Response to Referee comments (comments in black, response in blue)

Anonymous Referee #3
To date, Fuchs-Sutugin (F-S) approximation of mass-transport kinetics at the gas-particle interface with a fixed mass accommodation coefficient has been commonly in many models. In this work, the authors introduced an effective mass accommodation coefficient which considers penetration depth, surface accommodation coefficient, volatility, bulk diffusivity, and particle-phase reaction rate constant. They also suggest that this new coefficient and its dependence on these various parameters should be considered in the future when the F-S approximation is used to simulate gas-particle C1 interactions, in particular for viscous or semi-solid organic aerosols, which are commonly found in the atmosphere. The paper is well written and concise. It also provide new method for simulating SOA formation and evaluation. I support the publication of this work in ACP and have some minor comments below.

We thank this Referee for the review and positive evaluation of our manuscript.

Comments Line 194, "We simulate the temporal evolution of partitioning and equilibration of semivolatile organic compounds (SVOC) with C_0 = 100 µg m^{-3} and D_g = 0.1 cm^2 s^{-1} interacting with non-volatile seed particles with a number concentration of 5000 cm-3, an initial diameter of 200 nm, and a surface accommodation coefficient as = a(0) = 1. For the SVOC, we assume initial gas- and particle-phase concentrations of 2 µg m^{-3} and 0 µg m^{-3}, respectively. The particles are assumed to be either liquid with a bulk diffusion coefficient D_b = 10^{-7} cm^2 s^{-1} or semisolid with D_b = 10^{-15} cm^2 s^{-1}." Can the authors elaborate and justify why these parameters are chosen for their simulations? A typical condition for ambient conditions or laboratory studies?

We chose these values as they are typical values of SVOC volatility and viscosity for SOA based on previous measurements. In addition, these values were used in Zaveri et al. (2014) and it is easy to refer and compare with this study. We clarify this point in the revised manuscript.

Line 214, "For liquid particles with fast surface-bulk exchange and bulk diffusion (D_b = 10^{-7} cm^2 s^{-1}), a(x) remains close to as = a(0) = 1, and all models yield the same result of fast mass transfer from the gas to particle phase and equilibration within one second." Have the authors shown these results in the manuscript?

For simplicity and legibility of Fig. 1a, we do not show these results in the manuscript, but all model lines obtained for this scenario are overlapping as shown below. We clarify this point in the revised manuscript.
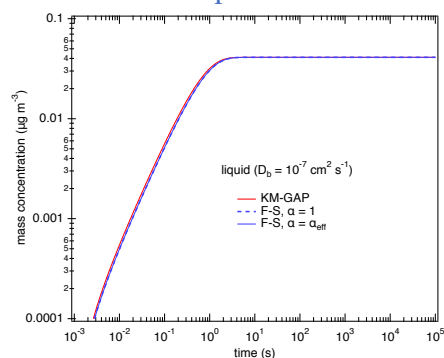
Line 235, "At long timescales (> 1 h), the partitioning is reasonable well captured by both the MOSAIC approximation using a two-film approach of bulk diffusion (Zaveri et al., 2014) as well as the simple F-S approximation accounting for the influence of penetration depth with the effective mass accommodation coefficient, aeff, newly introduced this study." Can the authors comment how the simple F-S approximation accounting for the influence of penetration depth with the effective mass accommodation coefficient can be improved or used at short timescales (< 1min)?

The F-S approach with α_{eff} underestimates partitioning at short timescales because the particle phase does not reach a quasi-steady state and corresponding bulk concentration gradient, whereas the application of α_{eff} is based on the assumption of an effective penetration depth of $r_p/5$ (Eq. 6). This is an inherent limitation for both the F-S approximation and the α_{eff} approach which are assuming a quasi-steady state. The time to reach a quasi-steady state depends on bulk diffusivity, particle radius, and particle-phase reaction rate coefficient (e.g., Fig. 5 in Zaveri et al., 2014). At shorter timescales, we recommend the use of kinetic multilayer models or similarly detailed modeling approaches that can resolve transient conditions. We clarify this point in the revised manuscript.

Line 253, "Model results for SVOC partitioning plus reactive uptake with different rate coefficients in semisolid aerosol particles are shown in Figure 2b." Can the authors C2 elaborate how to choose these first-order bulk reaction rate coefficients (kb = 0, 10-4, 10-3, 0.01, 0.1 s-1)?

The k_b value would vary for different compounds. A study has shown that chemical half-lives of highly oxygenated organic molecules are shorter than one hour (Krapf et al., 2016), corresponding to $k_b > \sim 2\times 10^{-4}$. First-order decomposition rate coefficients for organic hydroperoxides in SOA were reported in the range of $10^{-6} - 1.5\times 10^{-3}$ (Tong et al., 2016; Tong et al., 2018; Wei et al., 2020) and can be enhanced by photolysis (Badali et al., 2015; Epstein et al., 2014) or Fenton-like reactions in the presence of transition metal ions (Goldstein and Meyerstein, 1999). We add this aspect in the revised manuscript.

Line 263, "Figures 3a and 3b show how the effective mass accommodation coefficient aeff depends on volatility and bulk diffusivity as related to particle phase state and viscosity according to the Stokes-Einstein relation (Shiraiwa et al., 2011)." What are the timescale used in these simulations (e.g. < 1min, 1min to 1hr or > 1hr)? Would the simulated results affect by the timescale selected for the simulations (e.g. < 1min vs. > 1hr)?
Line 263, For Figure 4, what are the timescale used in these simulations (e.g. < 1min, 1min to 1hr or > 1hr)? Would the simulated results affect by the timescale chosen for the simulations (e.g. < 1min vs. > 1hr)?

The results presented in Figures 3 and 4 were not obtained by numerical simulations, but were calculated with the analytical equations Eq. 5 & 6 under the assumption of quasi-steady-state conditions. We clarify this point in the revised manuscript.

Summary and conclusions. In this section, can the authors discuss how an effective mass accommodation coefficient can be extended to apply for aerosols containing both inorganic and

organic species? How the phase separation and morphology can be accounted in the effective mass accommodation coefficient?

The penetration depth and related formulations presented in this study assume that organic particles (which can be mixed with inorganic components) are homogenous without considering potential gradients of bulk diffusivity. As pointed out, mixed organic-inorganic particles often undergo liquid-liquid phase separation. Additional work is necessary to develop advanced formulations for mass transfer of gas-phase species to particles with complex morphology. This aspect goes beyond the scope of this current study and may require further studies as stated in the last sentence of the manuscript:

"mixed organic-inorganic particles often undergo liquid-liquid phase separation at moderate and high RH (Krieger et al., 2012; You et al., 2014; Zuend and Seinfeld, 2012), and liquid-liquid phase separation can also occur for purely organic particles (Song et al., 2017). The interplay of particle phase state and phase separation can further impact SOA partitioning (Shiraiwa et al., 2013b). In such complex particle morphologies with multiple phases, gradients and discontinuities of diffusivity may occur within the particle bulk and require more advanced modeling approaches of gas-particle interaction kinetics to be addressed in future studies."