Camredon et al., 2007) ."

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