

Comments from the three reviewers are in blue, and our replies are in black. Changes to the manuscript are highlighted in red.

This manuscript presents compares gas phase diffusivities inferred from experiments (compilation of values reported in the literature) with gas phase diffusivities calculated using the method by Fuller et al. Based on the experimental values, preferred values are given and uncertainties are obtained by comparison within experimental data and with the calculated values. The authors use a kinetic multi-layer model (presented elsewhere) to give an example of gas phase diffusion on condensation of two different organic molecules chosen because of their atmospheric relevance and different volatility.

I find the topic and presented results very interesting. I recommend that the manuscript is published after some revisions. I find that the manuscript could be improved in terms of notation and explanations. In addition I find that the section where the multilayer model is used should be improved. My concerns and suggestions are outlined below and I hope the authors will find them useful in improving their manuscript.

Reply: We would like to thank referee 3 for recommending our manuscript for final publication. We have revised our manuscript according to his/her very helpful comments.

Major

I think something is missing in Equation (3b) – otherwise in the examples in Section 6 $C_{g,diff}$ would be ~ 0.5 in all cases? Should it be $1/(1+\gamma/\tau_{diff})$?

Reply: Referee 3 is right. We have corrected it.

The notation and the use of units is not quite consistent. In equation 1 the concentration of X in the gas phase is denoted $[X]_g$ and the unit is molecule cm^{-3} . In Figure 3 gas phase concentration of compound I is denoted $C_{g,VOC}$ and is presented in mass concentration units. I suggest to

include an equation similar to equation 1 but with mass units to make it easier for the reader to go from one notation and set of units to another.

Reply: We agree that the use of units is not very consistent. Using mass concentration in Eq. (1) will lead to the introduction of a new parameter (molar mass) and may reduce its readability. Instead, in the revised manuscript we have explained in the second paragraph of section 5 (P14, L25-29) why we use mass concentration.

It should be explained how [SS] can be calculated, or at least some references to where this is explained should be given.

Reply: We have added a sentence after Eq. (1) to explain how to calculate [SS] (P4, L15-18).

Page 5465 line 19: the effective uptake coefficient is introduced. I assume this value is replacing gamma in Equation 1 ?, but this should be stated explicitly.

Reply: The referee is correct. We have modified Eq. (1) to make it more explicit.

Equation (4): I suggest to explicitly write $\tau_{diff}(dp)$ to emphasize that the gas transport coefficient depends on particle size.

Reply: We understand ref. 3's concern, but would like to keep Eq. (4-5) separated, because combining them together may reduce the readability. In the revised manuscript we have added one sentence to emphasize the importance of particle size (P5, L16-18).

The example using the multilayer model: In general this is a very short section, but very interesting. To me it seems a bit as an "add-on" as it is now. I think this section could be better explained and several parameters should be varied (e.g. particle size, initial concentration of VOC). Estimation of volatilities of low-volatile organic molecules is highly uncertain, this should be acknowledged in the text.

Reply: Based on Ref. 3's comments, we have conducted several sensitivity studies by varying initial concentration of VOC and particle size. The initial concentration of VOC had little influence on effects of gas-phase diffusion (i.e. $C_{g,diff}$, gas-phase diffusion correction factor). The particle size has major influence on $C_{g,diff}$: the larger the particle size, the larger the effects of gas-phase diffusion. We have added the following sentence (P15, L9-11): “Additional model simulations with different seed particle size revealed that larger particle size leads to lower $C_{g,diff}$ value.” In addition, we clearly state the high uncertainty of the volatility of low-volatile compounds in the revised manuscript (P14, l20-21): “Volatility of $C_{20}H_{32}O_{12}$ is highly uncertain and it is assumed to be $1 \times 10^{-3} \mu\text{g m}^{-3}$.”

How is the “near surface gas phase” defined?

Reply: The distance of the near-surface gas-phase from the surface is comparable to the mean free path. We have clarified it in the revised manuscript (P4, L23-24).

In the example the authors have chosen a particle diameter of 300 nm. According to figure 2 this gives a Knudsen number of ~0.6 and using Equation 4 this corresponds to a gas transport coefficient of 1. Is this a special choice? If so, the reader should be made aware of this. Also – as mentioned above it would be relevant to make similar model runs varying one key parameter at a time and discuss the effects.

Reply: 300 nm was an arbitrary choice and we have conducted more simulations with different particle size. Please see the response above.

In the conclusion it says “We further suggest that all the compounds have very similar Knudsen numbers” – If I understand correctly this was not done for all compounds but only four organics?

Reply: We have done the calculations for a variety of inorganic and organic compounds which significantly differ in diffusivities. We stated “find” in the original manuscript, and we have

changed it to “suggest” in the revised version. Though we have not done the calculation for all the compounds (practically it is not possible considering the number of different compounds which may exist in the atmosphere), we are confident with our conclusion.

Minor:

Page 5472: “The differences between the measured and estimated diffusivities are typically <10%” – should it not say: The differences between the preferred and estimated diffusivities are typically <10% ?

Reply: Indeed it should be “measured diffusivities”. Every preferred value is based on measurement, but for many species included in our work there are multiple measurements.

Figure 3 caption: I think red and black has been exchanged in the explanation of figures 3.b and d.

Reply: corrected.