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

In this manuscript, the authors developed a database of diffusion coefficients for atmospheric trace gases. The goal is to use these diffusion coefficients to calculate the rate of gas uptake onto particles. The authors compiled an extensive list of coefficients and made recommendations, and also cited Fuller's method to estimate diffusivities. These diffusion coefficients are then applied to calculate Knudsen numbers and condensation into particles. The paper is clearly written, and the topic is relevant to the scope of Atmospheric Chemistry and Physics. This manuscript should be published after considering these comments:

Reply: We would like to thank ref. 1 for recommending our manuscript for final publication. We have revised the manuscript according to his/her very helpful comments.

- In general, the range in diffusion coefficients is small. It ranges from 30 to around 100 torr cm2 s-1, within 1 order of magnitude of each other. It seems to be that the difference between MVK and C20H32O12 is a result of its volatility, not diffusion coefficient. Why is it then an important task to compile gas-phase diffusion coefficients to correctly model condensation flux? It would seem to be that understanding volatility (which has an error of ~2 orders of magnitude) is more important, and we can assume an average gas phase diffusivity of ~0.1 cm2 s-1. Perhaps the authors can choose a better example?

Reply: We agree with the Ref. 1 that understanding the volatility is more important for predicting the formation of SOA. However, it does not mean that taking into account the effect of gas phase diffusion accurately is not important, especially considering that diffusion coefficients were arbitrarily used in some previous studies. Our work can definitely contribute to reduce the error in this aspect. In the revised manuscript (P5, L16-18), we have added one sentence to further explain the role of gas phase diffusion in heterogeneous reactions: "The effect of gas phase diffusion largely depends on the particle size and the uptake coefficient, as discussed in our previous work (Tang et al., 2014a)".

We have chosen MVK and C20H32O12 in our simulations because their uptake coefficients differ a lot due to their very different volatilities. As a result, the role of gas phase diffusion varies significantly for the two compounds, illustrating very well when gas phase diffusion can be important. In the revised manuscript, we have added one paragraph at the end of Section 6 (P15, L15-24) to explain/interpret our simulation results, and to discuss the role of gas phase diffusion on unreactive/reactive uptake and the formation of SOA.

- If gas phase diffusion becomes the limiting step for these ELVOCs to condense, can the authors comment on the relative importance of heterogeneous reactions of smaller molecules that lead to SOA formation (e.g. glyoxal dissolution and subsequent reactions) and larger molecules such as ELVOCs condensing onto particles? It would seem to me that heterogeneous reactions of smaller, more diffusive compounds can potentially be very important.

Reply: It is a very good point. We have added one paragraph (P15, L15-24) to discuss the heterogeneous uptake of smaller molecules due to reactions in the particle phase.

- In general, it would be nice to perform some sensitivity analysis. For example, the uncertainty in measurements could be used to investigate errors in the condensational flux. Also, the uncertainty in using Fuller's estimation (<10%) can also be investigated. My guess is that these uncertainties are quite small and have little overall effect on estimated condensational flux.

Reply: The simulations we did is to illustrate the role of gas phase diffusion in SOA formation. It has not been designed to simulate the formation of SOA in a realistic manner. However, we agree with Ref. 1 that it is helpful to investigate the error of uptake coefficients caused by uncertainties of estimated diffusivities using Fuller's method. We discussed the uncertainties of the derived uptake coefficients at the end of Section 5 (P13, L26-30). Indeed as Ref. 1 pointed out, the overall effect is quite small.

- Tables 1 and 2 list diffusivities of common gases. It would be useful to tabulate estimated diffusivities of common oxidation products, such as glyoxal, pinonic acid, IEPOX, which would be helpful for the SOA community. Fig. 2 lists some common oxidation products, but it would be nice to see the diffusion coefficients that went into the calculations listed in a table.

Reply: In the revised version (P11, L22-27) we have listed the diffusivities of common oxidation products which are important intermediates in SOA formation.

The purpose of Figure 2 is to show that Knudsen numbers are similar for inorganic and organic compounds which have very different diffusivities. We only include a few compounds in Figure 2 to ensure its readability.

Minor comments: - I suggest rearranging Tables 1 and 2 so that the compounds are listed in increasing molecular weights or carbon numbers from top to bottom, not left to right.

Reply: We respect Ref. 1's comments. However, we feel the way we arrange these tables assures better readability.

Pg. 5475 Equation (12): Particle diameter is usually in nm or um, so it may be more convenient to adjust the units in Dnorm (e.g. 150 torr um)

Reply: We agree with referee 1. In the revised manuscript, we have replaced D_{norm} with a new parameter, λ_{P}, which is equal to 100 nm atm^{-1}. It means that the mean free path of all gas molecules in 1 atm air is around 100 nm. Please see the revised manuscript for further details (P13, L17-L30).

- Pg. 5464 line 24: “condendation” should be “condensation”

- Pg. 5467 line 13: insert “in” into “: : :used our previous work: : :”

- Pg. 5469 line 9: “differenciate” should be “differentiate”

- Pg. 5472 line 8: “multifuntional" should be “multifunctional”

- Pg. 5472 line 12: “tropshere” should be “troposphere”

- Pg. 5473 line 13: “tempereatures” should be “temperatures”

- Pg. 5474 line 11: “sqaure" should be “square”

- Pg. 5476 line 4: “assumed to consists” should be “assumed to consist”

- Pg. 5477 line 5: “alcoholes” should be “alcohols”

- Pg. 5486 Table 2 caption: “multifuntional" should be “multifunctional”

- Pg. 5488 Table A1 caption: “fours” should be “four”

- Pg. 5492 Figure 3: The units on the y-axis label are not shown correctly. I am guessing that they should be in ug m-3.

Reply: Thanks. All typos have been corrected.

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

The manuscript compiles and discusses diffusion coefficients for organic trace gases, important in atmospheric chemistry. The compiled experimental data are compared to an estimation method by Fuller (in Reid et al. 1987). Based on both, preferred values are suggested. Details of the assessment are documented in IUPAC style in an extensive supplement. This all is excellent work and should be published in ACP. (I did not check the supplement for errors and typos.)

Reply: We would like to thank Ref. 2 for recommending our manuscript for final publication and very valuable comments. We have carefully addressed his/her comments in the revised manuscript.

The authors may to consider two suggestions before final publication:

The first regards the representation of the deviation between preferred (experimental) values and the Fuller estimation. In my opinion deviation between preferred and estimated values point to a systematic problem, not to a statistical one. In this case I suggest to give the deviation with the suited sign (to indicate if Fuller over- or underestimate D), and not with +/-.

Reply: We fully understand the concern of Ref. 2. In fact, the deviations between the measured and estimated diffusivities are presented for each measurement in the supplementary documents, in which we also discuss how we assign uncertainties to the recommended values in details. The deviation is defined as (D_e/D_m-1) in %, where D_e and D_m are the estimated and measured diffusivities. Therefore, we would like to keep our current method to assign the uncertainties. We also refer to the supplementary information for further details, and have added a sentence in the second paragraph of Section 4 after we mention the supplement for the first time.

The second point regards the notation “Knudsen number of molecules”. I think this is misleading. The Knudsen number is a scaling factor for a certain problem and relates a particle radius to a free mean path e.g. of a condensing vapor. Kn determines dynamic regime of uptake for a given particle size. Kn “tests” if diffusion to a large particle (quasi motionless) or molecular kinetics rules by rationing the free mean free path (lambda) of the vapor to particle diameter. What the authors find invariant is lambda of the molecules. If I pluck in some numbers in Eq. 12 and consider the definition of Kn as lambda / Rp (Particle radius) then at 760 torr the lambda for many molecules including small inorganic is about 100nm, a typical value.

Therefore de facto mean free path is invariant not the Knudsen number. Of course I am convinced the authors know all this, but I am warning because the semantics is misleading, although in praxis this has no effect.

Reply: We fully agree with referee 2’s comments on the notation and his/her insights into the Knudsen number and the mean free path.

In the revised manuscript, we have made the following changes: i) we have changed “Knudsen numbers of a trace gas” to “the Knudsen numbers for the uptake of a trace gas by particles”; ii) We have updated Eqs. (5) and (11) to include the mean free path, introduced λ_P (pressure normalized mean free path) to replace D_{norm} in Eq. (12) (P13, L18-30), and added a new figure to show that λ_P is constant for all gas molecules; iii) the title has been changed to “…Volume 2. Diffusivities of organic compounds, pressure normalized molecular mean free paths, and average Knudsen numbers for gas uptake calculations”, and the abstract has also been updated accordingly.

Minor comments:

page 5471, line 1-2: I suggest to mention one more time that "estimated refers to Fullers method. Something like: ": : :the difference between preferred and diffusivity estimated by Fuller's method, then: : :"

Reply: Done.

page 5471, line 8: it would be helpful to give the value estimated by Fuller here for better comparison.

Reply: Done.

page5473, line 1: Is the estimated diffusivity for larger carboxylic acids larger or smaller than the "measured". Could internal hydrogen bonding lead to "unusal" molecular morphologies/folding or similar?

Reply: Estimated diffusivities are smaller than measured values for some acids and larger than measured values for other acids. In the revised manuscript we have made it clear for each acids.

page 5474, line 9: I suggest to state that the mean free path, which is approximated by Dp(X)/c(X), is relatively invariant and as a consequence Kn is similar for many vapors for a given particle size.

Reply: It is a very good comment. As we mention above, in the revised manuscript we have introduced a new parameter, the pressure normalized mean free path, to replace D_{norm}.

Typos

page 5467, line 14: use plural, "inorganic compounds" page 5471, line 8: singular, diffusivity page 5474, line 11: square

Reply: corrected.

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 Cg,diff would be ~0.5 in all cases? Should it be 1/(1+gamma/Taudiff)?

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 Cg,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 1×10^{-3} μg 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.