We thank Referee 1 for the positive and constructive comments on our manuscript. We have carefully considered all suggestions and have revised the manuscript accordingly. In the following, we provide a point-by-point response to each comment.

**Referee comment:** The paper is a little redundant in spots and thus could be shortened (e.g., line 362).

**Authors' response:** We have revised Section 2.2.1 and 2.2.2 to remove redundant phrasing and streamline the description of the surrogate selection methods.

**Changes to the manuscript:** In Sect. 2.2.1 (formerly lines 326–328) the sentence "Figure S1 illustrates the binary solution ..." was deleted since the former Fig. S1 has been removed (see also response to a comment by referee 2). Lines 345–347, the sentence "Water represents a highly polar ..." was deleted as it is redundant given similar statements prior to that description. In Section 2.2.2, we have removed the sentences on line 362 and revised the following sentence to read: "The 2D space is subdivided into a number of grid cells (or clusters) based on the targeted reduction in system complexity."

**Referee comment:** It would be helpful to provide the reader with an approximate conversion from saturation vapor pressures (in Pa) to the C* variable (in $\mu g/m^3$) that is common in literature for VBS. I know this depends on molecular weight. I assumed a 200 g/mol molecular weight and used gas law to create an equivalent scale to help me interpret the figures. Maybe the authors could consider labeling an upper x-axis with C* assuming an average molecular weight.

**Authors' response:** We have included a secondary $x$-axis to the affected figures showing the approximate $C_j^\circ$ values (in $\mu g/m^3$) at the top corresponding to the main $x$-axis stating pure-component vapour pressure. Instead of simply assuming an average molar mass in the scale conversion, given the relatively large volatility and molar mass ranges covered, we opted for a slightly more accurate conversion by fitting a linear molar mass to vapour pressure relationship specific to the system of interest. This approach is now described in the supplement and the code to produce such conversions is included in the code repository of the 2D polarity–volatility framework. We also note that the $C_j^\circ$ values of individual compounds are computed and part of the output of the 2D framework.

**Changes to the manuscript:** Figures 7–11 were updated to include a secondary $x$-axis showing $C_j^\circ$ ($\mu g/m^3$) as alternative volatility metric. The caption of Fig. 7 was updated to include a brief description of this secondary axis. A new section S2 was added to the revised supplementary material document, describing the details of the conversion from a vapour pressure to an approximate $C_j^\circ$ axis. That section also includes a new Fig. S1 (included below), which shows the relationships between molar mass and saturation vapour pressure and between component saturation vapour concentration and saturation vapour pressure at $T = 298.15$ K for the two example SOA systems introduced in the main text.

**Referee comment:** The clustering algorithm seems like a novel approach with solid results. Even

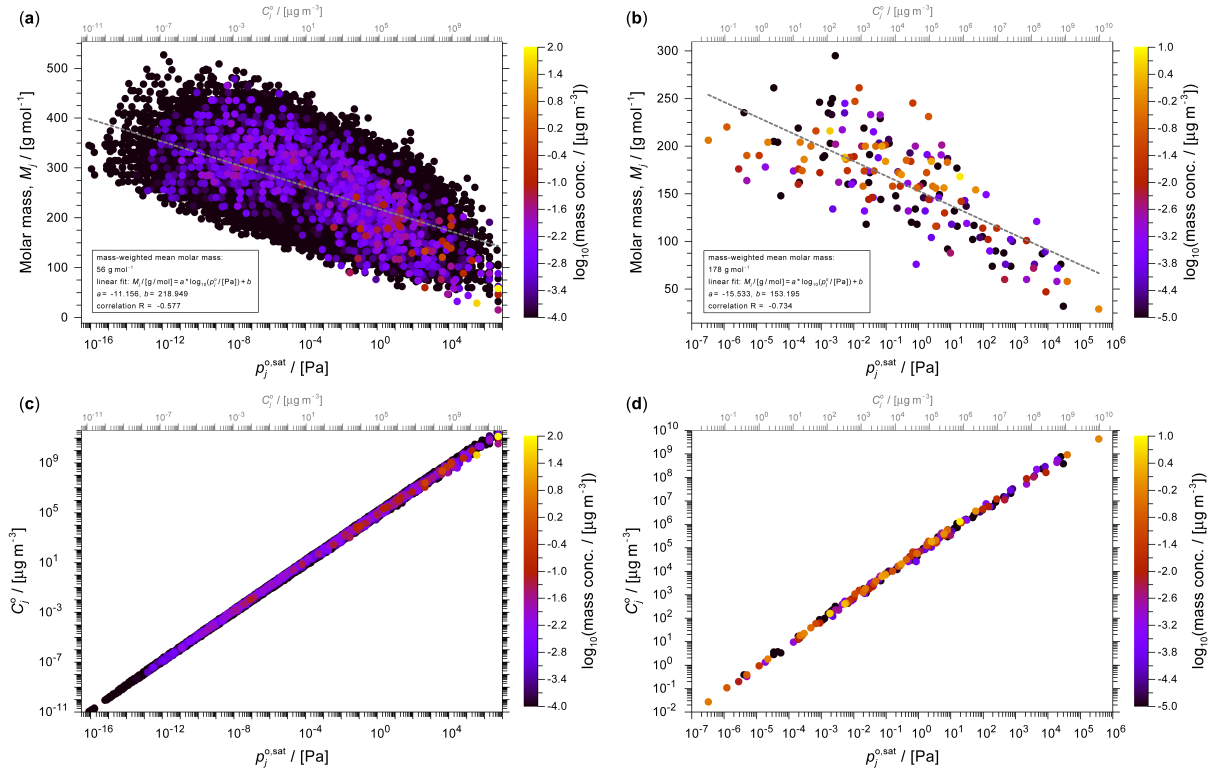


Figure S1: Relationships between molar mass and saturation vapour pressure (a, b) and between component saturation vapour concentration and saturation vapour pressure at $T = 298.15$ K. Panels (a) and (c) for the toluene-derived and panels (b) and (d) for $\alpha$-pinene-derived systems of gas-phase reaction products. The linear fit in (a, b) is used in the conversion from the lower to upper $x$-axes in these and related figures.

the simplified topologies give reasonable errors.

**Authors' response:** We thank the referee for this encouraging feedback and we agree that even relatively coarse resolutions give reasonable predictions. No changes were made in response to this comment.

**Referee comment:** I have one recommendation. I am wondering if the authors could create an additional table with results from a 1x3 parameter space. Where the 3 separates mass between water soluble, partially water soluble and water insoluble. Can the authors calculate the fraction of product mass and average ACR in these 3 bins for the two precursors (toluene, $\alpha$-pinene)? Or if the readers feel more appropriate using 4 polarity bins to better resolve partial solubility space. I think this info would be very helpful to constrain chemical transport models. Most chemical transport models only resolve the 1D volatility space. Or maybe the mass concentration data from Figures 8b and 10b can be provided in tables so that readers could manipulate data to their model needs.

**Authors' response:** Rather than presenting a 1×3 polarity binning in the manuscript, to address this comment, we suggest that readers can analyze the output from, e.g. the 6×3 or 10×5 (volatility × polarity) grid for a system of interest. Data tables related to Figs. 8b and 10b have been

added to the supplement. We add that the provided code repositories enable running the framework with a 1×3 grid space resolution, if desired. Furthermore, for the presented two systems (products from toluene or $\alpha$-pinene oxidation), the lumping framework output for different resolutions are provided in the Zenodo repository at `https://doi.org/10.5281/zenodo.17088391` (mentioned in the *Code and data availability* section). For example, the toluene-derived data for the 6×3 grid and weighted medoid lumping are provided in that Zenodo repository under toluene > ACR_P0 > lumping_output > 6by3 > "LumpedConc_1263_Weighted_Medoid_06x03.txt" and "SystemCompProp_1263_Weighted_Medoid_06x03.txt". Those two files also provide additional information about the properties of these surrogate compounds, e.g. their molar masses and $C_j^\circ$ values.

Concerning the interpretation of the ACR metric in terms of water soluble, partially water soluble and water insoluble fractions, we caution that in case of a phase-separated aerosol, a polar organic compound may still predominantly partition into the organic-rich phase when the aqueous electrolyte-rich phase is of high ionic strength and/or when the organic-rich phase is at least moderately polar and greater in mass than the electrolyte-rich phase. Thus, while the ACR of a component is a pure-component property and indicator of phase polarity preference, the precise partitioning preference will also be affected by the abundance and properties of the other compounds in a specific (two-phase) aerosol system, as discussed by Zuend et al. (2010).

**Changes to the manuscript:** We have added Tables S5 and S6 in the Supplement, which summarize the surrogate mass concentrations and their vapour pressure and ACR values for the 10×5 resolution cases shown in Figs. 8b and 10b in tabular form. These additions are referenced in the related Figure captions of 8b and 10b as well as in Section 3.3 of the revised manuscript.

**Referee comment:** Please check the grammar on line 351 after words "activity coefficients ..."

**Authors' response:** The sentence has been revised; a few words were missing by accident.

**Changes to the manuscript:** In Section 2.2.1, we corrected the sentence to read: "Of note, for a system consisting of tens of thousands of organic components, computing activity coefficients of all species simultaneously using the related multicomponent mixtures (containing as many components) with AIOMFAC, is prohibitively slow due to the many functional groups present, all the possible group–group and molecule–molecule interactions that need to be summed over and the associated computer memory requirements."

### References

Zuend, A., Marcolli, C., Peter, T., and Seinfeld, J. H.: Computation of liquid-liquid equilibria and phase stabilities: implications for RHdependent gas/particle partitioning of organic-inorganic aerosols, Atmos. Chem. Phys., 10, 7795–7820, https://doi.org/10.5194/acp-10- 7795-2010, 2010.