We thank Referee 2 for the thoughtful and constructive comments on our manuscript. We appreciate the detailed feedback and suggestions, which have helped us improve the clarity and completeness of the paper. In the following, we provide a point-by-point response to each comment.

Referee comment: Sect. 2.1.1: The detailed description of the S2AS tool is appreciated. For people not familiar with AIOMFAC use, it could be useful to explain what is the improvement over existing methods. How were organic molecules decomposed into AIOMFAC subgroups before S2AS? I am afraid to learn it had to be done manually, in which case the authors could highlight the reduction in potential human error (and the time saved!) as an additional benefit from this tool.

Authors' response: We will clarify the novelty of S2AS relative to previous practice. Before S2AS, assigning AIOMFAC functional subgroups to an organic molecule was indeed often a manual or semi-manual process, feasible only for relatively small sets of compounds. The UManSysProp online tool (Topping et al., 2016) did include some automated subgroup assignment functionality via SMARTS matching, but as we noted (Sect. 2.1.2), the SMARTS patterns and logic in UManSysProp differ from ours and are not directly transferable. In particular, there was no publicly available general tool to parse any SMILES into AIOMFAC subgroups comprehensively. Thus, S2AS provides a significant improvement by automating this mapping for large numbers of molecules, with a carefully curated priority list of SMARTS patterns and consistent handling of exceptions (Sect. 2.1.1 and the discussion of findings in Sect. 3.2.1). This reduces the potential for human error and saves a substantial amount of time when dealing with thousands of species.

Changes to the manuscript: In Section 2.1.1, at the end of the first paragraph about S2AS, we added: "Previously, AIOMFAC subgroup assignments for organic molecules had to be determined either manually (for small sets of structures) or using limited tool-specific pattern matching (e.g., the UManSysProp facility; Topping et al., 2016). Our S2AS tool automates this process for arbitrary molecules. It can process tens of thousands of compounds in a consistent way, whereas manual assignment would be prohibitively laborious and prone to errors or inconsistencies"

Referee comment: Fig. S1: The general effort to illustrate this paper is appreciated, but I fail to see the purpose of this figure. Unless I missed something, it seems very trivial and does not benefit the understanding of Sect. 2.2.1.

Authors' response: We agree with the referee that Fig. S1 does not add substantial value to the understanding of Section 2.2.1. We have therefore removed this figure from the Supplement.

Changes to the manuscript: Figure S1 has been removed from the Supplement and related text in Sect. S1 adjusted. The reference to this figure in Sect. 2.2.1 of the main text has also been deleted.

Referee comment: Sect 3.1: I guess the lumping and AIOMFAC calculations are carried out for the mixture obtained at the end of each of the example simulations. It would be important to clarify

this. With this remark come questions that may be addressed in the conclusion: how would this framework be applied when time-stepping through an atmospheric chemistry model? Would the surrogate species be recomputed at each call to AIOMFAC? How would the gas-aerosol partitioning of surrogate species be applied back to the explicit gas species to initialize the next gas chemistry timestep while ensuring mass conservation?

Authors' response: We confirm that the lumping and AIOMFAC calculations were performed on the product distributions at a specific (final) time of the simulations. We have clarified this in Section 3.1. We have added a new paragraph in the Conclusions section (see below) outlining several options for how the 2D framework could serve in offline or online applications of atmospheric chemistry and gas–particle partitioning models.

Changes to the manuscript: Section 3.1 (first paragraph) now includes a sentence clarifying that the lumping was applied to the data from the final time step of the mechanism simulations. The Conclusions section was expanded by the following paragraph.

We envision a few distinct options for future applications of this framework in different kinds of atmospheric chemistry models. (1) Within detailed chemical box or plume models, those that consider a large number of compounds and retain their molecular structure information, the computation of surrogates and subsequent gas–particle partitioning at each desired (output) time step may be the preferred option. (2) Alternatively, based on a separate offline calculation for a specific system, a fixed set of surrogate compounds could be determined with the 2D framework. Subsequently, at each time step, existing and newly formed compounds from the box model's chemical mechanism could be mapped to this conserved, predetermined set of surrogates using the closest normalized Euclidean distance to the various surrogates in the 2D space (similar to Eq. 6) to determine the surrogate to which a compound's mass will be lumped. (3) In the case of simplified chemical mechanisms, such as those often employed in large-scale chemical transport models, maintaining only a few organic aerosol surrogates or a 1D/2D VBS representation, the application differs since surrogate lumping during simulations is unnecessary. In that case, the 2D framework could serve in systematically generating sets of surrogate components after mechanism simulations (e.g. with GECKO-A) for targeted aerosol precursors (structure-resolved) or aid in generating 2D VBS bin-resolved (structure-agnostic) representations at desired polarity–volatility resolutions. In the latter case, the 2D lumping step may serve in assigning surrogates in the ACR vs. p° space and in translating the resulting surrogate mass concentrations into bin-based mass concentrations, e.g. in the O:C vs. C° coordinate space. In the case of atmospheric chemistry models that retain the molecular structure information of surrogates, we envision two options for invoking equilibrium gas–particle partitioning calculations during simulations. (i) Applying the gas–particle partitioning calculation offline at specific output times during a simulation while running the gas-phase chemical mechanism as if all material remained in the gas phase (no feedback from partitioning). (ii) Running the 2D lumping framework and the gas–particle partitioning method at every simulation time step, followed by treating the determined fractional surrogate amounts partitioned to the particle phase as partially or fully shielded from further gas-phase chemical reactions. The gas-phase fraction of a surrogate would then be applied to the list of associated compounds, updating their molecular

gas-phase concentrations prior to the next chemical reaction step in the simulation. Optionally, reactions in the condensed phase could be treated separately by a distinct mechanism.

Referee comment: Conclusion (l.708–715): I agree with the authors that this new framework is a good step toward reducing the complexity of organic chemistry models for application in large-scale models. However, as it is described here, this framework cannot be applied in a large-scale model because it still relies on the explicit description of gas phase organic chemistry. Could the authors please expand on how they see their tools being used in the future? For instance, could this framework be treated as a reference when creating future 2D-VBS-like parametrisations? Is there any chance this type of approach could be used to simplify the gas component?

Authors' response: We have expanded the Conclusions section to discuss how our framework may serve for developing reduced SOA mechanisms, including for 2D-VBS applications (see response to the previous comment). Regarding simplifying the gas component, we assume that this question is about simplifying a gas-phase chemical mechanism. Such applications are outside the scope of this study. However, existing and new approaches for systematic chemical mechanism reduction, such as the GENOA (Wang et al., 2022) and AMORE (Wiser et al., 2025) approaches, could potentially benefit from including our 2D lumping when categorizing reactants during mechanism reduction steps.

Changes to the manuscript: The Conclusions section was revised to include the following paragraph.

A computationally effective use of near-explicit gas-phase chemical mechanisms in atmospheric chemistry models benefits often from a tunable reduction in the complexity of the mechanism itself, both in terms of number of explicit species and number of reactions covered. Methods such as the GENerator of reduced Organic Aerosol mechanism (GENOA) (Wang et al., 2022) and the Automated MOdel REduction (AMORE) algorithm based on graph theory (Wiser et al., 2025) serve this purpose. When targeting SOA formation applications, AMORE v2.0 employs a 2D categorization based on the saturation vapour pressures and Henry's law constants of organic components, which is similar to the polarity–volatility space of our 2D framework. Further development of such rule-based mechanism reduction methods may therefore benefit from considering also our 2D framework for potential application in compound classification.

Referee comment: Technical Corrections:

1. l. 320: dipol-dipol → dipole-dipole.
   Authors' response: Corrected to dipole–dipole" in Section 2.2.1.
2. l. 350 "Of note, . . . AIOMFAC,": confusing sentence, missing a word?
   Authors' response: Revised the sentence for clarity as described in response to Referee #1.
3. l.611–612: this is repeating what is written in l. 608.
   Authors' response: Sentence on line 608 shortened to: "Additionally, the relatively small modelled SOA mass concentrations contribute to the observed metric fluctuations, since minor

absolute differences can result in larger relative errors."

4. l.716–719: a bit pompous, is this paragraph really needed?
   Authors' response: We deleted this final paragraph of the Conclusions section.

5. l.737: I am almost certain that Bernard Aumont is a professor, please check.
   Authors' response: This comment may reflect different norms. In the Acknowledgements, we had referred to "Dr. Bernard Aumont", which is consistent with common practice in acknowledgements. While Bernard Aumont holds a professorial position, we prefer to retain the current phrasing or simply refer to "Bernard Aumont" without a degree/title, whichever is preferred by the GMD journal. This aligns with typical conventions in scientific acknowledgements, where ranks/roles/titles such as "Professor" are often omitted or replaced with "Dr." to reflect earned academic degrees rather than job titles.

References

Wang, Z., Couvidat, F., and Sartelet, K.: GENerator of reduced Organic Aerosol mechanism (GENOA v1.0): an automatic generation tool of semi-explicit mechanisms, Geosci. Model Dev., 15, 8957–8982, https://doi.org/10.5194/gmd-15-8957-2022, 2022.

Wiser, F., Sen, S., Wang, Z., Lee-Taylor, J., Barsanti, K. C., Orlando, J., Westervelt, D. M., Henze, D. K., Fiore, A. M., Berman, A., Carter, R., and McNeill, V. F.: A graph theory-based algorithm for the reduction of atmospheric chemical mechanisms, PNAS Nexus, 4, 11, https://doi.org/10.1093/pnasnexus/pgaf273, 2025.