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Atmospheric Chemistry and Physics Discussions

Interactive comment on “Modeling the effect of non-ideality, dynamic mass transfer and viscosity on SOA formation in a 3-D air quality model” *by* Youngseob Kim et al.

Anonymous Referee #1

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Kim et al. investigated the effects of non-ideality, dynamic mass transfer and aerosol viscosity on SOA formation using an air quality model. Most of the current 3-D air quality models have not explicitly considered the role of kinetic partitioning and the interactions between organic and inorganic aerosol components. The organic aerosol community could certainly benefit from this modeling study. However, it is recommended that the manuscript be significantly improved before being published in ACP.

Major comments: (1) My major concern is related to the assumptions of particle viscosity. In the “Dynamic viscous” scenario, the particle diffusivity is assigned as a constant value, as low as 10-30 m2 s-1. I wonder is it necessary to assign such a small value

that almost stopes the bulk diffusion, which would rarely happen in the ambient air. In addition, particle viscosity should be a function of temperature, RH, and particle composition. However, in the sensitivity simulations in Section 4.5, the particle diffusivity is kept constant. Thus, it is hard to convince the readers that the results presented in Section 4.5 could reflect the impact of particle viscosity on SOA formation / evaporation in real atmospheric conditions.

(2) Although the development of the SOAP module has been published in Couovidat and Sartelet (2015), some treatments should be clarified when coupling SOAP into a 3-D model. Firstly, in Couovidat and Sartelet (2015), ISORROPIA is called prior to SOAP and the amount of particle water will adopt the value computed by ISORROPIA instead of SOAP when the water in SOAP is lower than the water in ISORROPIA. This treatment will cause uncertainties in particle water calculations as stated in Couovidat and Sartelet (2015). Has ISORROPIA been fully coupled with SOAP in the current 3-D modeling? If not, what is its impact on the results of SOA partitioning? I suggest adding a spatial distribution of particle water in Figure 3. Secondly, in Couovidat and Sartelet (2015), when treating the diffusion of organic compounds in spherical organic particles, it is assumed that the concentrations in one layer can be described independently from the concentrations in the other layers and the mass fraction (ratio of the mass of the layer over the mass of the particle) of layers must stay constant throughout the simulation. It seems the authors followed Couovidat and Sartelet (2015) and assumed that the transfer between the interface and the internal layer “can happen without diffusion to assure that the mass fraction of layers remain constant even during the growth or shrinking of the particle” (Page 13). This assumption may explain the unreasonable blue arrows in Figure 4 which is also questioned by the other referee. I agree with the related comments proposed by the other referee on Figure 4 and suggest revising this treatment in SOAP as the results in Figure 4 are contrary to previous kinetic simulations (please refer to Liu et al. PNAS, 2016; Mai et al. EST, 2015; Shiraiwa et al. GRL, 2012). For example, in Fig.4a, why compound A in the particle phase can not transfer from the core to the interface while compound A in the gas phase can transfer from

the interface to the core? Is it just because the mass ratios of the layers have to be maintained constant as the authors stated in the last paragraph in Section 3.3?

Thirdly, what is the time step adopted in the 3-D model? This is related whether the gas-particle partitioning has been reached equilibrium within the time step set in the 3-D model. Fourthly, the accommodation coefficient α is assumed as 0.5 in this study. Will the simulation results be changed if α is adopted as 1 (see the recent experiment study by Krechmer et al. 2017). Lastly, I agree with the comments proposed by the other referee that the authors stated that “the aerosols are assumed internally mixed” (Section 4.1) but the absorbing phases are still calculated separately (caq and cp stated in the last paragraph in Section 4.1).

(3) I have some concern about the presentation quality. For example: the title of Section 2.1 is “Composition of aerosols” but Section 2.1 is about gas-particle partitioning. In Section 2.1, the authors firstly stated that in SOAP the cp included cwater,p, while in the following Eq(2) cwater,p is calculated from cp. I am confused in Eq(2) cp includes cwater,p or not. Figure 2: the scenario of “SOAP-basic” is missing although it is compared with “SOAP-no_water” in the text (Page 8). Figure 2 shows the diurnal variations, but the text lacks the description why the differences of simulated SOA concentrations are sometimes large in daytime while sometimes are large in nighttime. Page 10 Section 3.1, the authors stated that the inorganic compounds influence activity coefficients “by middle-range and long-range interactions” but as already stated on Page 8, inorganic compounds also influence the short-range activity coefficient. Last sentence on Page 11, change “viscous compounds” to “viscous particles”. Last sentence on Page 13, “the compound A condenses onto the new layer created by the compound B” is not true. In my understanding, the layers in the model are kept same during SOA formation / evaporation. Section 4.2, besedes MFE and MFB, RMSE should also be defined. OM1 etc in Table 4 should also be defined. Section 4.3, it is better to describe Fig 6 from Fig 6a to Fig 6d instead of beginning from Fig 6d. The first sentence in Section 4.4, “influence” should be “influenced”.

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