

Response to referee #1

In this work, Li et al. proposed “equivalent uptake coefficient”, used this term to compare the relative importance of gas uptake onto aerosol surface versus group surface, and concluded that some uptake processes onto aerosol particles can be very important. The methodology is novel, and the results can be interesting for the atmospheric chemistry community. The manuscript can be accepted after the following comments are addressed.

Response: We thank the positive and constructive comments given by the referee #1, which are very helpful to improve the manuscript. Our response to each specific comment is presented below.

Detailed Comments and Responses:

1. P41, Table 1: There are some experimental studies (by Joel A Thornton, Jon Abbatt, Tim Bertram, and likely other) which explored the effect of organics on N_2O_5 uptake. In addition, there may be more studies on H_2O_2 uptake. Please check the IUPAC evaluation online as well as relevant literature.

Response: We thank the referee's comments. In the last version, we have already included the following experimental studies suggested by the referee in Table A.3 regarding N_2O_5 uptake on organics, i.e., Thornton et al. (2003), Griffiths et al. (2009), and Badger et al. (2006). In the revised manuscript, we have tried to complete the list by including Folkers et al. (2003), Gross et al. (2009) and Anttila et al. (2006) as in the new Table A.3.

For H_2O_2 , we checked the IUPAC evaluation online data and related literature. We now add one more measurement of H_2O_2 uptake on mineral dust (Zhao et al., 2011) in the revised manuscript. We are still unable to find more laboratory measurements of H_2O_2 uptake on aerosols other than mineral dust, thus more measurements are needed in the future.

2. P42, Table 2: I am not sure why the work by Wang et al. (2012) is used a representative example here. In fact, the uptake coefficients used by Wang et al. are far from being updated, and they mainly used uptake coefficients adopted by two modeling studies almost 20 years ago (Dentener et al., 1996; Zhang and Carmichael, 1999). For mineral dust in specific, the uptake coefficients used by Zhu et al. 2010 were updated values recommended by IUPAC. In addition, some of the studies which are cited as the sources of uptake coefficients measured by laboratory work are in fact pure modeling work, such as Bauer et al. (2004), Dentener et al. (1996), and so on. The author may consider updating this table.

Response: We thank the referee's comments. The scheme of Wang K et al. (2012) is taken as an example considering the large impact/applications of this scheme within the community (e.g., Wang et al., 2014; Li et al., 2015; Zheng et al., 2015). We update the table in the revised manuscript by including the scheme of Zhu et al. (2010) which uses updated values recommended by IUPAC. It should be addressed that we only provide examples of model

schemes that have considered the heterogeneous reactions to give an overall implication for modelers, rather than to give a complete overview of the parameterizations of uptake coefficients covering all modeling studies, which is out of the scope of this paper. To avoid misunderstanding, we revise the caption to “Examples of aerosol uptake coefficients used in atmospheric models”, add more illustrations in the footnote and main text, and move Table 2 to the supplement.

Uptake coefficients with sources listed as Dentener et al. (1996), Bauer et al. (2004), Song and Carmichael (2001) in the table are from model parameterization, and the specific laboratory measurements are not found in the literature. We have already noted this issue in the last version (see footnote ^b).

3. P3, L23-25: HO₂ uptake can be very important for tropospheric chemistry (George et al., 2013; Mao et al., 2013; Taketani et al., 2008; Thornton et al., 2008). Is there a reason why HO₂ has not been discussed in this paper?

Response: We agree that HO₂ uptake on aerosol is important for atmospheric chemistry. The motivation of our work is to compare the fluxes of dry deposition and aerosol uptake, which is difficult for radicals like HO₂ because the required parameters to calculate dry deposition are not available from current literature. The reason could be that the other pathways are too fast compared to the deposition of HO₂.

4. P2, L29-30: Very recently I reviewed heterogeneous reactions of mineral dust (Tang et al., 2017). Should this paper be cited here?

Response: We have included this review paper in the revised manuscript along with other original research articles.

5. P3, L12-15: Another convenient way to assess the relative importance of aerosol uptake and dry deposition is to calculate their lifetimes with respect to individual processes, as discussed by Tang et al. (2017).

Response: We agree with the referee that comparison of lifetimes between aerosol uptake and dry deposition is another feasible method, as discussed in Tang et al. (2017). The method proposed in our study share the same basic formulations with Tang et al. (2017), and velocities can be easily converted to lifetime.

6. P4, L24-26: This sentence is not easy to follow. I assume that the authors wanted to state that for smaller particles, gas phase diffusion would not be a limiting step and thus can be neglected. Please consider rephrasing it, and refer to Tang et al. (2014) for a comprehensive discussion on the role of gas phase diffusion.

Response: We thank the referee's comments. We have referred to the original formation of Jacob 2000 as follows:

“For atmospheric aerosols with a diameter of $\sim 0.2 \mu\text{m}$ or smaller, the related gaseous uptake tends to be limited by the free molecular collision rate (uptake rate $\rightarrow \omega\alpha A[X_g]/4$) (Jacob, 2000). Thus in the following analyses, we mainly focus on the discussion of γ_{eff} , and neglect the diffusion resistance in the gas phase.”

7. P12-13: In a paper published in 2017 (Tang et al., 2017), I provided a comprehensive and in depth discussion on the two factors the authors mentioned in Section 4.2, and would like to refer the authors to take a look at that paper.

Response: Thanks for the referee’s comments. We refer to this work in our discussion as follows:

“More than three orders of magnitudes of differences are derived by whether to consider the pores within the microstructure of solid aerosol surface or not (see Table A.1). Using the same method to calculate the available surface area may reconcile these differences (Tang et al., 2017).”

8. P14, L19-21 as well as related content in the abstract It is proposed that the following four groups of gas uptake onto aerosols can be important: 1) N_2O_5 on all types of aerosols, 2) HNO_3 and H_2O_2 on mineral dust, 3) O_3 on liquid organic aerosols; and 4) NO_2 , SO_2 , HNO_3 on sea salt aerosols. The four groups have some overlaps and not easy to follow. I would suggest re organizing them according to either types of gases or types of aerosol s

Response: This is re-organized to present the most intensive summary of our conclusion.

We will add the following table to better illustrate it. According to Table 2, there is no overlapped information and four bullets are already the minimum number of vectors from this matrix.

Table 2. Gas uptake processes that are potentially important compared to dry deposition across various environments (marked with ✓).

Gases	Mineral dust	Soot	Organic aerosol-solid	Organic aerosol-liquid	Sea salt aerosol
O_3				✓	
NO_2					✓
SO_2	✓				✓
N_2O_5	✓	✓	✓		✓
HNO_3	✓				✓
H_2O_2	✓				

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