

Interactive comment on “Mass Accommodation and Gas-Particle Partitioning in Secondary Organic Aerosols: Dependence on Diffusivity, Volatility, Particle-phase Reactions, and Penetration Depth” by Manabu Shiraiwa and Ulrich Pöschl

Anonymous Referee #4

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This manuscript introduces a quantity termed as "effective mass accommodation coefficient" which essentially accounts for particulate phase transport and reactions unlike the original mass accommodation coefficient introduced by Fuchs. The authors argue that such a quantity will be useful for e.g. large-scale modeling applications aiming to understand secondary organic aerosol (SOA) instead of having to conduct explicit calculations resolving the particulate phase and the gas-particle interface at different conditions. This manuscript is a welcome addition to the discussion on the kinetics related

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to SOA formation and growth, and fits well within the scope of ACP. I appreciate the clear distinction between the effective mass accommodation coefficient defined here and the "original" mass accommodation coefficient, as these concepts have unfortunately been often confused in recent literature dealing with SOA kinetics. I recommend publication in ACP after the following issues have been adequately addressed:

1. My main comment is related to whether it is appropriate to term the new coefficient an accommodation coefficient to correctly represent the targeted phenomena (i.e. the particle phase transport and chemistry at the interface region). According to the original Fuchs-Sutugin formulation (Eq. 2 in the manuscript) the accommodation coefficient is a quantity relevant in the kinetic regime and its impact on the mass flux towards the particle disappears at the limit of small Knudsen numbers (i.e. for large particles if pressure is assumed constant). Will the presented formulation of the effective mass accommodation coefficient give the correct dependence on the particle size? Is it physically correct that the effect of the effective accommodation coefficient also disappears at the limit of $Kn \rightarrow 0$? The authors should elaborate on this and justify their choice of representing the particle phase phenomena as an effective accommodation coefficient instead of a flux correction factor.

2. The authors end the abstract with a rather strong statement: "Our findings challenge the approach of traditional SOA models using the Fuchs-Sutugin approximation of mass transfer kinetics with a fixed mass accommodation coefficient regardless of particle phase state and penetration depth. The effective mass accommodation coefficient introduced in this study provides an efficient new way of accounting for the influence of volatility, diffusivity, and particle-phase reactions on SOA partitioning in process models as well as in regional and global air quality models." At the same time, the authors also acknowledge the fact that the particle-phase transport is only relevant at rather low RHs. While it is true that at some conditions (like in the free troposphere) the semi-solid state of the SOA is highly relevant, I would suggest softening the statements related to the implications of this study for global and regional SOA modeling

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when it comes to conditions representative of surface-level RH and temperature.

3. Lines 94-97: The authors state: "Molecular dynamics simulations (Julin et al., 2014; Von Domaros et al., 2020) and a recent SOA chamber study (Liu et al., 2019) suggest that the mass accommodation coefficients for semi-volatile organic molecules on organic substrates are close to unity. Measurement-derived mass accommodation coefficients reported from thermodenuder investigations of SOA volatility distributions, however, were one to three orders of magnitude lower (Kostenidou et al., 2018; Lee et al., 2010; Saleh et al., 2011)." I think it should be noted that e.g. the studies by Lee et al. and Saleh et al. have been subject to a relatively high uncertainty in the assumed saturation concentrations of the studied species (e.g. at the time of these studies the auto-oxidation reactions generating ELVOCs in SOA mixtures were not established like they are today). Therefore, I think these experimental studies studying complex SOA mixtures are hardly comparable to the more recent MD simulations and laboratory studies. Please revise to present a more relevant comparison.

4. The presented effective mass accommodation coefficient is dependent on a variable called the "penetration depth". How should this parameter be defined for ambient SOA mixtures? This is rather important for the usefulness of the proposed approach and further elaboration on this would be important in the discussion of the results.

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