Response to referee #2

The manuscript "Relative importance of gas uptake on aerosol and ground surfaces characterized by equivalent uptake coefficients" presented a theoretical approach to characterize the relative importance of uptake of trace gases on aerosols versus on ground. The authors proposed a new parameter "equivalent uptake coefficient" (γ_{eqv}) at which the flux of gas uptake on aerosols is equal to that on ground and derived γ_{eqv} under various environment (vertical velocity and particle surface concentration). By comparing γ_{eqv} with the effective uptake coefficient of gases on aerosols (γ_{eff}) reviewed from literature, the authors assessed the relative importance of gas uptake on aerosols to dry deposition. It was found that under urban environment, gas uptake on all types of aerosols (mineral dust, sea salt, organic aerosol, and soot) is important, while in pristine Amazonia forest the contribution of uptake on aerosols to gas loss is minor. N_2O_5 uptake on all types aerosol, HNO_3 and H_2O_2 on mineral aerosols, O_3 on liquid organic aerosol, NO_2, SO_2 and HNO_3 on sea salt aerosol are as important as dry deposition. The author also pointed out that H_2O_2 uptake on various aerosols need further laboratory studies and to be evaluated. The approach presented is a novel and convenient way to compare the relative importance of uptake of gases on aerosols with dry deposit. This manuscript is well written and easy to follow. And the discussion is well balanced. I have only a few minor comments, mainly to clarify some discussion. I recommend the direct publication of this manuscript on ACP after these minor comments are fixed.

Response: We thank the positive and constructive comments given by the referee #2, which are very helpful to improve the manuscript. Our response to each specific comment is presented below.

1. Pg. 4 line 18, a typical value of ω of 300 m s^{-1} is used. I understand this can simplify the equation and γ_{eqv}, since different gases have slightly different mean velocity, especially in order to get a clear picture as shown in Fig. 2. Are the γ_{eqv} values in Fig. 3-5 also calculated in this way? It might be helpful to briefly mention the influence of this simplification in the discussion part "Sect. 4.3".

Response: We thank the referee's comments. We applied the same formula in Fig. 2 and Fig. 3-5, i.e., the typical mean thermal velocity of 300 m s^{-1} was also used for Fig. 3-5. The biases due to this simplification are within 20% for calculations of γ_{eqv} for O_3, NO_2, SO_2 and HNO_3, and within 30% for H_2O_2 and N_2O_5. We add more discussion on this simplification in the revised manuscript as follows:

"We use a unified thermal velocity (300 m s^{-1}) for all gases, which will introduce positive biases of +4% ~ +30% for O_3, NO_2, SO_2, HNO_3, H_2O_2, and a negative bias of -24% for N_2O_5 in calculations of γ_{eqv} at the same temperature"

2. Pg. 10 line 11, I am curious why the authors mainly discussed the model schemes in the studies Liao and Seinfeld (2005) and Wang K et al. (2012) among other model studies including heterogeneous reactions.

Response: We thank the referee's comments. The scheme of Liao and Seinfeld (2005) and Wang K et al. (2012) were taken as an example here considering the large impact/applications of this scheme within the community (e.g., Monks et al., 2009; 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 (International Union of Pure and Applied Chemistry). It should be addressed that we only provide examples of model schemes here 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 update the table, 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.

3. Pg. 11 line 24, "…Sect. 3.5.1…", I guess that the authors meant "4.1.1". Also check line 26.

Response: Thanks for the careful reading and help. We correct it in the revised manuscript.

4. Pg. 13 line 27-Pg. 14 line 5, it might be helpful to also mention that the variability of aerosol surface concentration under each environment could also contribute to the variability of γ_{eqv}.

Response: We agree with the referee's comments that the variability of aerosol surface concentration can contribute to the variability of γ_{eqv}. We have included the following statement to emphasize it in the revised manuscript:

"In addition, the variability of aerosol surface area under each environment can also contribute to the variability of γ_{eqv}."

5. Pg. 14 line 25, it seems that one leading sentence is missing before "(a)…". Please double check.

Response: We appreciate the referee's careful reading and help. We add a leading sentence before the statements (a)~(c).

"There are several indications from this work of processes that should be addressed in future measurements and model implementations:"

6. Pg. 14 line 20, "…HNO_3 *and* H_2O_2 *on mineral…", according to Fig. 2 should* SO_2 *be also listed here?*

Response: As shown in Fig. 4, there are more than three orders of magnitude of variances in γ_{eff} for SO_2. γ_{eff} of mineral dust falls in the range of γ_{eqv} under high aerosol loadings or high mixing heights. The wide range of γ_{eff} for mineral dust (1.5×10^{-8} to 6.3×10^{-4}) is a big

challenge regarding its application in models. Considering this large variations, we add SO_2 uptake on mineral dust as one of the important processes compared to dry deposition, and further discuss the potential uncertainty of SO_2 in item (c) of the "Conclusion" section.

7. Pg. 38 line 6, "…the purple bar…" should be "blue bar".

Response: Revised.

References

Li, X., Zhang, Q., Zhang, Y., Zheng, B., Wang, K., Chen, Y., Wallington, T. J., Han, W., Shen, W., Zhang, X., and He, K.: Source contributions of urban PM2.5 in the Beijing–Tianjin–Hebei region: Changes between 2006 and 2013 and relative impacts of emissions and meteorology, Atmospheric Environment, 123, 229-239, https://doi.org/10.1016/j.atmosenv.2015.10.048, 2015.

Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R. S., Carslaw, K., Cooper, O. R., Dentener, F., Fowler, D., Fragkou, E., Frost, G. J., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H. C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I. S. A., Jenkin, M. E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M. G., Lee, J. D., Liousse, C., Maione, M., McFiggans, G., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J. J., O'Dowd, C. D., Palmer, P. I., Parrish, D. D., Petzold, A., Platt, U., Pöschl, U., Prévôt, A. S. H., Reeves, C. E., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van der Werf, G. R., Vautard, R., Vestreng, V., Vlachokostas, C., and von Glasow, R.: Atmospheric composition change – global and regional air quality, Atmospheric Environment, 43, 5268-5350, https://doi.org/10.1016/j.atmosenv.2009.08.021, 2009.

Wang, Y., Zhang, Q., Jiang, J., Zhou, W., Wang, B., He, K., Duan, F., Zhang, Q., Philip, S., and Xie, Y.: Enhanced sulfate formation during China's severe winter haze episode in January 2013 missing from current models, Journal of Geophysical Research: Atmospheres, 119, 10,425-410,440, 10.1002/2013JD021426, 2014.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmos. Chem. Phys., 15, 2031-2049, 10.5194/acp-15-2031-2015, 2015.

Zhu, S., Butler, T., Sander, R., Ma, J., and Lawrence, M. G.: Impact of dust on tropospheric chemistry over polluted regions: a case study of the Beijing megacity, Atmos. Chem. Phys., 10, 3855-3873, 10.5194/acp-10-3855-2010, 2010.