

The manuscript titled “Modeling studies of SOA formation from α -pinene ozonolysis” describes a kinetic gas-particle partitioning approach for simulations of SOA formation. Further the authors perform a range of sensitivity tests their they evaluate the sensitivity of this approach (model) to the particle phase bulk diffusion coefficient, the first order particle reaction rates and the inclusions of gas-phase HOM formation. The model is finally implemented for simulations of smog chamber SOA formation experiment.

General/major comments:

The manuscript is generally well written and addresses an important and interesting topic. However, the theory and assumptions behind the derived Eq. 2 and 3 is not easy to follow by only reading Sect. 2.2.2. E.g. for me it is not possible to understand what the Q_i steady state term stand for and what the overall gas-side mass transfer coefficient is ($K_{g,i}$). If the theory is taken directly from Zaveri et al., 2014 then I suggest that you remove Eq 2-4 from Sect 2.2.2 and only put the equations in Appendix A where you describe Q_i and $K_{g,i}$. In Sect. 2.2.2 you instead just mention that you have implemented and used the model/theory from Zaveri et al., 2014.

To me it is not clear what is new with the model approach presented in this study compared to Zaveri et al., 2014. The only new detail if I understand it correctly is the consideration of reversible particle phase reactions. The authors state that the kinetic gas-particle approach has been implemented in SPACCIM model and that it can be used for 3D-Eularian model simulations but this is never tested or evaluated. What is evaluated is the kinetic gas-particle approach from Zaveri et al., 2014 if I understand the manuscript correctly. If this is the case it should be clearly stated and the Sect 2.2.1 could be replaced with one sentence where it is stated that evaluated kinetic gas-particle approach from Zaveri et al., 2014 has been implemented in SPACCIM. But since I don't think you don't test how the approach is working or supposed to work in SPACCIM I don't see the point in describing this model in detail. E.g. you write in Sect. 2.2.1 that SPACCIM considers size-resolved particle and cloud droplet formation, evolution and evaporation using a one-dimentional sectional approach. But this is not used in the current study where you only consider one particle size at the time and if I understand it correctly you use a fixed particle radius in the model despite that you simulate SOA growth experiments where the particles growth over time. If you state that the tested approach will be used in the particle and cloud droplet size resolved model SPACCIM you as referee/reader want to see some results where you demonstrate how the model can simulate the particle number size distribution evolution during some SOA experiments and how the different model parameters (e.g. bulk diffusion coefficient and particle phase reaction rates) influence the particle number size distribution evolution. E.g. as in the study by Zaveri et al. (2014). There are a range of more advanced models for smog chamber SOA formation simulations such as KM-GAP and ADCHAM that the authors refer to and the model from Zaveri et al. (2014) which I think already have been implemented in regional and maybe even global chemistry transport models. Thus, the authors need to clearly demonstrate what is unique and

important with their approach. In the current form of the manuscript this is not clear.

I understand that the efficiency of the reactive uptake will be different for liquid and semi-solid particles but I don't understand how the phase-state can have such a tremendous influence on the SOA formation if the particle phase reactions are negligible slow (Fig 1b and 4c). To me the SOA phase state should not have a large influence on the SOA formation as long as the particle surface layer is composed of amorphous SOA material with the same composition as the SOA bulk. Then based on Raoult's law the saturation concentration of any gas-phase species above the particles would not differ between semi-solid and liquid particles. Even if the particles would be composed on solid crystalline salts (e.g. dry ammonium sulfate seed particles) organic molecules start to grow these particles if the gas-phase concentration of some organic species reaches above their pure-liquid saturation concentration. But the current model approach does not seem to capture this. I wonder if there is some fundamental assumption that is wrong/limiting the use of Eq. 2 and 3?

It is not clear how the SOA material that are formed after the heterogeneous reactions are treated in the model. Is it assumed to be non-volatile but still part of the amorphous SOA phase that allows more dissolution of SVOCs into the particle phase? This needs to be explained.

The model framework does not take into account the Kelvin effect if I understand this correct. If this is the case it cannot be used to study new particle formation. Please clarify and clearly state this if this is the case. I don't understand how you can assume that the particle radius is fixed. In any SOA new particle formation experiment (without seed) the particle size will grow from initially around 1 nm to larger sizes. I am skeptical to weather this model framework can handle this size dependent particle growth? Doesn't the model framework handle the gradual growth of the particles and can it take into account coagulation?

How realistic is it to represent a dimerization or oligomerization process as a first order reaction. Dimerization will involve two organic monomers. Please clearly explain what the first order particle formation reactions are supposed to represent in the model. I would also like to see some reference on what values of k_c that has been used in previous studies, I am sure that is also exists some experimental evidence of appropriate values and what reactions it may be.

In Sect. 3.4 you describe an approach of how to estimate a weighted particle-phase bulk diffusion coefficient that considers particle composition (Eq. 10). I don't understand this approach. Is D_m referring to the diffusion coefficient of the organic compounds in the particle-phase? Is it further assumed that the organic compounds are water-soluble then and that the particles are composed on one SOA+water+inorganics phase? I think that the particles often will be composed of several phases (e.g. one hydrophobic organic phase and one water + inorganics + some water soluble organics phase). I think that differences in D_{org} over time can also be due to particle phase oligomerization processes that gradually increase the average organic molecular mass. But this Eq. 10 does not

take this into consideration. To me Eq. 10 contains too many assumptions and is not evaluated properly to be able to be justified.

Minor comments:

Page 2, Line 17-18: I think this sentence needs to be reformulated.

Page 2, Line 25-28: This sentence is a bit hard to understand/follow. Can you reformulate it?

Page 2, last word, remove "the"

Page 3, Line 33. The molar yields are not 6 %.

Page 4, Line 2-3. I don't understand this sentence. I understand that the diffusion coefficient will depend on the composition but not how it depends on increasing organic matter. Indirectly it can of course be influenced by the organic mass since this in turn can influence the composition.

Sect 2.1. Here you describe the experiments and the instruments that were used. What I am missing is the information about the size of the smog chamber (volume) and the wall material (e.g. Teflon) and I also miss information about how the measurement results are used in the present manuscript. E.g. how were the measurements used to derive the SOA mass in Fig 9a? Also I miss a discussion concerning VOC and particle wall losses that is known to be important in chambers.

Sect. 3.5. Why did you decide to use $k_c = 10^{-2} \text{ s}^{-1}$ and $k_c = 10^{-3} \text{ s}^{-1}$ and $D_{\text{org}} = 10^{-12} \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{org}} = 10^{-14} \text{ m}^2 \text{ s}^{-1}$, respectively?

Table S4. I am missing a unit on the pure liquid saturation vapor pressures of the HOM. I also think that you should mention that these molecules only represent the HOMs but that the HOMs is a family of many organic molecules presumably formed from autoxidation with a wide range of volatility. Can you justify why you decided to use these specific HOM molecules and specify how they are assumed to be formed? It is not either clear how these three different HOM are used in the model because in the gas-phase mechanism you only seem to have one HOM molecule.

Eq. 1. Is A_i representing a concentration of a species or is A_i a solute as stated on Line 17? In Eq. 2 C is used to represent concentrations.

The title is a bit imprecise. There exist many model studies of SOA formation from α -pinene ozonolysis. Can you make it more precise?