

Response to comments of referees

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 N₂O₅ uptake. In addition, there may be more studies on H₂O₂ 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 N₂O₅ 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 H₂O₂, we checked the IUPAC evaluation online data and related literature. We now add one more measurement of H₂O₂ uptake on mineral dust (Zhao et al., 2011) in the revised manuscript. We are still unable to find more laboratory measurements of H₂O₂ 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 \checkmark).

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

References

- Anttila, T., Kiendler-Scharr, A., Tillmann, R., and Mentel, T. F.: On the Reactive Uptake of Gaseous Compounds by Organic-Coated Aqueous Aerosols: Theoretical Analysis and Application to the Heterogeneous Hydrolysis of N₂O₅, *The Journal of Physical Chemistry A*, 110, 10435-10443, 10.1021/jp062403c, 2006.
- Badger, C. L., Griffiths, P. T., George, I., Abbatt, J. P. D., and Cox, R. A.: Reactive Uptake of N₂O₅ by Aerosol Particles Containing Mixtures of Humic Acid and Ammonium Sulfate, *The Journal of Physical Chemistry A*, 110, 6986-6994, 10.1021/jp0562678, 2006.
- Folkers, M., Mentel, T. F., and Wahner, A.: Influence of an organic coating on the reactivity of aqueous aerosols probed by the heterogeneous hydrolysis of N₂O₅, *Geophys. Res. Lett.*, 30, 10.1029/2003GL017168, 2003.
- Griffiths, P. T., Badger, C. L., Cox, R. A., Folkers, M., Henk, H. H., and Mentel, T. F.: Reactive Uptake of N₂O₅ by Aerosols Containing Dicarboxylic Acids. Effect of Particle Phase, Composition, and Nitrate Content, *The Journal of Physical Chemistry A*, 113, 5082-5090, 10.1021/jp8096814, 2009.
- Gross, S., Iannone, R., Xiao, S., and Bertram, A. K.: Reactive uptake studies of NO₃ and N₂O₅ on alkenoic acid, alkanoate, and polyalcohol substrates to probe nighttime aerosol chemistry, *Physical Chemistry Chemical Physics*, 11, 7792-7803, 10.1039/B904741G, 2009.
- 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 PM_{2.5} 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.
- Thornton, J. A., Braban, C. F., and Abbatt, J. P. D.: N₂O₅ hydrolysis on sub-micron organic aerosols: the effect of relative humidity, particle phase, and particle size, *Physical Chemistry Chemical Physics*, 5, 4593-4603, 10.1039/B307498F, 2003.
- Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, *Atmos. Chem. Phys.*, 12, 10209-10237, 10.5194/acp-12-10209-2012, 2012.
- 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, 4104-4109, 10.1002/2013JD021426, 2014.

Zhao, Y., Chen, Z., Shen, X., and Zhang, X.: Kinetics and Mechanisms of Heterogeneous Reaction of Gaseous Hydrogen Peroxide on Mineral Oxide Particles, *Environmental Science & Technology*, 45, 3317-3324, 10.1021/es104107c, 2011.

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.

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, "...HNO₃ and H₂O₂ on mineral...", according to Fig. 2 should SO₂ be also listed here?

Response: As shown in Fig. 4, there are more than three orders of magnitude of variances in γ_{eff} for SO₂. 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 SO₂ uptake on mineral dust as one of the important processes compared to dry deposition, and further discuss the potential uncertainty of SO₂ 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 PM_{2.5} 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, 425–440, [10.1002/2013JD021426](https://doi.org/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](https://doi.org/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](https://doi.org/10.5194/acp-10-3855-2010), 2010.

Relative importance of gas uptake on aerosol and ground surfaces characterized by equivalent uptake coefficients

Meng Li^{1,*}, Hang Su^{2,1}, Guo Li¹, Nan Ma², Ulrich Pöschl¹, and Yafang Cheng¹

¹ Max Planck Institute for Chemistry, Mainz, 55118, Germany

5 ² Center for Air Pollution and Climate Change Research (APCC), Institute for Environmental and Climate Research (ECI), Jinan University, Guangzhou, 511443, China

*now at: Chemical Science Division, Earth System Research Laboratory, National Oceanic and Atmospheric Administration (NOAA), Boulder, Colorado 80305, United States

10 Correspondence to: Hang Su (h.su@mpic.de) and Yafang Cheng (yafang.cheng@mpic.de)

Abstract. Quantifying the relative importance of gas uptake on the ground and aerosol surfaces helps to determine which processes should be included in atmospheric chemistry models. Gas uptake by aerosols is often characterized by an effective uptake coefficient (γ_{eff}), whereas gas uptake on the ground is usually described by a deposition velocity (V_d). For efficient comparison, we introduce an equivalent uptake coefficient (γ_{eqv}) at which the uptake flux of aerosols would equal that on the ground surface. If γ_{eff} is similar to or larger than γ_{eqv} , aerosol uptake is important and should be included in atmospheric models. In this study, we compare uptake fluxes in the planetary boundary layer (PBL) for different reactive trace gases (O_3 , NO_2 , SO_2 , N_2O_5 , HNO_3 , H_2O_2), aerosol types (mineral dust, soot, organic aerosol, sea salt aerosol), environments (urban, agricultural land, Amazon forest, water body), seasons, and mixing heights.

For all investigated gases, γ_{eqv} ranges from $10^{-6} \sim 10^{-4}$ in polluted urban environments to $10^{-4} \sim 10^{-1}$ under pristine forest conditions. In urban areas, aerosol uptake is relevant for all species ($\gamma_{\text{eff}} \geq \gamma_{\text{eqv}}$) and should be considered in models. On the contrary, contributions of aerosol uptakes in Amazon forest are minor compared to the dry deposition. Phase state of aerosols could be one of the crucial factors influencing the uptake rates. Current models tend to underestimate the O_3 uptake on liquid organic aerosols which can be important especially over regions with $\gamma_{\text{eff}} \geq \gamma_{\text{eqv}}$. H_2O_2 uptakes on a variety of aerosols is yet to be measured at laboratory and evaluated.

25 Given the fact that most models have considered their uptakes on the ground surface, we suggest also considering the N_2O_5 uptake by all types of aerosols, HNO_3 and SO_2 uptake by mineral dust and sea salt aerosols, H_2O_2 uptake by mineral dust, NO_2 uptakes by sea salt aerosols and O_3 uptake by liquid organic aerosols in atmospheric models.

1 Introduction

Multiphase processes play an essential role in atmospheric chemistry and atmosphere-biosphere exchange (Ravishankara, 1997; Ammann et al., 1998; Gard et al., 1998; Usher et al., 2003; Bauer et al., 2004; Fowler et al., 2009; Kolb et al., 2010; Su et al., 2011, 2013; Herrmann, 2003, 2015; Pöhlker et al., 2012; Ammann et al., 2013; Oswald et al. 2013; Andreae et al., 2015; George et al., 2015; McNeill, 2015; Pöschl and Shiraiwa, 2015; Quinn et al., 2015; Weber et al., 2015; Cheng et al., 2016; Froehlich-Nowoisky et al. 2016; Lappalainen et al. 2016; Tang et al., 2016; Meusel et al. 2018). It not only affects the atmospheric trace gases concentrations but also modifies the properties of condensed phases, commonly known as the aging process (Song and Carmichael, 1999; Cheng et al., 2006, 2012; Rudich et al., 2007; Andreae 2009; Jimenez, et al., 2009; Gunthe et al., 2011; Ditas et al., 2018). In the planetary boundary layer, aerosols and ground provide two kinds of surfaces for multiphase reactions. In previous gas uptake studies, different formulations have been used to describe and parameterize the gas uptake processes (Wesely, 1989; Ravishankara, 1997; Jacob, 2000; Wesely and Hicks, 2000; Zhang et al., 2003; Ammann and Pöschl, 2007; Pöschl et al., 2007; Wesely, 2007).

A variety of ground surfaces, including vegetation, water, rock, road etc., can take up gaseous species through dry deposition, thus having significant impacts on the budget of these reactive gases and on the physicochemical properties of the ground surface itself (Lelieveld and Dentener, 2000; Ashmore, 2005). Dry deposition is one of the major removal pathways for most gaseous species and has been extensively parameterized in atmospheric models (Wesely and Hicks, 2000; Zhang et al., 2002, 2003). A resistance model, which consists of the aerodynamic resistance, quasi-laminar resistance and surface resistance, has been widely applied to calculate the dry deposition flux in global and regional atmospheric models (see Fig. 1, Wesely and Hicks, 2000; Wesely, 2007). The dry deposition velocity, V_d (in unit of cm s^{-1}) calculated as the reciprocal of the total resistance, is the key parameter to describe the uptake fluxes on the ground.

From late 1990s, the importance of reactive uptake of gases by aerosols has been commonly accepted (Ravishankara, 1997; Gard et al., 1998; Jacob, 2000). Gas uptake by aerosols not only influences the fate of reactive gases, but also changes the physio-chemical properties of atmospheric aerosols (Kolb et al., 2010). Taking account of the multiphase chemistry is proven a key factor to explain the observations and improve the model performances (Zhang and Carmichael, 1999; Song and Carmichael, 2001; Liao and Seinfeld, 2005; Wang et al., 2006; McNaughton et al., 2009; Wang X et al., 2012; Zheng B et al., 2015; Tang et al., 2017; Chen et al. 2018; Mu et al., 2018). Compared to dry deposition, the parameterization of gas uptake on aerosols is more challenging (Jacob, 2000; Pöschl and Shiraiwa, 2015). The mass transfer between gases and aerosols can be described by the resistance model in analogy to electrical circuit which decoupled the physio-chemical limitations in the gas phase, gas-surface interface and the bulk phase under (quasi-) steady state conditions (Schwartz and Freiberg, 1981; Schwartz, 1986; Kolb et al., 1995). A simplified scheme, which relies on the formulation of effective uptake coefficient (γ_{eff}) has been widely used in current atmospheric models (Jacob, 2000; Liao and Seinfeld, 2005; Wang K et al., 2012). Growing numbers of laboratory studies have reported γ_{eff} for various trace gases and aerosol particles that are

potentially important for atmospheric chemistry, such as O_3 , NO_2 , SO_2 , N_2O_5 , HNO_3 on mineral dust (Ullerstam et al., 2002; Mogili et al., 2006; Vlasenko et al., 2006; Wagner et al., 2008; Ndour et al., 2009), soot (Rogaski et al., 1997; Longfellow et al., 2000; Al-Abadleh and Grassian, 2000; Saathoff et al., 2001), and sea salt aerosols (Mochida et al., 2000; Gebel and Finlayson-Pitts, 2000; Hoffman et al., 2003; Thornton and Abbatt, 2005; Ye et al., 2010). A series of evaluations on the kinetic and photochemical data for the multiphase reactions were conducted afterwards (Crowley et al., 2010, 2013; Ammann et al., 2013; Burkholder et al., 2015). Pöschl et al. (2007) and the follow-up studies (e.g., Shiraiwa et al., 2010, 2011) developed a comprehensive kinetic model framework, enabling consistent and unambiguous descriptions of mass transfer and chemical reactions in aerosol systems.

However, which kind of surfaces is more important for gas uptake in the planetary boundary layer (PBL)? The answer is not straightforward because of the following reasons:

(1) though the surface of the Earth seems to be much larger than that of tiny aerosols, its contribution is diluted by the large volume of the PBL, resulting in a surface to volume ratio close to that of aerosol. For example, for a PBL height of 1000 m, the corresponding surface to volume ratio is $1000 \mu m^2 cm^{-3}$, comparable to aerosol surface area concentrations of $200 \sim 2000 \mu m^2 cm^{-3}$ for urban areas (Woo et al., 2001; Stanier et al., 2004; Wu et al., 2008, 2017; Ma and Birmili, 2015), and $200 \sim 1000 \mu m^2 cm^{-3}$ for rural environments (Ma et al., 2014; Ma and Birmili, 2015; Wu et al., 2017; Held et al., 2008).

(2) different formulations also hinder the comparison. As illustrated above, different schemes, formulations and terminologies are applied to calculate the uptake fluxes on ground and aerosols. The dry deposition velocity (V_d) is the fundamental parameter to describe the deposition process on the ground while the effective uptake coefficient (γ_{eff}) is used to describe the uptake fluxes on aerosols.

In this study, we conducted a comparative assessment of the gas uptake on both ground and aerosol surfaces. Our goal is to identify the prevailing multiphase process in the PBL, and especially those processes that have not yet been sufficiently considered in atmospheric models. Section 2 described the methods of calculation and comparison. We presented and discussed the main results in Section 3, which is followed by a summary of our major findings in Section 4.

2 Methods

In this work, we compared the relative importance of gas uptake by the ground and aerosols based on their uptake fluxes. In this comparison, resistance models were applied to calculate uptake fluxes on both ground and aerosol surfaces (see Fig. 1) as detailed below. The uptake fluxes of six reactive gases (O_3 , NO_2 , SO_2 , N_2O_5 , HNO_3 , H_2O_2) were calculated and compared for four typical land use categories (urban, agricultural land, Amazon forest, water) and five aerosol types (mineral dust, soot, solid organic aerosol, liquid organic aerosol, sea salt aerosol). These species were chosen considering their potential importance regarding dry deposition on the ground and uptake on aerosols within the troposphere.

2.1 Ground gas uptake

Dry deposition fluxes were calculated following the scheme and parameters of Wesely (1989) and Zhang et al. (2003). As shown in Fig. 1, the resistance model applied to characterize the dry deposition process includes the aerodynamic resistance (R_a), quasi-laminar resistance (R_b) and surface resistance (R_c). The basic equations for the flux calculations are:

$$5 \quad F_{grd} = -V_d[X_g] \times 10^{-2} \quad (1)$$

$$V_d = \frac{1}{R_{grd}} = \frac{1}{R_a + R_b + R_c} \quad (2)$$

where F_{grd} represents the gas deposition fluxes on various ground surfaces ($\text{mol m}^{-2} \text{s}^{-1}$); V_d represents the deposition velocity (cm s^{-1}); $[X_g]$ is the averaged gas concentration (mol m^{-3}); R_{grd} is the total resistance in the dry deposition process (s cm^{-1}), composed of R_a , R_b and R_c . The detailed equations and parameterization scheme for determination of R_a , R_b and R_c are provided in the supplement. A neutral meteorological condition was assumed in the calculation. We present the key input parameters and the calculated V_d in Table S1 and Table S2, respectively.

2.2 Aerosol gas uptake and the effective uptake coefficient (γ_{eff})

The net flux of gas X from gas phase to the condensed phase (J_{net} , $\text{mol m}^{-2} \text{s}^{-1}$) for one aerosol particle can be expressed as Eq. (3) under (quasi-) steady-state conditions (Pöschl et al., 2007):

$$15 \quad J_{net} = \frac{\omega \gamma_{eff}}{4} [X_g] \quad (3)$$

The effective uptake coefficient, γ_{eff} , represents the number of gas molecules taken by the aerosol particle divided by the number of those impacting onto the particle surface (Pöschl et al., 2007); ω is the mean thermal velocity (m s^{-1}), we use a typical value of 300 m s^{-1} in this study; $[X_g]$ is the averaged gas concentration far away from the aerosol surface (mol m^{-3}).

$$\frac{1}{\gamma_{eff}} = \frac{1}{\Gamma_g} + \frac{1}{\alpha} + \frac{1}{\Gamma_b} \quad (4)$$

20 As shown in Fig. 1, resistance models have been widely applied to quantify the mass transfer of gases to aerosol particles. For gas uptake on liquid droplets, following the resistance model as described by Eq. (4), the overall resistance $1/\gamma_{eff}$ is composed of three resistor terms due to gas diffusion ($1/\Gamma_g$), interfacial mass transfer ($1/\alpha$) and bulk diffusion and reaction ($1/\Gamma_b$) (Pöschl et al., 2007). The conductance of gas diffusion is commonly calculated based on $\Gamma_g = 8D_g\omega^{-1}d_p^{-1}$, where D_g is the diffusion coefficient of gas X in the gas phase ($\text{m}^2 \text{s}^{-1}$), and d_p represents the aerosol particle diameter. **For large particles and very fast uptake processes, the gas diffusion process can be a limiting factor for the overall uptake (Tang et al., 2014). For atmospheric aerosols with a diameter of $\sim 0.2 \mu\text{m}$, 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.**

Given a mixing height of h , and aerosol surface area density of A (particle surface area per unit volume of air, $\mu\text{m}^2 \text{cm}^{-3}$), the total uptake flux of gas X by aerosols (F_{aer} , $\text{mol m}^{-2} \text{s}^{-1}$) is:

$$F_{\text{aer}} = J_{\text{net}} Ah = \frac{\omega \gamma_{\text{eff}}}{4} Ah [X_g] \times 10^{-6} \quad (5)$$

where 10^{-6} is the unit conversion factor. We summarized the measured uptake coefficients for a variety of gas species and aerosol types at both initial state and steady state in Table 1 (details in Table A.1 ~ A.4). They are mainly derived from the measured values in literatures, reviewed data of IUPAC (International Union of Pure and Applied Chemistry) Task Group on Atmospheric Chemical Kinetic Data Evaluation (Crowley et al., 2010, 2013; Ammann et al., 2013; available at <http://iupac.pole-ether.fr/>), and NASA-JPL (Jet Propulsion Laboratory, Burkholder et al., 2015) (see references in Table A.1 ~ A.4). As we focus on PBL, those γ_{eff} measured at room temperatures ($\sim 298\text{K}$) are mainly presented. Gas uptakes at very low temperature (e.g., polar region, stratosphere) are out of scope of this study and should be explored in future work.

Though the initial and steady-state uptake coefficients are listed, it should be noted that the values at initial state may not be appropriate for direct application in chemical transport models (CTMs) considering the subsequent surface saturation and depletion of reactants for several cases (e.g., on mineral dust and soot, Ndour et al., 2009; Stephens et al., 1986; Ammann et al. 1998; Kalberer et al. 1999). In general, the upper limit and lower limit are determined based on those derived using the geometric surface and the BET (Brunauer-Emmett-Teller) surface, respectively. Preferences are given to those measured at steady state using ambient aerosols, or recommended values by the IUPAC group with relatively high reliability. As shown in Table A.1, more than 3 orders of magnitude of variances are found for SO_2 and O_3 uptake on mineral dust depending on the gas concentration and aerosol components (Michel et al., 2002, 2003; Mogili et al., 2006; Ullerstam et al., 2002, 2003; Li et al., 2006). Large discrepancies also exist for SO_2 and HNO_3 uptake on soot (Longfellow et al., 2000; Saathoff et al., 2001; Xu et al., 2015). For H_2O_2 , limited measurements of γ_{eff} have been conducted for aerosols apart from mineral dust and soot.

2.3 Uptake coefficient at equivalent flux (γ_{eqv})

To help the evaluation, we define an uptake coefficient at equivalent flux γ_{eqv} . Here, γ_{eqv} is the effective uptake coefficient on aerosols when the ground flux equals the aerosol flux within the PBL. When $\gamma_{\text{eff}} > \gamma_{\text{eqv}}$, the aerosol surfaces are more important than the ground surfaces regarding the gas uptake and vice versa. By letting F_{grd} equal F_{aer} , we can derive the expression of γ_{eqv} as below.

$$\gamma_{\text{eqv}} = \frac{4 V_d}{3 Ah} \times 10^2 \quad (6)$$

and in a typical mixing height of 300m, we have

$$\gamma_{\text{eqv}} = \frac{V_d}{2.25A} \quad (7)$$

According to Eq. (6), γ_{eqv} is proportional to V_d , and is inversely proportional to aerosol surface area density and the mixing height. We calculated a series of γ_{eqv} for a variety of gas species, land use categories, seasons, aerosol surface area densities (A) and mixing heights (h).

As defined, γ_{eqv} reflects the relative importance of gas uptake on aerosols compared to those on the ground surfaces. Larger γ_{eqv} indicates higher probability for gases to deposit on the ground rather than on aerosols for further chemical reactions on surface and bulk, and vice versa. Low dry deposition velocities and high loadings of aerosols providing large amounts of surface reaction sites can benefit gas uptake on aerosols. The derived γ_{eqv} and γ_{eff} from laboratory measurements are compared in Sect. 3.

3. Results and discussion

To estimate the possible range of γ_{eqv} for different environments, we designed different scenarios with mixing height h varying between 100 m and 1.0 km (a typical value of 300m), and A varying with land use categories as follows:

(a) Range of A . We set the range of A based on measurements in various environments collected in literature. A are in the range of $200 \sim 2000 \mu\text{m}^2 \text{cm}^{-3}$ for aerosols in the urban area (Woo et al., 2001; Stanier et al., 2004; Wu et al., 2008, 2017; Ma and Birmili, 2015), $200 \sim 1000 \mu\text{m}^2 \text{cm}^{-3}$ in agricultural land (sub-urban and rural, Held et al., 2008; Su et al. 2008; Ma et al., 2014; Ma and Birmili, 2015; Wu et al., 2017), $8 \sim 700 \mu\text{m}^2 \text{cm}^{-3}$ in Amazon forest (Zhou et al., 2002; Rissler et al., 2006; Pöschl et al., 2010; Andreae et al. 2015), and $20 \sim 200 \mu\text{m}^2 \text{cm}^{-3}$ for sea salt aerosols (SSA, O'Dowd et al., 1997; Ghan et al., 1998; Lewis and Schwartz, 2004).

(b) Typical A (corresponding to the typical γ_{eqv} in Fig. 3 ~ Fig. 5). We use $1050 \mu\text{m}^2 \text{cm}^{-3}$ for the urban environment (Wang et al., 2017), $230 \mu\text{m}^2 \text{cm}^{-3}$ for the agricultural land (Held et al., 2008), $46 \mu\text{m}^2 \text{cm}^{-3}$ for the Amazon forest (Rissler et al., 2006), and $76 \mu\text{m}^2 \text{cm}^{-3}$ for SSA (canonical distribution at wind speed of 10 m s^{-1} , Lewis and Schwartz, 2004).

It should be noted that the above ranges and the typical values of A are derived from current available experiments to support our analyses and discussions in this study, but still cannot cover all cases of particle distributions in the world.

Figure 2 shows the calculated γ_{eqv} over a range of dry deposition velocity and aerosol surface area densities at a mixing height of 300m. V_d for different scenarios were calculated based on the resistance scheme illustrated above, showing a range of $0.01 \sim 2.3 \text{ cm s}^{-1}$, with lowest for NO_2 and highest for N_2O_5 and HNO_3 (details in Table S2). Aerosol surface area densities covered a range of $8.6 \mu\text{m}^2 \text{cm}^{-3}$ to $2139 \mu\text{m}^2 \text{cm}^{-3}$, from pristine rainforest to polluted megacities. We show the calculated γ_{eqv} at typical conditions (typical A as described above, $h=300\text{m}$) by season in Table S3 and detailed illustrated γ_{eqv} for each gas species in sections below. As shown in Fig.2, γ_{eqv} decreases with increase of A , which is closely related to the air pollution level, and increases with increasing V_d .

For small V_d ($\leq 0.1 \text{ cm s}^{-1}$), γ_{eqv} lie in the range of $10^{-5} \sim 10^{-4}$ for clean regions, such as Leipzig, Melpitz, Pittsburgh, and reduced to $10^{-6} \sim 10^{-5}$ under polluted cities including Beijing and Wangdu. This low dry deposition can be found for NO_2 above the urban ground (0.03 cm s^{-1} , seasonal mean), and O_3 , NO_2 , SO_2 and H_2O_2 on water body (0.07 cm s^{-1} , 0.01 cm s^{-1} , 0.03 cm s^{-1} , and 0.08 cm s^{-1} , respectively). The downward shift of γ_{eqv} with larger aerosol surface area density suggests an increasing importance of gas uptake in polluted areas than clean areas.

With the increase of V_d ($> 0.1 \text{ cm s}^{-1}$), γ_{eqv} increases to $10^{-5} \sim 10^{-2}$ accordingly. In pristine region of Amazon forest, γ_{eqv} can reach up to 10^{-2} . The lowest γ_{eqv} is 2.1×10^{-5} during haze events with high concentrations of fine particulate matter and surface area in the PBL ($A=2139 \mu\text{m}^2 \text{ cm}^{-3}$). In this study, this range of V_d covers most of the investigated cases, including O_3 , SO_2 , H_2O_2 on urban/Amazon forest/agricultural land, NO_2 on agricultural land/Amazon forest, and N_2O_5 , HNO_3 on all land use types (see Table S2). Thus we can derive a general criterion of $\gamma_{\text{eff}} > 10^{-5}$ conservatively for aerosol uptake to compete with the dry deposition.

In the following, we further compared γ_{eqv} to the laboratory measurements of γ_{eff} for different reactive gases (O_3 , NO_2 , SO_2 , N_2O_5 , HNO_3 , H_2O_2). The uptake coefficients at initial state are in general 1~3 magnitudes higher than those at steady-state (see Table 1 and Fig. 3~5). Considering the timescale of gas uptake by aerosols in the real world and applications in models, we mainly focused on the comparisons of γ_{eqv} and the steady-state γ_{eff} in the following discussions.

3.1 O_3

Under typical conditions (typical A by land use, $h=300\text{m}$, as illustrated above), γ_{eqv} for O_3 are determined between 9.2×10^{-5} and 2.2×10^{-3} , lowest in urban and highest in the Amazon forest. The extended range is $1.4 \times 10^{-5} \sim 3.8 \times 10^{-2}$, varying with particle area densities and mixing heights (Fig. 3). There are overlaps between γ_{eqv} and γ_{eff} for liquid organic aerosols among all investigated typical environments, and other types of aerosols under favorable circumstances for aerosol uptake in urban. γ_{eff} lie below γ_{eqv} for other combinations of aerosol types and land use categories.

We can only expect comparable uptake between ground and aerosol surfaces of mineral dust, soot, solid organic aerosol, and SSA at high aerosol loadings in urban (e.g., $A=1400 \mu\text{m}^2 \text{ m}^{-3}$, Beijing) and/or high mixing layers (e.g., $h=1.0 \text{ km}$). Combined with the measured uptake coefficients which lie in the range of 1.0×10^{-7} to 1.6×10^{-4} for soot, 1.1×10^{-5} to 3.0×10^{-3} for liquid organic aerosols and 1.3×10^{-6} to 1.0×10^{-4} for SSA, we can expect high uptake fluxes of O_3 on these three kinds of aerosols when corresponding γ_{eff} larger than 10^{-4} for other ground surfaces.

Complexity comes from the organic aerosols, of which the phase state has a large impact on the uptake and is subject to the temperature, relative humidity and particle size (see Fig. 3) (Virtanen et al. 2010; Cheng et al. 2015). For liquid organic aerosols, the measured γ_{eff} show large variances from 10^{-5} to 10^{-3} and corresponding γ_{eqv} fall into this range, demonstrating that O_3 uptake on aerosols is comparable to that on the ground. Thus, multiphase reactions of O_3 on liquid organic aerosols

should be included in atmospheric models. This is also consistent with the findings of Mu et al. (2018), which demonstrates the importance of the phase state of aerosols in multiphase reactions and transport of polycyclic aromatic hydrocarbons to improve the model performances at both regional and global scales.

Shiraiwa et al. (2017) shows the global map of SOA (secondary organic aerosol) phase state at the earth's surface. SOA in Southern China, Amazon forest and South Africa are mainly in liquid phase within PBL. For these regions, the comparable uptake fluxes for O_3 on liquid organic aerosols compared to the dry deposition demonstrate the importance of aerosol uptake. Dry deposition is one of the major sinking pathways for O_3 (Ganzeveld and Lelieveld, 1995). The uptakes of O_3 by aerosols are expected to contribute comparable sink fluxes as dry deposition regionally. Inclusion of the O_3 uptake by organic aerosols in these regions will increase the deposition rate of O_3 on aerosols, affect its lifetime, and further affect the fate of HO_x , NO_x through chemical reactions in the gas phase.

3.2 NO_2

For NO_2 , γ_{eqv} are generally above the upper limit of γ_{eff} in urban, agricultural land and forest environments, as shown in Figure 4, demonstrating that the ground surfaces are of greater importance than aerosols. Overlaps are found for SSA on various land use types and also for liquid organic aerosols under the urban environment.

NO_2 tend to deposit on ground surface instead of on mineral dust particles, soot and solid organic aerosols. As reviewed in Table A.1~A.3, the effective uptake coefficient of NO_2 on these three kinds of aerosols are at magnitudes of $< 10^{-6}$ under steady-state conditions. For A ranging from $46 \mu m^2 cm^{-3}$ (Amazon) to $1050 \mu m^2 cm^{-3}$ (Wangdu) and mixing height of 300 m, γ_{eqv} of NO_2 lie between 1.4×10^{-5} and 1.3×10^{-3} , 1~3 orders of magnitudes larger than γ_{eff} on these three kinds of aerosols. Increasing the PBL mixing height and aerosol surface area may reduce γ_{eqv} by ~1-2 magnitudes, but are still above the measured γ_{eff} at steady state.

The reactive uptake coefficients of NO_2 by SSA were quantified in the range of 10^{-6} to 10^{-4} , demonstrating the ability of ambient sea salt aerosols to take in chemical species like NO_2 (Harrison and Collins, 1998; Yabushita et al., 2009; Ye et al., 2010). The high uptake coefficients observed for SSA ($6.0 \times 10^{-7} - 3.0 \times 10^{-4}$) are probably attributed to the reactions of Cl^- with dissolved NO_2 in aqueous phase (Msibi et al., 1993; Harrison and Collins, 1998; Yabushita et al., 2009). The overlapped values of γ_{eqv} and γ_{eff} , show that the NO_2 uptake by SSA is comparable to the uptake by land surface or water body in coastal areas and therefore should be taken into account in atmospheric models.

Another important process is the NO_2 uptake on liquid organic aerosols (γ_{eff} in the range of $2.2 \times 10^{-7} - 7.0 \times 10^{-6}$) in urban area of high A . As shown in Fig. 4, the lower limit of γ_{eqv} in urban is $\sim 2.2 \times 10^{-6}$, lying in the range of γ_{eff} . The uptake coefficients of NO_2 on pure water are estimated around $10^{-7} \sim 10^{-6}$ driven by low solubility and slow hydrolysis rates (Kleffmann et al., 1998; Gutzwiller et al., 2002; Ammann et al., 2005; Komiyama and Inoue, 1980). Harrison and Collins

(1998) reported a high γ_{eff} of $5.4 \sim 5.8 \times 10^{-4}$ for NO_2 uptake on ammonium sulfate aerosols at high RH (RH=50%, 85%). Presence of reactants such as inorganics of HSO_3^- or phenolic compounds in aqueous aerosols can promote the uptake significantly through chemical reactions with dissolved NO_2 to $10^{-5} \sim 10^{-4}$ (Msibi et al., 1993; Lee and Tang, 1998; Spindler et al. 2003; Ammann et al., 2005; Yabushita et al., 2009; Su et al. 2008; Cheng et al., 2016). Multiple measurements and modeling work have also pointed out that high alkalinity of aqueous aerosols is key to promote the reactions and further increase the NO_2 uptake rates (Ammann et al., 2005; Herrmann et al., 2015; Cheng et al., 2016). Therefore, the NO_2 uptake on alkaline aqueous aerosols containing organic/inorganic reactants is competitive in the urban atmosphere, and should be detailed addressed in models. In Amazon forest, where A is too low ($46 \mu\text{m}^2 \text{cm}^{-3}$), corresponding to a γ_{eqv} on the order of 10^{-3} , even a high γ_{eff} of 10^{-4} is not sufficient to compete with the uptake by the ground surfaces.

In summary, the NO_2 uptake coefficients on liquid aerosol droplets can vary by three orders of magnitude with aerosol compositions ($10^{-7} \sim 10^{-4}$). On liquid organic aerosols and sea salt aerosols, the uptake can reach up to $10^{-6} \sim 10^{-4}$ through chemical reactions (Abbatt and Waschewsky, 1998; Ammann et al., 2005; Yabushita et al., 2009), significantly larger than the uptake on pure water of $10^{-7} \sim 10^{-6}$ (Lee and Tang, 1988; Kleffmann et al., 1998; Gutzwiller et al., 2002). For liquid ammonium sulfate aerosols, discrepancies with two orders of magnitude ($10^{-6} \sim 10^{-4}$) in γ_{eff} are found with reasons unexplained yet (Harrison and Collins, 1998; Tan et al., 2016). Considering these variances, aerosol components are important to parameterize the γ_{eff} in atmospheric models.

3.3 SO_2

The calculated γ_{eqv} of SO_2 vary between 1.0×10^{-4} and 2.1×10^{-3} for land surfaces, and 1.7×10^{-4} above water body under typical conditions. As shown in Fig. 4, the SO_2 uptake by mineral dust is comparable to the ground uptake in urban, and under favorable conditions over agricultural land and water body. For soot, aerosol uptake is magnitudes lower than those on the ground ($\gamma_{\text{eqv}} \geq \gamma_{\text{eff}}$), thus is unimportant for SO_2 . For SSA, γ_{eff} of $3.2 \times 10^{-3} \sim 1.7 \times 10^{-2}$ has been reported for SO_2 at aerosol pH of 5.4~6.6, which is high enough to compete with dry depositions over most environments (Gebel et al., 2000). Additional reactions of SO_2 and O_3 in alkaline solutions are found to promote the SO_2 uptake and form sulfate on SSA at first stage (Laskin et al., 2003). However, aerosol acidification due to production of H^+ has been suggest to quickly suppress the oxidation process in the real world (Alexander et al., 2005). We suggest including both the SO_2 uptake on SSA and the aerosol acidification process in models.

The extended range of γ_{eqv} is $1.6 \times 10^{-5} \sim 1.6 \times 10^{-3}$, $5.5 \times 10^{-5} \sim 2.8 \times 10^{-3}$, and $1.9 \times 10^{-5} \sim 1.9 \times 10^{-3}$ for urban, agricultural land and water body, respectively. γ_{eff} of mineral dust falls in this range 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, because it can be affected by the presence of oxidant, phase state, components of the tested dust and the use of surface area in calculation

(Huss et al., 1982; Ullerstam et al., 2003; Li et al., 2006; Alexander et al., 2009; Zhang et al., 2018). We further discuss the SO₂ uptake on mineral dust by different conditions as below.

Under dry conditions (as reviewed in Table A.1), γ_{eff} are measured on the order of $10^{-7} \sim 10^{-4}$ (Goodman et al., 2001; Usher et al., 2002; Ullerstam et al., 2003; Adams et al., 2005; Li et al., 2006). IUPAC recommended an averaged value of 4×10^{-5} for atmospheric modeling, based on measurements using airborne aerosols (Usher et al., 2002; Adams et al., 2005).

In environments with high RH, water can enhance or inhibit the uptake by affecting reactive sites, varying with experimental conditions (Huang et al., 2015; Zhang et al., 2018). On the other hand, the uptake rate can be improved by several factors and/or aqueous chemical reactions, such as presence of O₃, H₂O₂, and transition metal ions (TMIs), which strongly depends on the aerosol pH (Jayne et al., 1990; Li et al., 2006; Cheng et al., 2016; Zhang et al., 2018). The initial γ_{eff} of SO₂ on pure water can reach as high as $10^{-3} \sim 0.1$ varying with pH (Gardner et al., 1987; Worsnop et al., 1989; Jayne et al., 1990; Ponche et al., 1993). Depending on aerosol pH and oxidant concentrations, the regimes of SO₂ uptake and sulfate formation may transit from TMI- or H₂O₂-dominated regime to NO₂- or O₃-dominated regime (Cheng et al., 2016). In this case, the SO₂ uptakes on aqueous aerosols are expected to play the dominant roles over dry deposition under specific circumstances such as the haze event (He et al., 2014; Cheng et al., 2016), which should be quantified combining in-situ measurements and atmospheric modeling.

As shown in the examples of Table S4, several model schemes adopt an γ_{eff} of $\sim 10^{-4}$ (Liao and Seinfeld, 2005; Wang K et al., 2012), around one order of magnitude higher than the measured values on low RH conditions (Usher et al., 2002; Ullerstam et al., 2003; Adams et al., 2005; Li et al., 2006). For example, in Liao and Seinfeld (2005), γ_{eff} is 3.0×10^{-4} for RH < 50%, and 0.1 for RH \geq 50% (see Table S4 with references). At low RH, the uptake coefficient commonly used in model is based on the dry deposition measurement of SO₂ on calcareous soils, cements and Fe₂O₃, rather than laboratory measured γ_{eff} values that have been recommended by IUPAC. The reason for this divergence is unclear and we are in favor of using the IUPAC recommended γ_{eff} (e.g., Zhu et al., 2010, as shown in Table S4). The high uptake coefficient in model at high RH is based on two assumptions: fast oxidation of SO₂ by O₃ in the aqueous phase and high alkalinity in the dust aerosols. Thus this γ_{eff} should be applied with caveats that these prerequisites have been fulfilled, especially when extending it for other type of aerosols (Zheng et al. 2015).

3.4 N₂O₅, HNO₃, and H₂O₂

N₂O₅, HNO₃, and H₂O₂ demonstrate their high uptake ability on atmospheric aerosols, as shown in Fig. 5. For N₂O₅, the similar or higher values of γ_{eff} over γ_{eqv} demonstrate that the multiphase uptake by all types of aerosols are as important as or even more important than dry deposition. The N₂O₅ uptake by aerosols has been widely included in models (Bauer et al.,

2004; Liao and Seinfeld, 2005; Wang K et al., 2012). The uptake of HNO_3 and H_2O_2 by mineral dust and HNO_3 by SSA are important given the overlap between γ_{eff} and γ_{eqv} , thus should also be detailed characterized in atmospheric models.

For N_2O_5 , the measured uptake coefficients are $4.8 \times 10^{-3} \sim 0.20$ for mineral dust, $4.0 \times 10^{-5} \sim 6.3 \times 10^{-3}$ for soot, and $6.4 \times 10^{-3} \sim 3.9 \times 10^{-2}$ for SSA, which are comparable to or 1~2 magnitudes higher than the calculated γ_{eqv} of $9.3 \times 10^{-4} \sim 2.1 \times 10^{-2}$ under typical conditions (details in Table A.1~A.4). For other kinds of aqueous aerosols, e.g., ammonium sulfate aerosols with high RH, N_2O_5 can also be taken up very efficiently with γ_{eff} of $10^{-3} \sim 10^{-2}$ (Kane et al., 2001; Schötze and Herrman, 2002; Hallquist et al., 2003; Badger et al., 2006). The importance of N_2O_5 and HNO_3 uptake by aerosols has been sufficiently addressed in previous studies (Evans and Jacob, 2005; Liao and Seinfeld, 2005; Stadtler et al., 2018). In current CTMs, γ_{eff} of N_2O_5 is explicitly calculated as a function of temperature and RH, of which the relation was determined from laboratory experiments (Kane et al., 2001; Bauer et al., 2004; Liao and Seinfeld, 2005).

The extended range of γ_{eqv} for HNO_3 is $1.5 \times 10^{-4} \sim 1.5 \times 10^{-2}$ (urban), $1.5 \times 10^{-4} \sim 7.7 \times 10^{-3}$ (agricultural land), $4.2 \times 10^{-4} \sim 3.7 \times 10^{-1}$ (Amazon) and $7.0 \times 10^{-4} \sim 7.0 \times 10^{-2}$ (water), which are within or below the range of γ_{eff} for mineral dust and SSA. The higher γ_{eff} of 1.0×10^{-3} to 0.21 for mineral dust and of 5.0×10^{-4} to 0.25 for SSA demonstrated a more important role of aerosol uptake than that of the ground surfaces. The uptake of HNO_3 on soot and solid organic aerosols appear to be less important. The HNO_3 uptake on mineral dust have been implemented in current models with an uptake coefficient of 0.1, or between 1.1×10^{-3} and 0.2, consistent with the range of experimentally determined γ_{eff} reviewed in this study (Liao and Seinfeld, 2005; Wang K et al., 2012).

The study on the uptake of H_2O_2 by aerosols is rather limited compared to other trace gases aforementioned. The reported γ_{eff} on dust and ambient aerosol samples suggest aerosol uptake is more important than that by the ground surface. The measured uptake coefficients of H_2O_2 on mineral dust are in the range of $1.0 \times 10^{-5} \sim 9.4 \times 10^{-4}$, overlapped with the calculated γ_{eqv} of $1.5 \times 10^{-4} \sim 3.0 \times 10^{-3}$ under typical conditions. Ambient aerosols collected in urban area show similar γ_{eff} of H_2O_2 ($8.1 \times 10^{-5} \sim 4.6 \times 10^{-4}$) to mineral dust (Wu et al., 2015). The aerosol chemistry of H_2O_2 in the troposphere is complex and unclear (Liang et al., 2013; Li et al., 2016). In some cases, a net emission of H_2O_2 from aerosol surfaces was speculated instead of an uptake or adsorption as a result of HO_x radicals cycling (Liang et al., 2013; Li et al., 2016). Most models only parameterize the H_2O_2 uptake by dust particles (Dentener et al., 1996; Wang K et al., 2012). The uptake by other aerosol types hasn't been considered due to limited experimental data. More laboratory kinetic measurements are thus needed. Since ambient aerosol samples show a γ_{eff} similar to that of dust particles (Wu et al., 2015; Pradhan et al., 2010ab; Zhou et al., 2016), we suggest adopting the γ_{eff} of dust particles and applying it to all aerosol types before new kinetic data become available.

4. Discussion

In this section, we address several important issues based on the comparisons. Large variability found in the measured γ_{eff} for SO_2 and NO_2 are discussed in Sect. 4.1. How to apply the measured γ_{eff} in atmospheric models to represent the reactivity of heterogeneous reactions still remains an open question. Regarding this, we discuss the underlying important factors that should be taken into account in Sect. 4.2. Outlooks and limitation of this work are provided in Sect. 4.3.

4.1 Large variability of γ_{eff} for SO_2 and NO_2

Notably, there is a large variability in the reviewed γ_{eff} of SO_2 uptake by dust particles (as discussed in Sect. 3.2). For SO_2 uptake by dust particles, more than three orders of magnitude of differences are found for its uptake by mineral dust ($10^{-8} \sim 10^{-4}$, steady state), which may be attributed to several factors such as the experimental particle substrates, co-existing oxidants (O_3 , H_2O_2 , NO_2), RH, measurement techniques and surface area used in data processing (Ullerstam et al., 2003; Li et al., 2006; Huang et al., 2015). For example, a γ_{eff} of 1.6×10^{-4} was derived for SO_2 uptake on Al_2O_3 powder (Usher et al., 2002). The uptake coefficient was reduced by one order of magnitude to $1.6 \sim 6.6 \times 10^{-5}$ using ambient aerosols of Chinese loess / Saharan dust (Usher et al., 2002; Ullerstam et al., 2003; Adams et al., 2005), indicating that the particle substrate is key in investigating SO_2 uptake. Similarly, through cross comparisons between other different investigations shown in Table A.1, we anticipate that the above factors can all contribute to this large discrepancy. As recommended by IUPAC, an uptake coefficient of 4×10^{-5} based on airborne measurements is suggested to use in models on low RH conditions. For high RH, we suggest determining γ_{eff} with information of aerosol pH because of the high correlation between them as illustrated in Sect. 3.3.

For NO_2 uptake on liquid aerosol droplets, three orders of magnitudes of differences are found ($10^{-7} \sim 10^{-4}$), varying significantly with aerosol compositions. On pure water, the uptake is measured at $10^{-7} \sim 10^{-6}$ (Lee and Tang, 1988; Kleffmann et al., 1998; Gutzwiller et al., 2002). On liquid organic aerosols and sea salt aerosols, the uptake can be effectively accelerated to $10^{-6} \sim 10^{-4}$ through multiphase reactions (Abbatt and Waschewsky, 1998; Ammann et al., 2005; Yabushita et al., 2009). For ammonium sulfate aerosols, large discrepancies of $10^{-6} \sim 10^{-4}$ for the initial γ_{eff} are found with reasons unexplained yet (Harrison and Collins, 1998; Tan et al., 2016). Based on the reviewed measurements, we suggest using a relatively high uptake coefficient ($\sim 10^{-4}$) for aqueous aerosols containing reactants, and a lower value ($< 10^{-6}$) for other cases.

4.2 Initial vs steady state, geometric vs BET

Measurements of effective uptake coefficients revealed the instantly fast uptake at the initial state and gradually declined due to the saturation of surface reaction sites and loss of reactive substances (Hanisch and Crowley, 2003). The uptake at the initial state can be faster of orders of magnitudes higher than that at the steady state for aerosols (see Table 1 and Table

A.1~A.4). The time scale reaching surface saturation/equilibrium is dependent on the reaction system. For gas-aqueous particle surface, the timescale to establish equilibrium for the investigated species is less than 1s (Seinfeld and Pandis, pp 554-557, 2006). For dust particles, it can take hours for complete saturation (Judeikis et al., 1978; Goodman et al., 2001). Fine particles with diameters $<10\ \mu\text{m}$ have lifetimes of several days in the atmosphere (Prospero, 1999; Lee et al., 2009).

5 Thus using uptake coefficients at steady state maybe more representative in models, unless we can assume that the uptake process is not limited by surface accommodation and reactions (like HNO_3 , Goodman et al., 2000), typically when the gas concentration is low enough so the surface passivation is negligible compared to the lifetime of aerosols in the atmosphere (Hanisch and Crowley, 2003). Gas uptake on fresh aerosols may reach or even surpass the level of the ground near emitting sources. Using a uniform uptake coefficient in atmospheric models may not be enough to reflect the deactivation process of

10 the multiphase gas uptake during aerosol aging, considering the large range of γ_{eff} varying with time.

In addition, γ_{eff} are measured and reported based on the geometric surface or/and the BET surface. 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).** In this study, γ_{eff} with revised BET surface are generally used as the lower limit, and those using the geometric

15 surface as the upper limit. Whether using BET area as a correction in the calculation of γ_{eff} or not remains discrepancy when applied in models (Hanisch and Crowley, 2001ab; Underwood et al., 2001ab). This discrepancy from measurements may come from the differences in experimental samples (airborne particles vs powder). To solve this issue, more studies on the reactive surface area for ambient aerosols are needed to guide the data processing and model parameterization.

4.3 Outlooks and limitations

20 We can conclude that phase state is a crucial factor influencing the uptake rates. The uptake rates of O_3 and NO_2 in liquid organic aerosols are 1~3 orders of magnitudes higher than on solid / semi-solid surfaces. In regions with high RH and sufficient source of organic compounds (e.g., Amazon forest, southern China), the gas uptake is anticipated to have considerable effect on concentrations. The effect is yet to be evaluated combined with further model simulations.

Measurement of uptake by ambient aerosols is crucial to reconcile lab experiments and modeling results, especially for gas

25 with limited investigation conducted (e.g., H_2O_2). Currently limited work has been done to address the uptake of H_2O_2 by aerosol particles other than mineral dust (Liao and Seinfeld, 2005; Pradhan et al., 2010 ab; Wang K et al., 2012; Zhou et al., 2016). Because ambient aerosol samples show a γ_{eff} comparable to that of dust particles, we recommend similar gamma values of $1.0 \times 10^{-5} \sim 9.4 \times 10^{-4}$ for H_2O_2 uptake by other types of aerosol, which will lead a larger sink in the atmospheric budget of H_2O_2 .

Considering the complexity of multiple factors affecting the uptake rates, such as temperature, RH, gas concentration, aerosol pH, and aerosol state (fresh or aged), establishing a look-up table for γ_{eff} based on available factors above should be a feasible way to implement the gas uptake processes in atmospheric models (Mu et al., 2018).

There are limitations for the comparisons conducted in this study. 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. The ambient parameters to calculate the dry deposition velocities (temperature, radiation) refer to the standard meteorological database for construction in northern China (Zhang, 2004), which may introduce uncertainties for analyses of other areas. In addition, the variability of aerosol surface area under each environment can also contribute to the variability of γ_{eqv} . We mainly focused on the uptake fluxes at room temperature ($\sim 298\text{K}$). The gas uptakes at very low temperature (e.g., polar region, stratosphere) are out of scope of this study but should be further explored concerning its potentially large impact. The real ambient multiphase processes are much more complex than the laboratory measurements nevertheless they use airborne aerosols. Ambient on-line measurements of γ_{eff} will favor the model parameterization and improve our understanding of the multiphase processes within PBL in the real world (Li et al., 2019). Moreover, more gaseous and aerosol species such as VOCs and bioaerosols should also be investigated (Zhou et al., 1996; Wagner et al., 2002; Fried et al., 2003; Beck et al., 2013; Li et al., 2014; Li et al. 2016; Ouyang et al., 2016; Liu et al. 2017; Meusel et al. 2017).

5. Conclusions

In this work, we investigated the relative importance of gas uptake fluxes on ground and aerosols for six reactive trace gases (O_3 , NO_2 , SO_2 , N_2O_5 , HNO_3 , H_2O_2), various environments, aerosol types and mixing heights. The purpose is to identify aerosol uptake process which is equally or more important than the dry deposition on ground surfaces but has not been adequately addressed in models.

For efficient comparison, we derived a criterion, γ_{eqv} , to identify which kind of surface is dominant in gas uptake. For investigated gas species, γ_{eqv} generally lie in the magnitude of 10^{-4} , and can be extended to lower values in polluted areas and/or low dry deposition velocities. Especially, γ_{eqv} lie in the range of $10^{-6} \sim 10^{-4}$ in polluted urban environments and $10^{-4} \sim 10^{-1}$ under pristine forest conditions. The effective uptake coefficient (γ_{eff}) derived from experiments are reviewed and compared with γ_{eqv} . Notably, the gas uptake by aerosols is comparable and should be considered in models when γ_{eff} is equal to or higher than γ_{eqv} . In urban environments, aerosol uptake is important for all combinations of gases and aerosols, favored by the high particle surface densities. On the contrary, the contribution of aerosol uptake is minor compared to dry deposition for gases in the Amazon forest.

The following gas uptake by aerosols can be as important as the dry deposition processes and should be considered in atmospheric models: N_2O_5 on all types of aerosols, HNO_3 and SO_2 on mineral dust and sea salt aerosols, H_2O_2 on mineral dust, NO_2 on sea salt aerosols and O_3 on liquid organic aerosols ($\gamma_{\text{eff}} \geq \gamma_{\text{eqv}}$, as shown in Table 2). The gas uptake on mineral dust for most gases and sea salt aerosols uptake of SO_2 and NO_2 have already been parameterized in a series of models. The processes of H_2O_2 uptake on mineral dust and O_3 on liquid aerosols haven't received enough attention unfortunately. For other combinations of gas species and aerosols, the ground tends to be the dominant surface rather than aerosols to take up trace gases within PBL.

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

- (a) It is indicated that the multiphase processes for O_3 on liquid organic aerosols are underestimated in current atmospheric models. For regions with high RH and the existence of organic aerosols at liquid state such as Southern China, Amazon forest and South Africa, the multiphase uptakes of O_3 by aerosols are expected to contribute comparable sinking fluxes as dry deposition. Compared to the relatively low uptakes on (semi-) solid organic aerosols, we can conclude that phase state is a crucial factor influencing the uptake rates.
- (b) Large uncertainties should be addressed for the comparison results of SO_2 and NO_2 . There are more than three orders of magnitude of variances in γ_{eff} for SO_2 on mineral dust and NO_2 on aqueous aerosols. Under low RH circumstances, dry deposition tends to dominate the gas uptake rather than aerosols. However, for cases in high RH, the contributions of aerosols should be cautiously determined with full considerations of the aerosol component, aerosol pH, etc.
- (c) H_2O_2 uptake on a variety of aerosols is needed to be measured and evaluated. It's shown that the H_2O_2 uptake on dust is comparable or even more important than that by the ground surface ($\gamma_{\text{eff}} \geq \gamma_{\text{eqv}}$). Measurements using ambient aerosols suggest that the uptake on aerosols other than mineral dust should be of similar magnitude.

Data availability. All parameters to calculate V_d , the aerosol surface area densities (A), and the laboratory measurements of γ_{eff} are derived from peer-reviewed literature or publicly available database (as illustrated in the main text).

Author contribution. H.S. and Y.C. designed the research. M.L. performed the research with input from H.S., Y.C., and N.M.. U.P. and G.L. discussed the results. M.L., H.S. and Y.C. wrote the manuscript with input from all-co-authors.

Competing interests. The authors declare no conflict of interests.

Acknowledgements. We acknowledge the National Natural Science Foundation of China (91644218), National Key Research and Development Program of China (Grant 2017YFC0210104), and Guangdong Innovative and Entrepreneurial

Research Team Program (2016ZT06N263). This work was supported by the Max Planck Society (MPG). Y.C. also acknowledges the Minerva Program of MPG.

References

- Abbatt, J. P. D., Lee, A. K. Y., and Thornton, J. A.: Quantifying trace gas uptake to tropospheric aerosol: recent advances and remaining challenges, *Chemical Society Reviews*, 41, 6555-6581, doi: 10.1039/C2CS35052A, 2012.
- 5 Abbatt, J. P. D., and Waschewsky, G. C. G.: Heterogeneous Interactions of HOBr, HNO₃, O₃, and NO₂ with Deliquescent NaCl Aerosols at Room Temperature, *The Journal of Physical Chemistry A*, 102, 3719-3725, doi: 10.1021/jp980932d, 1998.
- Adams, J. W., Rodriguez, D., and Cox, R. A.: The uptake of SO₂ on Saharan dust: a flow tube study, *Atmos. Chem. Phys.*, 5, 2679-2689, doi: 10.5194/acp-5-2679-2005, 2005.
- 10 Andreae, M. O.: A New Look at Aging Aerosols, *Science*, 326, 1493, doi: 10.1126/science.1183158, 2009.
- Al-Abadleh, H. A., and Grassian, V. H.: Heterogeneous Reaction of NO₂ on Hexane Soot: A Knudsen Cell and FT-IR Study, *The Journal of Physical Chemistry A*, 104, 11926-11933, doi: 10.1021/jp002918i, 2000.
- Alexander, B., Park, R. J., Jacob, D. J., and Gong, S.: Transition metal-catalyzed oxidation of atmospheric sulfur: Global implications for the sulfur budget, *Journal of Geophysical Research: Atmospheres*, 114, doi: 10.1029/2008JD010486, 15 2009.
- Alexander, B., Park, R. J., Jacob, D. J., Li, Q. B., Yantosca, R. M., Savarino, J., Lee, C. C. W., and Thiemens, M. H.: Sulfate formation in sea-salt aerosols: Constraints from oxygen isotopes, *Journal of Geophysical Research: Atmospheres*, 110, doi: 10.1029/2004JD005659, 2005.
- Ammann, M., Cox, R. A., Crowley, J. N., Jenkin, M. E., Mellouki, A., Rossi, M. J., Troe, J., and Wallington, T. J.: 20 Evaluated kinetic and photochemical data for atmospheric chemistry: Volume VI – heterogeneous reactions with liquid substrates, *Atmos. Chem. Phys.*, 13, 8045-8228, doi: 10.5194/acp-13-8045-2013, 2013.
- Ammann, M., Kalberer, M., Jost, D. T., Tobler, L., Rössler, E., Piguet, D., Gägeler, H. W., and Baltensperger, U.: Heterogeneous production of nitrous acid on soot in polluted air masses, *Nature*, 395, 157, doi: 10.1038/25965, 1998.
- Ammann, M., and Pöschl, U.: Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions 25 – Part 2: Exemplary practical applications and numerical simulations, *Atmos. Chem. Phys.*, 7, 6025-6045, doi: 10.5194/acp-7-6025-2007, 2007.
- Ammann, M., Rössler, E., Strekowski, R., and George, C.: Nitrogen dioxide multiphase chemistry: Uptake kinetics on aqueous solutions containing phenolic compounds, *Physical Chemistry Chemical Physics*, 7, 2513-2518, doi: 10.1039/B501808K, 2005.

- Andreae, M. O., Acevedo, O. C., Araùjo, A., Artaxo, P., Barbosa, C. G. G., Barbosa, H. M. J., Brito, J., Carbone, S., Chi, X., Cintra, B. B. L., da Silva, N. F., Dias, N. L., Dias-Júnior, C. Q., Ditas, F., Ditz, R., Godoi, A. F. L., Godoi, R. H. M., Heimann, M., Hoffmann, T., Kesselmeier, J., Könemann, T., Krüger, M. L., Lavric, J. V., Manzi, A. O., Lopes, A. P., Martins, D. L., Mikhailov, E. F., Moran-Zuloaga, D., Nelson, B. W., Nölscher, A. C., Santos Nogueira, D., Piedade, M. T. F., Pöhlker, C., Pöschl, U., Quesada, C. A., Rizzo, L. V., Ro, C. U., Ruckteschler, N., Sá, L. D. A., de Oliveira Sá, M., Sales, C. B., dos Santos, R. M. N., Saturno, J., Schöngart, J., Sörgel, M., de Souza, C. M., de Souza, R. A. F., Su, H., Targhetta, N., Tóta, J., Trebs, I., Trumbore, S., van Eijck, A., Walter, D., Wang, Z., Weber, B., Williams, J., Winderlich, J., Wittmann, F., Wolff, S., and Yáñez-Serrano, A. M.: The Amazon Tall Tower Observatory (ATTO): overview of pilot measurements on ecosystem ecology, meteorology, trace gases, and aerosols, *Atmos. Chem. Phys.*, 15, 10723-10776, doi: 10.5194/acp-15-10723-2015, 2015.
- Angelini, M. M., Garrard, R. J., Rosen, S. J., and Hinrichs, R. Z.: Heterogeneous Reactions of Gaseous HNO₃ and NO₂ on the Clay Minerals Kaolinite and Pyrophyllite, *The Journal of Physical Chemistry A*, 111, 3326-3335, doi: 10.1021/jp0672656, 2007.
- Anttila, T., Kiendler-Scharr, A., Tillmann, R., and Mentel, T. F.: On the Reactive Uptake of Gaseous Compounds by Organic-Coated Aqueous Aerosols: Theoretical Analysis and Application to the Heterogeneous Hydrolysis of N₂O₅, *The Journal of Physical Chemistry A*, 110, 10435-10443, doi: 10.1021/jp062403c, 2006.
- Arens, F., Gutzwiller, L., Baltensperger, U., Gäggeler, H. W., and Ammann, M.: Heterogeneous Reaction of NO₂ on Diesel Soot Particles, *Environmental Science & Technology*, 35, 2191-2199, doi: 10.1021/es000207s, 2001.
- Ashmore M, R.: Assessing the future global impacts of ozone on vegetation, *Plant, Cell & Environment*, 28, 949-964, doi: 10.1111/j.1365-3040.2005.01341.x, 2005.
- Badger, C. L., Griffiths, P. T., George, I., Abbatt, J. P. D., and Cox, R. A.: Reactive Uptake of N₂O₅ by Aerosol Particles Containing Mixtures of Humic Acid and Ammonium Sulfate, *The Journal of Physical Chemistry A*, 110, 6986-6994, doi: 10.1021/jp0562678, 2006.
- Bauer, S. E., Balkanski, Y., Schulz, M., Hauglustaine, D. A., and Dentener, F.: Global modeling of heterogeneous chemistry on mineral aerosol surfaces: Influence on tropospheric ozone chemistry and comparison to observations, *Journal of Geophysical Research: Atmospheres*, 109, n/a-n/a, doi: 10.1029/2003JD003868, 2004.
- Beck, I., Jochner, S., Gilles, S., McIntyre, M., Buters, J. T. M., Schmidt-Weber, C., Behrendt, H., Ring, J., Menzel, A., and Traidl-Hoffmann, C.: High Environmental Ozone Levels Lead to Enhanced Allergenicity of Birch Pollen, *PLOS ONE*, 8, e80147, doi: 10.1371/journal.pone.0080147, 2013.
- Behnke, W., George, C., Scheer, V., and Zetzsch, C.: Production and decay of ClNO₂ from the reaction of gaseous N₂O₅ with NaCl solution: Bulk and aerosol experiments, *Journal of Geophysical Research: Atmospheres*, 102, 3795-3804, doi: 10.1029/96JD03057, 1997.

- Berkemeier, T., Steimer, S. S., Krieger, U. K., Peter, T., Poschl, U., Ammann, M., and Shiraiwa, M.: Ozone uptake on glassy, semi-solid and liquid organic matter and the role of reactive oxygen intermediates in atmospheric aerosol chemistry, *Physical Chemistry Chemical Physics*, 18, 12662-12674, doi: 10.1039/C6CP00634E, 2016.
- Börensén, C., Kirchner, U., Scheer, V., Vogt, R., and Zellner, R.: Mechanism and Kinetics of the Reactions of NO₂ or HNO₃ with Alumina as a Mineral Dust Model Compound, *The Journal of Physical Chemistry A*, 104, 5036-5045, doi: 10.1021/jp994170d, 2000.
- Burkholder, J. B., Abbatt, J. P. D., Huie, R. E., Kurylo, M. J., Wilmoth, D. M., Sander, S. P., Barker, J. R., Kolb, C. E., Orkin, V. L., and Wine, P. H.: Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 18, JPL Publication 15-10, 2015.
- Chang, R. Y. W., Sullivan, R. C., and Abbatt, J. P. D.: Initial uptake of ozone on Saharan dust at atmospheric relative humidities, *Geophys. Res. Lett.*, 32, doi: 10.1029/2005GL023317, 2005.
- Chen, Y., Wolke, R., Ran, L., Birmili, W., Spindler, G., Schröder, W., Su, H., Cheng, Y., Tegen, I., and Wiedensohler, A.: A parameterization of the heterogeneous hydrolysis of N₂O₅ for mass-based aerosol models: improvement of particulate nitrate prediction, *Atmos. Chem. Phys.*, 18, 673-689, doi: 10.5194/acp-18-673-2018, 2018.
- Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H., Zeng, L. M., Liu, S., Gnauk, T., Brüggemann, E., and He, L. Y.: Mixing state of elemental carbon and non-light-absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, *Journal of Geophysical Research: Atmospheres*, 111, doi: 10.1029/2005JD006929, 2006.
- Cheng, Y., Su, H., Koop, T., Mikhailov, E., and Pöschl, U.: Size dependence of phase transitions in aerosol nanoparticles, *Nature Communications*, 6, 5923, doi: 10.1038/ncomms6923, 2015.
- Cheng, Y. F., Su, H., Rose, D., Gunthe, S. S., Berghof, M., Wehner, B., Achtert, P., Nowak, A., Takegawa, N., Kondo, Y., Shiraiwa, M., Gong, Y. G., Shao, M., Hu, M., Zhu, T., Zhang, Y. H., Carmichael, G. R., Wiedensohler, A., Andreae, M. O., and Pöschl, U.: Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization, *Atmos. Chem. Phys.*, 12, 4477-4491, doi: 10.5194/acp-12-4477-2012, 2012.
- Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Pöschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, *Science Advances*, 2, e1601530, doi: 10.1126/sciadv.1601530, 2016.
- Crowley, J. N., Ammann, M., Cox, R. A., Hynes, R. G., Jenkin, M. E., Mellouki, A., Rossi, M. J., Troe, J., and Wallington, T. J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume V – heterogeneous reactions on solid substrates, *Atmos. Chem. Phys.*, 10, 9059-9223, doi: 10.5194/acp-10-9059-2010, 2010.
- Crowley, J. N., Ammann, M., Cox, R. A., Hynes, R. G., Jenkin, M. E., Mellouki, A., Rossi, M. J., Troe, J., and Wallington, T. J.: Corrigendum to "Evaluated kinetic and photochemical data for atmospheric chemistry: Volume V –

- heterogeneous reactions on solid substrates" published in *Atmos. Chem. Phys.* 10, 9059–9223, 2010, *Atmos. Chem. Phys.*, 13, 7359–7359, doi: 10.5194/acp-13-7359-2013, 2013.
- de Gouw, J. A., and Lovejoy, E. R.: Reactive uptake of ozone by liquid organic compounds, *Geophys. Res. Lett.*, 25, 931–934, doi: 10.1029/98GL00515, 1998.
- 5 de Haan, D. O., and Finlayson-Pitts, B. J.: Knudsen Cell Studies of the Reaction of Gaseous Nitric Acid with Synthetic Sea Salt at 298 K, *The Journal of Physical Chemistry A*, 101, 9993–9999, doi: 10.1021/jp972450s, 1997.
- Dentener, F. J., Carmichael, G. R., Zhang, Y., Lelieveld, J., and Crutzen, P. J.: Role of mineral aerosol as a reactive surface in the global troposphere, *Journal of Geophysical Research: Atmospheres*, 101, 22869–22889, doi: 10.1029/96JD01818, 1996.
- 10 Disselkamp, R. S., Carpenter, M. A., Cowin, J. P., Berkowitz, C. M., Chapman, E. G., Zaveri, R. A., and Laulainen, N. S.: Ozone loss in soot aerosols, *Journal of Geophysical Research: Atmospheres*, 105, 9767–9771, doi:10.1029/1999JD901189, 2000.
- Ditas, J., Ma, N., Zhang, Y., Assmann, D., Neumaier, M., Riede, H., Karu, E., Williams, J., Scharffe, D., Wang, Q., Saturno, J., Schwarz, J. P., Katich, J. M., McMeeking, G. R., Zahn, A., Hermann, M., Brenninkmeijer, C. A. M., Andreae, M. O.,
- 15 Pöschl, U., Su, H., and Cheng, Y.: Strong impact of wildfires on the abundance and aging of black carbon in the lowermost stratosphere, *Proceedings of the National Academy of Sciences*, 115, E11595, doi: 10.1073/pnas.1806868115, 2018.
- Dupart, Y., Fine, L., D’Anna, B., and George, C.: Heterogeneous uptake of NO₂ on Arizona Test Dust under UV-A irradiation: An aerosol flow tube study, *Aeolian Research*, 15, 45–51, doi: <https://doi.org/10.1016/j.aeolia.2013.10.001>,
- 20 2014.
- El Zein, A., Romanias, M. N., and Bedjanian, Y.: Heterogeneous Interaction of H₂O₂ with Arizona Test Dust, *The Journal of Physical Chemistry A*, 118, 441–448, doi: 10.1021/jp409946j, 2014.
- Evans, M. J., and Jacob, D. J.: Impact of new laboratory studies of N₂O₅ hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and OH, *Geophys. Res. Lett.*, 32, doi: 10.1029/2005GL022469, 2005.
- 25 Fenidel, W., Matter, D., Burtscher, H., and Schmidt-Ott, A.: Interaction between carbon or iron aerosol particles and ozone, *Atmospheric Environment*, 29, 967–973, doi: [https://doi.org/10.1016/1352-2310\(95\)00038-Z](https://doi.org/10.1016/1352-2310(95)00038-Z), 1995.
- Fenter, F. F., Caloz, F., and Rossi, M. J.: Experimental evidence for the efficient “dry deposition” of nitric acid on calcite, *Atmospheric Environment*, 29, 3365–3372, doi: [https://doi.org/10.1016/1352-2310\(95\)00183-Y](https://doi.org/10.1016/1352-2310(95)00183-Y), 1995.
- Folkers, M., Mentel, T. F., and Wahner, A.: Influence of an organic coating on the reactivity of aqueous aerosols probed by
- 30 the heterogeneous hydrolysis of N₂O₅, *Geophys. Res. Lett.*, 30, doi: 10.1029/2003GL017168, 2003.
- Fowler, D., Pilegaard, K., Sutton, M. A., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., Schjoerring, J. K., Granier, C., Neftel, A., Isaksen, I. S. A., Laj, P., Maione, M., Monks, P. S., Burkhardt, J., Daemmgen,

- U., Neiryneck, J., Personne, E., Wichink-Kruit, R., Butterbach-Bahl, K., Flechard, C., Tuovinen, J. P., Coyle, M., Gerosa, G., Loubet, B., Altimir, N., Gruenhage, L., Ammann, C., Cieslik, S., Paoletti, E., Mikkelsen, T. N., Ro-Poulsen, H., Cellier, P., Cape, J. N., Horváth, L., Loreto, F., Niinemets, Ü., Palmer, P. I., Rinne, J., Misztal, P., Nemitz, E., Nilsson, D., Pryor, S., Gallagher, M. W., Vesala, T., Skiba, U., Brüggemann, N., Zechmeister-Boltenstern, S., Williams, J., O'Dowd, C., Facchini, M. C., de Leeuw, G., Flossman, A., Chaumerliac, N., and Erisman, J. W.: Atmospheric composition change: Ecosystems–Atmosphere interactions, *Atmospheric Environment*, 43, 5193-5267, doi: <http://dx.doi.org/10.1016/j.atmosenv.2009.07.068>, 2009.
- Fried, A., Crawford, J., Olson, J., Walega, J., Potter, W., Wert, B., Jordan, C., Anderson, B., Shetter, R., Lefer, B., Blake, D., Blake, N., Meinardi, S., Heikes, B., O'Sullivan, D., Snow, J., Fuelberg, H., Kiley, C. M., Sandholm, S., Tan, D., Sachse, G., Singh, H., Faloon, I., Harward, C. N., and Carmichael, G. R.: Airborne tunable diode laser measurements of formaldehyde during TRACE-P: Distributions and box model comparisons, *Journal of Geophysical Research: Atmospheres*, 108, doi: 10.1029/2003JD003451, 2003.
- Frinak, E. K., Wermeille, S. J., Mashburn, C. D., Tolbert, M. A., and Pursell, C. J.: Heterogeneous Reaction of Gaseous Nitric Acid on γ -Phase Iron(III) Oxide, *The Journal of Physical Chemistry A*, 108, 1560-1566, doi: 10.1021/jp030807o, 2004.
- Fröhlich-Nowoisky, J., Kampf, C. J., Weber, B., Huffman, J. A., Pöhlker, C., Andreae, M. O., Lang-Yona, N., Burrows, S. M., Gunthe, S. S., Elbert, W., Su, H., Hoor, P., Thines, E., Hoffmann, T., Després, V. R., and Pöschl, U.: Bioaerosols in the Earth system: Climate, health, and ecosystem interactions, *Atmospheric Research*, 182, 346-376, doi: <https://doi.org/10.1016/j.atmosres.2016.07.018>, 2016.
- Ganzeveld, L., and Lelieveld, J.: Dry deposition parameterization in a chemistry general circulation model and its influence on the distribution of reactive trace gases, *Journal of Geophysical Research: Atmospheres*, 100, 20999-21012, doi: 10.1029/95JD02266, 1995.
- Gard, E. E., Kleeman, M. J., Gross, D. S., Hughes, L. S., Allen, J. O., Morrical, B. D., Fergenson, D. P., Dienes, T., E. Gälli, M., Johnson, R. J., Cass, G. R., and Prather, K. A.: Direct Observation of Heterogeneous Chemistry in the Atmosphere, *Science*, 279, 1184, 1998.
- Gardner, J. A., Watson, L. R., Adewuyi, Y. G., Davidovits, P., Zahniser, M. S., Worsnop, D. R., and Kolb, C. E.: Measurement of the mass accommodation coefficient of SO₂ (g) on water droplets, *Journal of Geophysical Research: Atmospheres*, 92, 10887-10895, doi: 10.1029/JD092iD09p10887, 1987.
- Gebel, M. E., Finlayson-Pitts Barbara, J., and Ganske Jane, A.: The uptake of SO₂ on synthetic sea salt and some of its components, *Geophys. Res. Lett.*, 27, 887-890, doi: 10.1029/1999GL011152, 2000.
- George, C., Ammann, M., D'Anna, B., Donaldson, D. J., and Nizkorodov, S. A.: Heterogeneous Photochemistry in the Atmosphere, *Chemical Reviews*, 115, 4218-4258, doi: 10.1021/cr500648z, 2015.

- George, C., Ponche, J. L., Mirabel, P., Behnke, W., Scheer, V., and Zetzsch, C.: Study of the Uptake of N₂O₅ by Water and NaCl Solutions, *The Journal of Physical Chemistry*, 98, 8780-8784, doi: 10.1021/j100086a031, 1994.
- Ghan, S. J., Guzman, G., and Abdul-Razzak, H.: Competition between Sea Salt and Sulfate Particles as Cloud Condensation Nuclei, *Journal of the Atmospheric Sciences*, 55, 3340-3347, doi: 10.1175/1520-0469(1998)055<3340:CBSSAS>2.0.CO;2, 1998.
- Goodman, A. L., Li, P., Usher, C. R., and Grassian, V. H.: Heterogeneous Uptake of Sulfur Dioxide On Aluminum and Magnesium Oxide Particles, *The Journal of Physical Chemistry A*, 105, 6109-6120, doi: 10.1021/jp004423z, 2001.
- Goodman, A. L., Underwood, G. M., and Grassian, V. H.: A laboratory study of the heterogeneous reaction of nitric acid on calcium carbonate particles, *Journal of Geophysical Research: Atmospheres*, 105, 29053-29064, doi: 10.1029/2000JD900396, 2000.
- Griffiths, P. T., Badger, C. L., Cox, R. A., Folkers, M., Henk, H. H., and Mentel, T. F.: Reactive Uptake of N₂O₅ by Aerosols Containing Dicarboxylic Acids. Effect of Particle Phase, Composition, and Nitrate Content, *The Journal of Physical Chemistry A*, 113, 5082-5090, doi: 10.1021/jp8096814, 2009.
- Gross, S., Iannone, R., Xiao, S., and Bertram, A. K.: Reactive uptake studies of NO₃ and N₂O₅ on alkenoic acid, alkanolate, and polyalcohol substrates to probe nighttime aerosol chemistry, *Physical Chemistry Chemical Physics*, 11, 7792-7803, doi: 10.1039/B904741G, 2009.
- Gustafsson, R. J., Orlov, A., Griffiths, P. T., Cox, R. A., and Lambert, R. M.: Reduction of NO₂ to nitrous acid on illuminated titanium dioxide aerosol surfaces: implications for photocatalysis and atmospheric chemistry, *Chemical Communications*, 3936-3938, doi: 10.1039/B609005B, 2006.
- Guimbaud, C., Arens, F., Gutzwiller, L., Gäggeler, H. W., and Ammann, M.: Uptake of HNO₃ to deliquescent sea-salt particles: a study using the short-lived radioactive isotope tracer ¹³N, *Atmos. Chem. Phys.*, 2, 249-257, doi: 10.5194/acp-2-249-2002, 2002.
- Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M., Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, *Atmos. Chem. Phys.*, 11, 11023-11039, doi: 10.5194/acp-11-11023-2011, 2011.
- Gutzwiller, L., George, C., Rössler, E., and Ammann, M.: Reaction Kinetics of NO₂ with Resorcinol and 2,7-Naphthalenediol in the Aqueous Phase at Different pH, *The Journal of Physical Chemistry A*, 106, 12045-12050, doi: 10.1021/jp026240d, 2002.
- Hallquist, M., Stewart, D. J., Stephenson, S. K., and Anthony Cox, R.: Hydrolysis of N₂O₅ on sub-micron sulfate aerosols, *Physical Chemistry Chemical Physics*, 5, 3453-3463, doi: 10.1039/B301827J, 2003.

- Hanisch, F., and Crowley, J. N.: Heterogeneous Reactivity of Gaseous Nitric Acid on Al_2O_3 , CaCO_3 , and Atmospheric Dust Samples: A Knudsen Cell Study, *The Journal of Physical Chemistry A*, 105, 3096-3106, doi: 10.1021/jp001254+, 2001a.
- 5 Hanisch, F., and Crowley, J. N.: The heterogeneous reactivity of gaseous nitric acid on authentic mineral dust samples, and on individual mineral and clay mineral components, *Physical Chemistry Chemical Physics*, 3, 2474-2482, doi: 10.1039/B101700O, 2001b.
- Hanisch, F., and Crowley, J. N.: Ozone decomposition on Saharan dust: an experimental investigation, *Atmos. Chem. Phys.*, 3, 119-130, doi: 10.5194/acp-3-119-2003, 2003.
- 10 Harrison, R. M., and Collins, G. M.: Measurements of Reaction Coefficients of NO_2 and HONO on Aerosol Particles, *Journal of Atmospheric Chemistry*, 30, 397-406, doi: 10.1023/A:1006094304069, 1998.
- He, H., Wang, Y., Ma, Q., Ma, J., Chu, B., Ji, D., Tang, G., Liu, C., Zhang, H., and Hao, J.: Mineral dust and NO_x promote the conversion of SO_2 to sulfate in heavy pollution days, *Scientific Reports*, 4, 4172, doi: 10.1038/srep04172, 2014.
- Hearn, J. D., Lovett, A. J., and Smith, G. D.: Ozonolysis of oleic acid particles: evidence for a surface reaction and secondary reactions involving Criegee intermediates, *Physical Chemistry Chemical Physics*, 7, 501-511, doi: 15 10.1039/B414472D, 2005.
- Herrmann, H.: Kinetics of Aqueous Phase Reactions Relevant for Atmospheric Chemistry, *Chemical Reviews*, 103, 4691-4716, doi: 10.1021/cr020658q, 2003.
- Herrmann, H., Schaefer, T., Tilgner, A., Styler, S. A., Weller, C., Teich, M., and Otto, T.: Tropospheric Aqueous-Phase Chemistry: Kinetics, Mechanisms, and Its Coupling to a Changing Gas Phase, *Chemical Reviews*, 115, 4259-4334, doi: 20 10.1021/cr500447k, 2015.
- Hoffman, R. C., Kaleuati, M. A., and Finlayson-Pitts, B. J.: Knudsen Cell Studies of the Reaction of Gaseous HNO_3 with NaCl Using Less than a Single Layer of Particles at 298 K: A Modified Mechanism, *The Journal of Physical Chemistry A*, 107, 7818-7826, doi: 10.1021/jp030611o, 2003.
- 25 Held, A., Zerrath, A., McKeon, U., Fehrenbach, T., Niessner, R., Plass-Dülmer, C., Kaminski, U., Berresheim, H., and Pöschl, U.: Aerosol size distributions measured in urban, rural and high-alpine air with an electrical low pressure impactor (ELPI), *Atmospheric Environment*, 42, 8502-8512, doi: <https://doi.org/10.1016/j.atmosenv.2008.06.015>, 2008.
- Huang, L., Zhao, Y., Li, H., and Chen, Z.: Kinetics of Heterogeneous Reaction of Sulfur Dioxide on Authentic Mineral Dust: Effects of Relative Humidity and Hydrogen Peroxide, *Environmental Science & Technology*, 49, 10797-10805, doi: 10.1021/acs.est.5b03930, 2015.
- 30 Huss, A., Lim, P. K., and Eckert, C. A.: Oxidation of aqueous sulfur dioxide. 1. Homogeneous manganese(II) and iron(II) catalysis at low pH, *The Journal of Physical Chemistry*, 86, 4224-4228, doi: 10.1021/j100218a027, 1982.

- Il'in, S. D.: Study of heterogeneous ozone loss on materials typical of atmospheric aerosol species, *Sov. J. Chem. Phys.*, 8, 1858-1880, 1991.
- Jacob, D. J.: Heterogeneous chemistry and tropospheric ozone, *Atmospheric Environment*, 34, 2131-2159, doi: [https://doi.org/10.1016/S1352-2310\(99\)00462-8](https://doi.org/10.1016/S1352-2310(99)00462-8), 2000.
- 5 Jayne, J. T., Gardner, J. A., Davidovits, P., Worsnop, D. R., Zahniser, M. S., and Kolb, C. E.: The effect of H₂O₂ content on the uptake of SO₂(g) by aqueous droplets, *Journal of Geophysical Research: Atmospheres*, 95, 20559-20563, doi: 10.1029/JD095iD12p20559, 1990.
- Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimo, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525, doi: 10.1126/science.1180353, 2009.
- 10 Johnson, E. R., Sciegienka, J., Carlos-Cuellar, S., and Grassian, V. H.: Heterogeneous Uptake of Gaseous Nitric Acid on Dolomite (CaMg(CO₃)₂) and Calcite (CaCO₃) Particles: A Knudsen Cell Study Using Multiple, Single, and Fractional Particle Layers, *The Journal of Physical Chemistry A*, 109, 6901-6911, doi: 10.1021/jp0516285, 2005.
- 20 Judeikis, H. S., Stewart, T. B., and Wren, A. G.: Laboratory studies of heterogeneous reactions of SO₂, *Atmospheric Environment* (1967), 12, 1633-1641, doi: [https://doi.org/10.1016/0004-6981\(78\)90312-8](https://doi.org/10.1016/0004-6981(78)90312-8), 1978.
- Kalberer, M., Ammann, M., Arens, F., Gäggeler, H. W., and Baltensperger, U.: Heterogeneous formation of nitrous acid (HONO) on soot aerosol particles, *Journal of Geophysical Research: Atmospheres*, 104, 13825-13832, doi: 10.1029/1999JD900141, 1999.
- 25 Kalberer, M., Tabor, K., Ammann, M., Parrat, Y., Weingartner, E., Piguet, D., Rössler, E., Jost, D. T., Türlér, A., Gäggeler, H. W., and Baltensperger, U.: Heterogeneous Chemical Processing of ¹³NO₂ by Monodisperse Carbon Aerosols at Very Low Concentrations, *The Journal of Physical Chemistry*, 100, 15487-15493, doi: 10.1021/jp9606974, 1996.
- Kamm, S., Möhler, O., Naumann, K. H., Saathoff, H., and Schurath, U.: The heterogeneous reaction of ozone with soot aerosol, *Atmospheric Environment*, 33, 4651-4661, doi: [https://doi.org/10.1016/S1352-2310\(99\)00235-6](https://doi.org/10.1016/S1352-2310(99)00235-6), 1999.
- 30 Kane, S. M., Caloz, F., and Leu, M.-T.: Heterogeneous Uptake of Gaseous N₂O₅ by (NH₄)₂SO₄, NH₄HSO₄, and H₂SO₄ Aerosols, *The Journal of Physical Chemistry A*, 105, 6465-6470, doi: 10.1021/jp010490x, 2001.

- Karagulian, F., and Rossi, M. J.: Heterogeneous Chemistry of the NO₃ Free Radical and N₂O₅ on Decane Flame Soot at Ambient Temperature: Reaction Products and Kinetics, *The Journal of Physical Chemistry A*, 111, 1914-1926, doi: 10.1021/jp0670891, 2007.
- Karagulian, F., Santschi, C., and Rossi, M. J.: The heterogeneous chemical kinetics of N₂O₅ on CaCO₃ and other atmospheric mineral dust surrogates, *Atmos. Chem. Phys.*, 6, 1373-1388, doi: 10.5194/acp-6-1373-2006, 2006.
- King, M. D., Thompson, K. C., Ward, A. D., Pfrang, C., and Hughes, B. R.: Oxidation of biogenic and water-soluble compounds in aqueous and organic aerosol droplets by ozone: a kinetic and product analysis approach using laser Raman tweezers, *Faraday Discussions*, 137, 173-192, doi: 10.1039/B702199B, 2008.
- Kirchner, U., Scheer, V., and Vogt, R.: FTIR Spectroscopic Investigation of the Mechanism and Kinetics of the Heterogeneous Reactions of NO₂ and HNO₃ with Soot, *The Journal of Physical Chemistry A*, 104, 8908-8915, doi: 10.1021/jp0005322, 2000.
- Kleffmann, J. r., Becker, K. H., Lackhoff, M., and Wiesen, P.: Heterogeneous conversion of NO₂ on carbonaceous surfaces, *Physical Chemistry chemical Physics*, 5443-5450, 1999.
- Kleffmann, J., Becker, K. H., and Wiesen, P.: Heterogeneous NO₂ conversion processes on acid surfaces: possible atmospheric implications, *Atmospheric Environment*, 32, 2721-2729, doi: [https://doi.org/10.1016/S1352-2310\(98\)00065-X](https://doi.org/10.1016/S1352-2310(98)00065-X), 1998.
- Knopf, D. A., Anthony, L. M., and Bertram, A. K.: Reactive Uptake of O₃ by Multicomponent and Multiphase Mixtures Containing Oleic Acid, *The Journal of Physical Chemistry A*, 109, 5579-5589, doi: 10.1021/jp0512513, 2005.
- Kolb, C. E., Worsnop, D. R., Zahniser, M. S., Davidovits, P., Keyser, L. F., Leu, M. T., Molina, M. J., Hanson, D. R., Ravishankara, A. R., Williams, L. R., and Tolbert, M. A.: LABORATORY STUDIES OF ATMOSPHERIC HETEROGENEOUS CHEMISTRY, in: *Progress and Problems in Atmospheric Chemistry*, Advanced Series in Physical Chemistry, Volume 3, WORLD SCIENTIFIC, 771-875, 1995.
- Kolb, C. E., Cox, R. A., Abbatt, J. P. D., Ammann, M., Davis, E. J., Donaldson, D. J., Garrett, B. C., George, C., Griffiths, P. T., Hanson, D. R., Kulmala, M., McFiggans, G., Pöschl, U., Riipinen, I., Rossi, M. J., Rudich, Y., Wagner, P. E., Winkler, P. M., Worsnop, D. R., and O' Dowd, C. D.: An overview of current issues in the uptake of atmospheric trace gases by aerosols and clouds, *Atmos. Chem. Phys.*, 10, 10561-10605, doi: 10.5194/acp-10-10561-2010, 2010.
- Komiyama, H., and Inoue, H.: 20 Absorption of nitrogen oxides into water, *Chemical Engineering Science*, 35, 154-161, doi: [https://doi.org/10.1016/0009-2509\(80\)80082-0](https://doi.org/10.1016/0009-2509(80)80082-0), 1980.
- Lappalainen, H. K., Kerminen, V. M., Petäjä, T., Kurten, T., Baklanov, A., Shvidenko, A., Bäck, J., Vihma, T., Alekseychik, P., Andreae, M. O., Arnold, S. R., Arshinov, M., Asmi, E., Belan, B., Bobylev, L., Chalov, S., Cheng, Y., Chubarova, N., de Leeuw, G., Ding, A., Dobrolyubov, S., Dubtsov, S., Dyukarev, E., Elansky, N., Eleftheriadis, K., Esau, I., Filatov, N., Flint, M., Fu, C., Glezer, O., Gliko, A., Heimann, M., Holtslag, A. A. M., Hörrak, U., Janhunen, J., Juhola, S., Järvi,

- L., Järvinen, H., Kanukhina, A., Konstantinov, P., Kotlyakov, V., Kieloaho, A. J., Komarov, A. S., Kujansuu, J., Kukkonen, I., Duplissy, E. M., Laaksonen, A., Laurila, T., Lihavainen, H., Lisitzin, A., Mahura, A., Makshtas, A., Mareev, E., Mazon, S., Matishov, D., Melnikov, V., Mikhailov, E., Moiseev, D., Nigmatulin, R., Noe, S. M., Ojala, A., Pihlatie, M., Popovicheva, O., Pumpanen, J., Regerand, T., Repina, I., Shcherbinin, A., Shevchenko, V., Sipilä, M.,
5 Skorokhod, A., Spracklen, D. V., Su, H., Subetto, D. A., Sun, J., Terzhevik, A. Y., Timofeyev, Y., Troitskaya, Y., Tynkkynen, V. P., Kharuk, V. I., Zaytseva, N., Zhang, J., Viisanen, Y., Vesala, T., Hari, P., Hansson, H. C., Matvienko, G. G., Kasimov, N. S., Guo, H., Bondur, V., Zilitinkevich, S., and Kulmala, M.: Pan-Eurasian Experiment (PEEX): towards a holistic understanding of the feedbacks and interactions in the land–atmosphere–ocean–society continuum in the northern Eurasian region, *Atmos. Chem. Phys.*, 16, 14421-14461, doi: 10.5194/acp-16-14421-2016, 2016.
- 10 Laskin, A., Gaspar, D. J., Wang, W., Hunt, S. W., Cowin, J. P., Colson, S. D., and Finlayson-Pitts, B. J.: Reactions at Interfaces As a Source of Sulfate Formation in Sea-Salt Particles, *Science*, 301, 340, doi: 10.1126/science.1085374, 2003.
- Lee, J. H., and Tang, I. N.: Accommodation coefficient of gaseous NO₂ on water surfaces, *Atmospheric Environment* (1967), 22, 1147-1151, doi: [https://doi.org/10.1016/0004-6981\(88\)90344-7](https://doi.org/10.1016/0004-6981(88)90344-7), 1988.
- 15 Lee, Y. H., Chen, K., and Adams, P. J.: Development of a global model of mineral dust aerosol microphysics, *Atmos. Chem. Phys.*, 9, 2441-2458, doi: 10.5194/acp-9-2441-2009, 2009.
- Lelieveld, J., and Dentener, F. J.: What controls tropospheric ozone?, *Journal of Geophysical Research: Atmospheres*, 105, 3531-3551, doi: 10.1029/1999JD901011, 2000.
- Lelièvre, S., Bedjanian, Y., Laverdet, G., and Le Bras, G.: Heterogeneous Reaction of NO₂ with Hydrocarbon Flame Soot,
20 *The Journal of Physical Chemistry A*, 108, 10807-10817, doi: 10.1021/jp0469970, 2004a.
- Lelièvre, S., Bedjanian, Y., Pouvesle, N., Delfau, J.-L., Vovelle, C., and Le Bras, G.: Heterogeneous reaction of ozone with hydrocarbon flame soot, *Physical Chemistry Chemical Physics*, 6, 1181-1191, doi: 10.1039/B316895F, 2004b.
- Lewis, E. R., and Schwartz, S. E.: *Sea salt Aerosol Production: Mechanisms, Methods, Measurements, and Models*, American Geophysical Union, Washington, DC, 2004.
- 25 Li, G., Cheng, Y., Kuhn, U., Xu, R., Yang, Y., Meusel, H., Wang, Z., Ma, N., Wu, Y., Li, M., Williams, J., Hoffmann, T., Ammann, M., Pöschl, U., Shao, M., and Su, H.: Physicochemical uptake and release of volatile organic compounds by soil in coated-wall flow tube experiments with ambient air, *Atmos. Chem. Phys.*, 19, 2209-2232, doi: 10.5194/acp-19-2209-2019, 2019.
- Li, G., Su, H., Li, X., Kuhn, U., Meusel, H., Hoffmann, T., Ammann, M., Pöschl, U., Shao, M., and Cheng, Y.: Uptake of
30 gaseous formaldehyde by soil surfaces: a combination of adsorption/desorption equilibrium and chemical reactions, *Atmos. Chem. Phys.*, 16, 10299-10311, doi: 10.5194/acp-16-10299-2016, 2016.

- Li, H., Chen, Z., Huang, L., and Huang, D.: Organic peroxides' gas-particle partitioning and rapid heterogeneous decomposition on secondary organic aerosol, *Atmos. Chem. Phys.*, 16, 1837-1848, doi: 10.5194/acp-16-1837-2016, 2016.
- Li, H. J., Zhu, T., Zhao, D. F., Zhang, Z. F., and Chen, Z. M.: Kinetics and mechanisms of heterogeneous reaction of NO₂ on CaCO₃ surfaces under dry and wet conditions, *Atmos. Chem. Phys.*, 10, 463-474, doi: 10.5194/acp-10-463-2010, 2010.
- Li, L., Chen, Z. M., Zhang, Y. H., Zhu, T., Li, J. L., and Ding, J.: Kinetics and mechanism of heterogeneous oxidation of sulfur dioxide by ozone on surface of calcium carbonate, *Atmos. Chem. Phys.*, 6, 2453-2464, doi: 10.5194/acp-6-2453-2006, 2006.
- Li, X., Rohrer, F., Brauers, T., Hofzumahaus, A., Lu, K., Shao, M., Zhang, Y. H., and Wahner, A.: Modeling of HCHO and CHOCHO at a semi-rural site in southern China during the PRIDE-PRD2006 campaign, *Atmos. Chem. Phys.*, 14, 12291-12305, doi: 10.5194/acp-14-12291-2014, 2014.
- Liu, F., Lakey, P. S. J., Berkemeier, T., Tong, H., Kunert, A. T., Meusel, H., Cheng, Y., Su, H., Fröhlich-Nowoisky, J., Lai, S., Weller, M. G., Shiraiwa, M., Pöschl, U., and Kampf, C. J.: Atmospheric protein chemistry influenced by anthropogenic air pollutants: nitration and oligomerization upon exposure to ozone and nitrogen dioxide, *Faraday Discussions*, 200, 413-427, doi: 10.1039/C7FD00005G, 2017.
- Liu, Y., Cain, J. P., Wang, H., and Laskin, A.: Kinetic Study of Heterogeneous Reaction of Deliquesced NaCl Particles with Gaseous HNO₃ Using Particle-on-Substrate Stagnation Flow Reactor Approach, *The Journal of Physical Chemistry A*, 111, 10026-10043, doi: 10.1021/jp072005p, 2007.
- Liu, Y., Gibson, Cain, Wang, H., Grassian, and Laskin, A.: Kinetics of Heterogeneous Reaction of CaCO₃ Particles with Gaseous HNO₃ over a Wide Range of Humidity, *The Journal of Physical Chemistry A*, 112, 1561-1571, doi: 10.1021/jp076169h, 2008.
- Liao, H., and Seinfeld, J. H.: Global impacts of gas-phase chemistry-aerosol interactions on direct radiative forcing by anthropogenic aerosols and ozone, *Journal of Geophysical Research: Atmospheres*, 110, doi: 10.1029/2005JD005907, 2005.
- Longfellow, C. A., Ravishankara, A. R., and Hanson, D. R.: Reactive and nonreactive uptake on hydrocarbon soot: HNO₃, O₃, and N₂O₅, *Journal of Geophysical Research: Atmospheres*, 105, 24345-24350, doi: 10.1029/2000JD900297, 2000.
- Ma, N., and Birmili, W.: Estimating the contribution of photochemical particle formation to ultrafine particle number averages in an urban atmosphere, *Science of The Total Environment*, 512-513, 154-166, doi: <https://doi.org/10.1016/j.scitotenv.2015.01.009>, 2015.

- Ma, N., Birmili, W., Müller, T., Tuch, T., Cheng, Y. F., Xu, W. Y., Zhao, C. S., and Wiedensohler, A.: Tropospheric aerosol scattering and absorption over central Europe: a closure study for the dry particle state, *Atmos. Chem. Phys.*, 14, 6241-6259, doi: 10.5194/acp-14-6241-2014, 2014.
- McNeill, V. F.: Aqueous Organic Chemistry in the Atmosphere: Sources and Chemical Processing of Organic Aerosols, *Environmental Science & Technology*, 49, 1237-1244, doi: 10.1021/es5043707, 2015.
- McNaughton, C. S., Clarke, A. D., Kapustin, V., Shinozuka, Y., Howell, S. G., Anderson, B. E., Winstead, E., Dibb, J., Scheuer, E., Cohen, R. C., Wooldridge, P., Perring, A., Huey, L. G., Kim, S., Jimenez, J. L., Dunlea, E. J., DeCarlo, P. F., Wennberg, P. O., Crounse, J. D., Weinheimer, A. J., and Flocke, F.: Observations of heterogeneous reactions between Asian pollution and mineral dust over the Eastern North Pacific during INTEX-B, *Atmos. Chem. Phys.*, 9, 8283-8308, doi: 10.5194/acp-9-8283-2009, 2009.
- Meusel, H., Elshorbany, Y., Kuhn, U., Bartels-Rausch, T., Reinmuth-Selzle, K., Kampf, C. J., Li, G., Wang, X., Lelieveld, J., Pöschl, U., Hoffmann, T., Su, H., Ammann, M., and Cheng, Y.: Light-induced protein nitration and degradation with HONO emission, *Atmos. Chem. Phys.*, 17, 11819-11833, doi: 10.5194/acp-17-11819-2017, 2017.
- Msibi, I. M., Shi, J. P., and Harrison, R. M.: Accommodation coefficient for trace gas uptake using deposition profile measurement in an annular reactor, *Journal of Atmospheric Chemistry*, 17, 339-351, doi: 10.1007/BF00696853, 1993.
- Mochida, M., Hirokawa, J., and Akimoto, H.: Unexpected large uptake of O₃ on sea salts and the observed Br₂ formation, *Geophys. Res. Lett.*, 27, 2629-2632, doi: 10.1029/1999GL010927, 2000.
- Mogili, P. K., Kleiber, P. D., Young, M. A., and Grassian, V. H.: Heterogeneous Uptake of Ozone on Reactive Components of Mineral Dust Aerosol: An Environmental Aerosol Reaction Chamber Study, *The Journal of Physical Chemistry A*, 110, 13799-13807, doi: 10.1021/jp063620g, 2006.
- Moise, T., and Rudich, Y.: Reactive uptake of ozone by proxies for organic aerosols: Surface versus bulk processes, *Journal of Geophysical Research: Atmospheres*, 105, 14667-14676, doi: 10.1029/2000JD900071, 2000.
- Morris, J. W., Davidovits, P., Jayne, J. T., Jimenez, J. L., Shi, Q., Kolb, C. E., Worsnop, D. R., Barney, W. S., and Cass, G.: Kinetics of submicron oleic acid aerosols with ozone: A novel aerosol mass spectrometric technique, *Geophys. Res. Lett.*, 29, 71-71-71-74, doi: 10.1029/2002GL014692, 2002.
- Ndour, M., D'Anna, B., George, C., Ka, O., Balkanski, Y., Kleffmann, J., Stemmler, K., and Ammann, M.: Photoenhanced uptake of NO₂ on mineral dust: Laboratory experiments and model simulations, *Geophys. Res. Lett.*, 35, doi: 10.1029/2007GL032006, 2008.
- Ndour, M., Nicolas, M., D'Anna, B., Ka, O., and George, C.: Photoreactivity of NO₂ on mineral dusts originating from different locations of the Sahara desert, *Physical Chemistry Chemical Physics*, 11, 1312-1319, doi: 10.1039/B806441E, 2009.

- Mendez, M., Visez, N., Gosselin, S., Crenn, V., Riffault, V., and Petitprez, D.: Reactive and Nonreactive Ozone Uptake during Aging of Oleic Acid Particles, *The Journal of Physical Chemistry A*, 118, 9471-9481, doi: 10.1021/jp503572c, 2014.
- Meusel, H., Tamm, A., Kuhn, U., Wu, D., Leifke, A. L., Fiedler, S., Ruckteschler, N., Yordanova, P., Lang-Yona, N., Pöhlker, M., Lelieveld, J., Hoffmann, T., Pöschl, U., Su, H., Weber, B., and Cheng, Y.: Emission of nitrous acid from soil and biological soil crusts represents an important source of HONO in the remote atmosphere in Cyprus, *Atmos. Chem. Phys.*, 18, 799-813, doi: 10.5194/acp-18-799-2018, 2018.
- Michel, A. E., Usher, C. R., and Grassian, V. H.: Heterogeneous and catalytic uptake of ozone on mineral oxides and dusts: A Knudsen cell investigation, *Geophys. Res. Lett.*, 29, 10-11-10-14, doi: 10.1029/2002GL014896, 2002.
- Michel, A. E., Usher, C. R., and Grassian, V. H.: Reactive uptake of ozone on mineral oxides and mineral dusts, *Atmospheric Environment*, 37, 3201-3211, doi: [https://doi.org/10.1016/S1352-2310\(03\)00319-4](https://doi.org/10.1016/S1352-2310(03)00319-4), 2003.
- Moise, T., and Rudich, Y.: Reactive uptake of ozone by proxies for organic aerosols: Surface versus bulk processes, *Journal of Geophysical Research: Atmospheres*, 105, 14667-14676, doi: 10.1029/2000JD900071, 2000.
- Mu, Q., Shiraiwa, M., Octaviani, M., Ma, N., Ding, A., Su, H., Lammel, G., Pöschl, U., and Cheng, Y.: Temperature effect on phase state and reactivity controls atmospheric multiphase chemistry and transport of PAHs, *Science Advances*, 4, 2018.
- Nicolas, M., Ndour, M., Ka, O., D'Anna, B., and George, C.: Photochemistry of Atmospheric Dust: Ozone Decomposition on Illuminated Titanium Dioxide, *Environ. Sci. & Technol.*, 43, 7437-7442, doi: 10.1021/es901569d, 2009.
- Ndour, M., D'Anna, B., George, C., Ka, O., Balkanski, Y., Kleffmann, J., Stemmler, K., and Ammann, M.: Photoenhanced uptake of NO₂ on mineral dust: Laboratory experiments and model simulations, *Geophys. Res. Lett.*, 35, doi: 10.1029/2007GL032006, 2008.
- Ndour, M., Nicolas, M., D'Anna, B., Ka, O., and George, C.: Photoreactivity of NO₂ on mineral dusts originating from different locations of the Sahara desert, *Physical Chemistry Chemical Physics*, 11, 1312-1319, doi: 10.1039/B806441E, 2009.
- O'Dowd, C. D., Smith, M. H., Consterdine, I. E., and Lowe, J. A.: Marine aerosol, sea-salt, and the marine sulphur cycle: a short review, *Atmospheric Environment*, 31, 73-80, doi: [https://doi.org/10.1016/S1352-2310\(96\)00106-9](https://doi.org/10.1016/S1352-2310(96)00106-9), 1997.
- Oswald, R., Behrendt, T., Ermel, M., Wu, D., Su, H., Cheng, Y., Breuninger, C., Moravek, A., Mougín, E., Delon, C., Loubet, B., Pommerening-Röser, A., Sörgel, M., Pöschl, U., Hoffmann, T., Andreae, M. O., Meixner, F. X., and Trebs, I.: HONO Emissions from Soil Bacteria as a Major Source of Atmospheric Reactive Nitrogen, *Science*, 341, 1233, doi: 10.1126/science.1242266, 2013.
- Ouyang, Y., Xu, Z., Fan, E., Li, Y., and Zhang, L.: Effect of nitrogen dioxide and sulfur dioxide on viability and morphology of oak pollen, *International Forum of Allergy & Rhinology*, 6, 95-100, doi: 10.1002/alr.21632, 2016.

- Pöhlker, C., Wiedemann, K. T., Sinha, B., Shiraiwa, M., Gunthe, S. S., Smith, M., Su, H., Artaxo, P., Chen, Q., Cheng, Y., Elbert, W., Gilles, M. K., Kilcoyne, A. L. D., Moffet, R. C., Weigand, M., Martin, S. T., Pöschl, U., and Andreae, M. O.: Biogenic Potassium Salt Particles as Seeds for Secondary Organic Aerosol in the Amazon, *Science*, 337, 1075, doi: 10.1126/science.1223264, 2012.
- 5 Pöschl, U., Letzel, T., Schauer, C., and Niessner, R.: Interaction of Ozone and Water Vapor with Spark Discharge Soot Aerosol Particles Coated with Benzo[a]pyrene: O₃ and H₂O Adsorption, Benzo[a]pyrene Degradation, and Atmospheric Implications, *The Journal of Physical Chemistry A*, 105, 4029-4041, doi: 10.1021/jp004137n, 2001.
- Pöschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P.,
- 10 Rose, D., Schneider, J., Su, H., Zorn, S. R., Artaxo, P., and Andreae, M. O.: Rainforest Aerosols as Biogenic Nuclei of Clouds and Precipitation in the Amazon, *Science*, 329, 1513, 2010.
- Pöschl, U., Rudich, Y., and Ammann, M.: Kinetic model framework for aerosol and cloud surface chemistry and gas-particle interactions – Part 1: General equations, parameters, and terminology, *Atmos. Chem. Phys.*, 7, 5989-6023, doi: 10.5194/acp-7-5989-2007, 2007.
- 15 Pöschl, U., and Shiraiwa, M.: Multiphase Chemistry at the Atmosphere–Biosphere Interface Influencing Climate and Public Health in the Anthropocene, *Chemical Reviews*, 115, 4440-4475, doi: 10.1021/cr500487s, 2015.
- Ponche, J. L., George, C., and Mirabel, P.: Mass transfer at the air/water interface: Mass accommodation coefficients of SO₂, HNO₃, NO₂ and NH₃, *Journal of Atmospheric Chemistry*, 16, 1-21, doi: 10.1007/BF00696620, 1993.
- Pradhan, M., Kalberer, M., Griffiths, P. T., Braban, C. F., Pope, F. D., Cox, R. A., and Lambert, R. M.: Uptake of Gaseous
- 20 Hydrogen Peroxide by Submicrometer Titanium Dioxide Aerosol as a Function of Relative Humidity, *Environmental Science & Technology*, 44, 1360-1365, doi: 10.1021/es902916f, 2010a.
- Pradhan, M., Kyriakou, G., Archibald, A. T., Papageorgiou, A. C., Kalberer, M., and Lambert, R. M.: Heterogeneous uptake of gaseous hydrogen peroxide by Gobi and Saharan dust aerosols: a potential missing sink for H₂O₂ in the troposphere, *Atmos. Chem. Phys.*, 10, 7127-7136, doi: 10.5194/acp-10-7127-2010, 2010b.
- 25 Prince, A. P., Wade, J. L., Grassian, V. H., Kleiber, P. D., and Young, M. A.: Heterogeneous reactions of soot aerosols with nitrogen dioxide and nitric acid: atmospheric chamber and Knudsen cell studies, *Atmospheric Environment*, 36, 5729-5740, doi: [https://doi.org/10.1016/S1352-2310\(02\)00626-X](https://doi.org/10.1016/S1352-2310(02)00626-X), 2002.
- Prospero, J. M.: Long-range transport of mineral dust in the global atmosphere: Impact of African dust on the environment of the southeastern United States, *Proceedings of the National Academy of Sciences*, 96, 3396, doi:
- 30 10.1073/pnas.96.7.3396, 1999.
- Quinn, P. K., Collins, D. B., Grassian, V. H., Prather, K. A., and Bates, T. S.: Chemistry and Related Properties of Freshly Emitted Sea Spray Aerosol, *Chemical Reviews*, 115, 4383-4399, doi: 10.1021/cr500713g, 2015.

- Ravishankara, A. R.: Heterogeneous and Multiphase Chemistry in the Troposphere, *Science*, 276, 1058, 1997.
- Rissler, J., Vestin, A., Swietlicki, E., Fisch, G., Zhou, J., Artaxo, P., and Andreae, M. O.: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, *Atmos. Chem. Phys.*, 6, 471-491, doi: 10.5194/acp-6-471-2006, 2006.
- 5 Rogaski, C. A., Golden, D. M., and Williams, L. R.: Reactive uptake and hydration experiments on amorphous carbon treated with NO₂, SO₂, O₃, HNO₃, and H₂SO₄, *Geophys. Res. Lett.*, 24, 381-384, doi: 10.1029/97GL00093, 1997.
- Romanias, M. N., El Zein, A., and Bedjanian, Y.: Heterogeneous Interaction of H₂O₂ with TiO₂ Surface under Dark and UV Light Irradiation Conditions, *The Journal of Physical Chemistry A*, 116, 8191-8200, doi: 10.1021/jp305366v, 2012.
- Romanias, M. N., El Zein, A., and Bedjanian, Y.: Uptake of hydrogen peroxide on the surface of Al₂O₃ and Fe₂O₃,
 10 *Atmospheric Environment*, 77, 1-8, doi: <https://doi.org/10.1016/j.atmosenv.2013.04.065>, 2013.
- Rudich, Y., Donahue, N. M., and Mentel, T. F.: Aging of Organic Aerosol: Bridging the Gap Between Laboratory and Field Studies, *Annual Review of Physical Chemistry*, 58, 321-352, doi: 10.1146/annurev.physchem.58.032806.104432, 2007.
- Saathoff, H., Naumann, K. H., Riemer, N., Kamm, S., Möhler, O., Schurath, U., Vogel, H., and Vogel, B.: The loss of NO₂, HNO₃, NO₃/N₂O₅, and HO₂/HOONO₂ on soot aerosol: A chamber and modeling study, *Geophys. Res. Lett.*, 28,
 15 1957-1960, doi:10.1029/2000GL012619, 2001.
- Sadanaga, Y., Hirokawa, J., and Akimoto, H.: Formation of molecular chlorine in dark condition: Heterogeneous reaction of ozone with sea salt in the presence of ferric ion, *Geophys. Res. Lett.*, 28, 4433-4436, doi: 10.1029/2001GL013722, 2001.
- Sage, A. M., Weitkamp, E. A., Robinson, A. L., and Donahue, N. M.: Reactivity of oleic acid in organic particles: changes in
 20 oxidant uptake and reaction stoichiometry with particle oxidation, *Physical Chemistry Chemical Physics*, 11, 7951-7962, doi: 10.1039/B904285G, 2009.
- Salgado-Muñoz, M. S., and Rossi, M. J.: Heterogeneous reactions of HNO₃ with flame soot generated under different combustion conditions. Reaction mechanism and kinetics, *Physical Chemistry Chemical Physics*, 4, 5110-5118, doi: 10.1039/B203912P, 2002.
- 25 Santschi, C., and Rossi, M. J.: Uptake of CO₂, SO₂, HNO₃ and HCl on Calcite (CaCO₃) at 300 K: Mechanism and the Role of Adsorbed Water, *The Journal of Physical Chemistry A*, 110, 6789-6802, doi: 10.1021/jp056312b, 2006.
- Saul, T. D., Tolocka, M. P., and Johnston, M. V.: Reactive Uptake of Nitric Acid onto Sodium Chloride Aerosols Across a Wide Range of Relative Humidities, *The Journal of Physical Chemistry A*, 110, 7614-7620, doi: 10.1021/jp060639a, 2006.
- 30 Schweitzer, F., Mirabel, P., and George, C.: Multiphase Chemistry of N₂O₅, ClNO₂, and BrNO₂, *The Journal of Physical Chemistry A*, 102, 3942-3952, doi: 10.1021/jp980748s, 1998.

- Schwartz, S. E.: Mass-Transport Considerations Pertinent to Aqueous Phase Reactions of Gases in Liquid-Water Clouds, Chemistry of Multiphase Atmospheric Systems, Berlin, Heidelberg, 1986, 415-471,
- Schwartz, S. E., and Freiberg, J. E.: Mass-transport limitation to the rate of reaction of gases in liquid droplets: Application to oxidation of SO₂ in aqueous solutions, Atmospheric Environment (1967), 15, 1129-1144, doi: [https://doi.org/10.1016/0004-6981\(81\)90303-6](https://doi.org/10.1016/0004-6981(81)90303-6), 1981.
- Schütze, M., and Herrmann, H.: Determination of phase transfer parameters for the uptake of HNO₃, N₂O₅ and O₃ on single aqueous drops, Physical Chemistry Chemical Physics, 4, 60-67, doi: 10.1039/B106078N, 2002.
- Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: From air pollution to climate change, John Wiley & Sons, New York, USA, 2006.
- Seisel, S., Böresen, C., Vogt, R., and Zellner, R.: Kinetics and mechanism of the uptake of N₂O₅ on mineral dust at 298 K, Atmos. Chem. Phys., 5, 3423-3432, doi: 10.5194/acp-5-3423-2005, 2005.
- Shiraiwa, M., Ammann, M., Koop, T., and Pöschl, U.: Gas uptake and chemical aging of semisolid organic aerosol particles, Proceedings of the National Academy of Sciences, 108, 11003, 2011.
- Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, 8, 15002, 10.1038/ncomms15002, doi: <https://www.nature.com/articles/ncomms15002#supplementary-information>, 2017.
- Shiraiwa, M., Pfrang, C., and Pöschl, U.: Kinetic multi-layer model of aerosol surface and bulk chemistry (KM-SUB): the influence of interfacial transport and bulk diffusion on the oxidation of oleic acid by ozone, Atmos. Chem. Phys., 10, 3673-3691, doi: 10.5194/acp-10-3673-2010, 2010.
- Smith, M. H., Park, P. M., and Consterdine, I. E.: Marine aerosol concentrations and estimated fluxes over the sea, Quarterly Journal of the Royal Meteorological Society, 119, 809-824, doi: 10.1002/qj.49711951211, 1993.
- Smith, G. D., Woods, E., DeForest, C. L., Baer, T., and Miller, R. E.: Reactive Uptake of Ozone by Oleic Acid Aerosol Particles: Application of Single-Particle Mass Spectrometry to Heterogeneous Reaction Kinetics, The Journal of Physical Chemistry A, 106, 8085-8095, doi: 10.1021/jp020527t, 2002.
- Song, C. H., and Carmichael, G. R.: The aging process of naturally emitted aerosol (sea-salt and mineral aerosol) during long range transport, Atmospheric Environment, 33, 2203-2218, doi: [https://doi.org/10.1016/S1352-2310\(98\)00301-X](https://doi.org/10.1016/S1352-2310(98)00301-X), 1999.
- Song, C. H., and Carmichael, G. R.: A three-dimensional modeling investigation of the evolution processes of dust and sea-salt particles in east Asia, Journal of Geophysical Research: Atmospheres, 106, 18131-18154, doi:10.1029/2000JD900352, 2001.
- Spindler, G., Hesper, J., Brüggemann, E., Dubois, R., Müller, T., and Herrmann, H.: Wet annular denuder measurements of nitrous acid: laboratory study of the artefact reaction of NO₂ with S(IV) in aqueous solution and comparison with field measurements, Atmospheric Environment, 37, 2643-2662, doi: [https://doi.org/10.1016/S1352-2310\(03\)00209-7](https://doi.org/10.1016/S1352-2310(03)00209-7), 2003.

- Stadtler, S., Simpson, D., Schröder, S., Taraborrelli, D., Bott, A., and Schultz, M.: Ozone impacts of gas-aerosol uptake in global chemistry transport models, *Atmos. Chem. Phys.*, 18, 3147-3171, doi: 10.5194/acp-18-3147-2018, 2018.
- Stanier, C. O., Khlystov, A. Y., and Pandis, S. N.: Ambient aerosol size distributions and number concentrations measured during the Pittsburgh Air Quality Study (PAQS), *Atmospheric Environment*, 38, 3275-3284, doi: <https://doi.org/10.1016/j.atmosenv.2004.03.020>, 2004.
- Stemmler, K., Vlasenko, A., Guimbaud, C., and Ammann, M.: The effect of fatty acid surfactants on the uptake of nitric acid to deliquesced NaCl aerosol, *Atmos. Chem. Phys.*, 8, 5127-5141, doi: 10.5194/acp-8-5127-2008, 2008.
- Stephens, S., Rossi, M. J., and Golden, D. M.: The Heterogeneous Reaction of Ozone on Carbonaceous Surfaces, *International Journal of Chemical Kinetics*, 18, 1133-1149, doi: 10.1002/kin.550181004, 1986.
- 10 Stewart, D. J., Griffiths, P. T., and Cox, R. A.: Reactive uptake coefficients for heterogeneous reaction of N₂O₅ with submicron aerosols of NaCl and natural sea salt, *Atmos. Chem. Phys.*, 4, 1381-1388, doi: 10.5194/acp-4-1381-2004, 2004.
- Su, H., Cheng, Y. F., Cheng, P., Zhang, Y. H., Dong, S., Zeng, L. M., Wang, X., Slanina, J., Shao, M., and Wiedensohler, A.: Observation of nighttime nitrous acid (HONO) formation at a non-urban site during PRIDE-PRD2004 in China, *Atmospheric Environment*, 42, 6219-6232, doi: <https://doi.org/10.1016/j.atmosenv.2008.04.006>, 2008.
- 15 Su, H., Cheng, Y. F., Shao, M., Gao, D. F., Yu, Z. Y., Zeng, L. M., Slanina, J., Zhang, Y. H., and Wiedensohler, A.: Nitrous acid (HONO) and its daytime sources at a rural site during the 2004 PRIDE-PRD experiment in China, *Journal of Geophysical Research: Atmospheres*, 113, doi: 10.1029/2007JD009060, 2008.
- Su, H., Cheng, Y., Oswald, R., Behrendt, T., Trebs, I., Meixner, F. X., Andreae, M. O., Cheng, P., Zhang, Y., and Pöschl, U.: Soil Nitrite as a Source of Atmospheric HONO and OH Radicals, *Science*, 333, 1616, 2011.
- 20 Su, H., Cheng, Y., and Pöschl, U.: The Exchange of Soil Nitrite and Atmospheric HONO: A Missing Process in the Nitrogen Cycle and Atmospheric Chemistry, in: *Disposal of Dangerous Chemicals in Urban Areas and Mega Cities: Role of Oxides and Acids of Nitrogen in Atmospheric Chemistry*, edited by: Barnes, I. and Rudzinski, K. J., Springer Netherlands, Dordrecht, 93–99, 2013.
- 25 Sullivan, R. C., Thornberry, T., and Abbatt, J. P. D.: Ozone decomposition kinetics on alumina: effects of ozone partial pressure, relative humidity and repeated oxidation cycles, *Atmos. Chem. Phys.*, 4, 1301-1310, doi: 10.5194/acp-4-1301-2004, 2004.
- Tan, F., Tong, S., Jing, B., Hou, S., Liu, Q., Li, K., Zhang, Y., and Ge, M.: Heterogeneous reactions of NO₂ with CaCO₃–(NH₄)₂SO₄ mixtures at different relative humidities, *Atmos. Chem. Phys.*, 16, 8081-8093, doi: 10.5194/acp-16-8081-2016, 2016.
- 30

- Tang, M. J., Cox, R. A., and Kalberer, M.: Compilation and evaluation of gas phase diffusion coefficients of reactive trace gases in the atmosphere: volume 1. Inorganic compounds, *Atmos. Chem. Phys.*, 14, 9233-9247, doi: 10.5194/acp-14-9233-2014, 2014.
- 5 Tang, M., Cziczo, D. J., and Grassian, V. H.: Interactions of Water with Mineral Dust Aerosol: Water Adsorption, Hygroscopicity, Cloud Condensation, and Ice Nucleation, *Chemical Reviews*, 116, 4205-4259, doi: 10.1021/acs.chemrev.5b00529, 2016.
- Tang, M. J., Huang, X., Lu, K., Ge, M., Li, Y., Cheng, P., Zhu, T., Ding, A., Zhang, Y., Gligorovski, S., Song, W., Ding, X., Bi, X., and Wang, X.: Heterogeneous reactions of mineral dust aerosol: implications for tropospheric oxidation capacity, *Atmos. Chem. Phys.*, 17, 11727-11777, doi: 10.5194/acp-17-11727-2017, 2017.
- 10 Tang, M. J., Thieser, J., Schuster, G., and Crowley, J. N.: Kinetics and mechanism of the heterogeneous reaction of N₂O₅ with mineral dust particles, *Physical Chemistry Chemical Physics*, 14, 8551-8561, doi: 10.1039/C2CP40805H, 2012.
- Tang, M. J., Schuster, G., and Crowley, J. N.: Heterogeneous reaction of N₂O₅ with illite and Arizona test dust particles, *Atmos. Chem. Phys.*, 14, 245-254, doi: 10.5194/acp-14-245-2014, 2014.
- 15 Thornton, J. A., and Abbatt, J. P. D.: N₂O₅ Reaction on Submicron Sea Salt Aerosol: Kinetics, Products, and the Effect of Surface Active Organics, *The Journal of Physical Chemistry A*, 109, 10004-10012, doi: 10.1021/jp054183t, 2005.
- Thornton, J. A., Braban, C. F., and Abbatt, J. P. D.: N₂O₅ hydrolysis on sub-micron organic aerosols: the effect of relative humidity, particle phase, and particle size, *Physical Chemistry Chemical Physics*, 5, 4593-4603, doi: 10.1039/B307498F, 2003.
- 20 Tolocka, M. P., Saul, T. D., and Johnston, M. V.: Reactive Uptake of Nitric Acid into Aqueous Sodium Chloride Droplets Using Real-Time Single-Particle Mass Spectrometry, *The Journal of Physical Chemistry A*, 108, 2659-2665, doi: 10.1021/jp036612y, 2004.
- Ullerstam, M., Johnson, M. S., Vogt, R., and Ljungström, E.: DRIFTS and Knudsen cell study of the heterogeneous reactivity of SO₂ and NO₂ on mineral dust, *Atmos. Chem. Phys.*, 3, 2043-2051, doi: 10.5194/acp-3-2043-2003, 2003.
- 25 Ullerstam, M., Vogt, R., Langer, S., and Ljungstrom, E.: The kinetics and mechanism of SO₂ oxidation by O₃ on mineral dust, *Physical Chemistry Chemical Physics*, 4, 4694-4699, doi: 10.1039/B203529B, 2002.
- Umann, B., Arnold, F., Schaal, C., Hanke, M., Uecker, J., Aufmhoff, H., Balkanski, Y., and Van Dingenen, R.: Interaction of mineral dust with gas phase nitric acid and sulfur dioxide during the MINATROC II field campaign: First estimate of the uptake coefficient from atmospheric data, *Journal of Geophysical Research: Atmospheres*, 110, doi:10.1029/2005JD005906, 2005.
- 30 Underwood, G. M., Li, P., Al-Abadleh, H., and Grassian, V. H.: A Knudsen Cell Study of the Heterogeneous Reactivity of Nitric Acid on Oxide and Mineral Dust Particles, *The Journal of Physical Chemistry A*, 105, 6609-6620, doi: 10.1021/jp002223h, 2001a.

- Underwood, G. M., Song, C. H., Phadnis, M., Carmichael, G. R., and Grassian, V. H.: Heterogeneous reactions of NO₂ and HNO₃ on oxides and mineral dust: A combined laboratory and modeling study, *Journal of Geophysical Research: Atmospheres*, 106, 18055-18066, doi: 10.1029/2000JD900552, 2001b.
- Usher, C. R., Al-Hosney, H., Carlos-Cuellar, S., and Grassian, V. H.: A laboratory study of the heterogeneous uptake and oxidation of sulfur dioxide on mineral dust particles, *Journal of Geophysical Research: Atmospheres*, 107, ACH 16-11-ACH 16-19, doi: 10.1029/2002JD002051, 2002.
- Usher, C. R., Michel, A. E., and Grassian, V. H.: Reactions on Mineral Dust, *Chemical Reviews*, 103, 4883-4940, doi: 10.1021/cr020657y, 2003.
- Vlasenko, A., Sjogren, S., Weingartner, E., Stemmler, K., Gäggeler, H. W., and Ammann, M.: Effect of humidity on nitric acid uptake to mineral dust aerosol particles, *Atmos. Chem. Phys.*, 6, 2147-2160, doi: 10.5194/acp-6-2147-2006, 2006.
- Vogt, R., and Finlayson-Pitts, B. J.: A Diffuse Reflectance Infrared Fourier Transform Spectroscopic Study of the Surface Reaction of NaCl with Gaseous NO₂ and HNO₃, *The Journal of Physical Chemistry*, 98, 3747-3755, doi: 10.1021/j100065a033, 1994.
- Wagner, C., Hanisch, F., Holmes, N., de Coninck, H., Schuster, G., and Crowley, J. N.: The interaction of N₂O₅ with mineral dust: aerosol flow tube and Knudsen reactor studies, *Atmos. Chem. Phys.*, 8, 91-109, doi: 10.5194/acp-8-91-2008, 2008.
- Wagner, V., von Glasow, R., Fischer, H., and Crutzen, P. J.: Are CH₂O measurements in the marine boundary layer suitable for testing the current understanding of CH₄ photooxidation?: A model study, *Journal of Geophysical Research: Atmospheres*, 107, ACH 3-1-ACH 3-14, doi: 10.1029/2001JD000722, 2002.
- Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, *Atmos. Chem. Phys.*, 12, 10209-10237, doi: 10.5194/acp-12-10209-2012, 2012.
- Wang, X., Wang, W., Yang, L., Gao, X., Nie, W., Yu, Y., Xu, P., Zhou, Y., and Wang, Z.: The secondary formation of inorganic aerosols in the droplet mode through heterogeneous aqueous reactions under haze conditions, *Atmospheric Environment*, 63, 68-76, doi: <https://doi.org/10.1016/j.atmosenv.2012.09.029>, 2012.
- Wang, Y., Zhuang, G., Sun, Y., and An, Z.: The variation of characteristics and formation mechanisms of aerosols in dust, haze, and clear days in Beijing, *Atmospheric Environment*, 40, 6579-6591, doi: <https://doi.org/10.1016/j.atmosenv.2006.05.066>, 2006.
- Wagner, C., Schuster, G., and Crowley, J. N.: An aerosol flow tube study of the interaction of N₂O₅ with calcite, Arizona dust and quartz, *Atmospheric Environment*, 43, 5001-5008, doi: <https://doi.org/10.1016/j.atmosenv.2009.06.050>, 2009.
- Weber, B., Wu, D., Tamm, A., Ruckteschler, N., Rodríguez-Caballero, E., Steinkamp, J., Meusel, H., Elbert, W., Behrendt, T., Sörgel, M., Cheng, Y., Crutzen, P. J., Su, H., and Pöschl, U.: Biological soil crusts accelerate the nitrogen cycle

- through large NO and HONO emissions in drylands, *Proceedings of the National Academy of Sciences*, 112, 15384, doi: 10.1073/pnas.1515818112, 2015.
- Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, *Atmospheric Environment* (1967), 23, 1293-1304, doi: [https://doi.org/10.1016/0004-6981\(89\)90153-4](https://doi.org/10.1016/0004-6981(89)90153-4), 1989.
- 5 Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, *Atmospheric Environment*, 41, 52-63, doi: <http://dx.doi.org/10.1016/j.atmosenv.2007.10.058>, 2007.
- Wesely, M. L., and Hicks, B. B.: A review of the current status of knowledge on dry deposition, *Atmospheric Environment*, 34, 2261-2282, doi: [https://doi.org/10.1016/S1352-2310\(99\)00467-7](https://doi.org/10.1016/S1352-2310(99)00467-7), 2000.
- Woo, K. S., Chen, D. R., Pui, D. Y. H., and McMurry, P. H.: Measurement of Atlanta Aerosol Size Distributions: Observations of Ultrafine Particle Events, *Aerosol Science and Technology*, 34, 75-87, doi: 10.1080/02786820120056, 2001.
- 10 Worsnop, D. R., Zahniser, M. S., Kolb, C. E., Gardner, J. A., Watson, L. R., Van Doren, J. M., Jayne, J. T., and Davidovits, P.: The temperature dependence of mass accommodation of sulfur dioxide and hydrogen peroxide on aqueous surfaces, *The Journal of Physical Chemistry*, 93, 1159-1172, doi: 10.1021/j100340a027, 1989.
- 15 Wu, Z., Hu, M., Lin, P., Liu, S., Wehner, B., and Wiedensohler, A.: Particle number size distribution in the urban atmosphere of Beijing, China, *Atmospheric Environment*, 42, 7967-7980, doi: <https://doi.org/10.1016/j.atmosenv.2008.06.022>, 2008.
- Wu, Z. J., Ma, N., Größ, J., Kecorius, S., Lu, K. D., Shang, D. J., Wang, Y., Wu, Y. S., Zeng, L. M., Hu, M., Wiedensohler, A., and Zhang, Y. H.: Thermodynamic properties of nanoparticles during new particle formation events in the atmosphere of North China Plain, *Atmospheric Research*, 188, 55-63, doi: <https://doi.org/10.1016/j.atmosres.2017.01.007>, 2017.
- 20 Xu, W., Li, Q., Shang, J., Liu, J., Feng, X., and Zhu, T.: Heterogeneous oxidation of SO₂ by O₃-aged black carbon and its dithiothreitol oxidative potential, *Journal of Environmental Sciences*, 36, 56-62, doi: <https://doi.org/10.1016/j.jes.2015.02.014>, 2015.
- 25 Yabushita, A., Enami, S., Sakamoto, Y., Kawasaki, M., Hoffmann, M. R., and Colussi, A. J.: Anion-Catalyzed Dissolution of NO₂ on Aqueous Microdroplets, *The Journal of Physical Chemistry A*, 113, 4844-4848, doi: 10.1021/jp900685f, 2009.
- Ye, C., Li, H., Zhu, T., Shang, J., Zhang, Z., and Zhao, D.: Heterogeneous reaction of NO₂ with sea salt particles, *Science China Chemistry*, 53, 2652-2656, doi: 10.1007/s11426-010-4159-9, 2010.
- 30 Zhang, L., Brook, J. R., and Vet, R.: On ozone dry deposition—with emphasis on non-stomatal uptake and wet canopies, *Atmospheric Environment*, 36, 4787-4799, doi: [http://dx.doi.org/10.1016/S1352-2310\(02\)00567-8](http://dx.doi.org/10.1016/S1352-2310(02)00567-8), 2002.

- Zhang, Y., and Carmichael, G. R.: The Role of Mineral Aerosol in Tropospheric Chemistry in East Asia—A Model Study, *Journal of Applied Meteorology*, 38, 353-366, doi: 10.1175/1520-0450(1999)038<0353:TROMAI>2.0.CO;2, 1999.
- Zhang, Y., Tong, S., Ge, M., Jing, B., Hou, S., Tan, F., Chen, Y., Guo, Y., and Wu, L.: The influence of relative humidity on the heterogeneous oxidation of sulfur dioxide by ozone on calcium carbonate particles, *Science of The Total Environment*, 633, 1253-1262, doi: <https://doi.org/10.1016/j.scitotenv.2018.03.288>, 2018.
- Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, *Atmos. Chem. Phys.*, 3, 2067-2082, doi: 10.5194/acp-3-2067-2003, 2003.
- Zhang, Q.: China Standard Meteorological Database for Construction (CD-ROM), China Mechanical Industry Press, Beijing, 2004.
- 10 Zhao, Y., Chen, Z., Shen, X., and Zhang, X.: Kinetics and Mechanisms of Heterogeneous Reaction of Gaseous Hydrogen Peroxide on Mineral Oxide Particles, *Environmental Science & Technology*, 45, 3317-3324, doi: 10.1021/es104107c, 2011.
- 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 15 2013 haze episode in North China, *Atmos. Chem. Phys.*, 15, 2031-2049, doi: 10.5194/acp-15-2031-2015, 2015.
- Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Pöschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, *Atmos. Chem. Phys.*, 15, 2969-2983, doi: 10.5194/acp-15-2969-2015, 2015.
- Zhou, J., Swietlicki, E., Hansson, H. C., and Artaxo, P.: Submicrometer aerosol particle size distribution and hygroscopic growth measured in the Amazon rain forest during the wet season, *Journal of Geophysical Research: Atmospheres*, 107, LBA 22-21-LBA 22-10, doi: 10.1029/2000JD000203, 2002.
- 20 Zhou, L., Wang, W., Ge, M., and Tong, S.: Heterogeneous uptake of gaseous hydrogen peroxide on mineral dust, *Journal of Environmental Sciences*, 40, 44-50, doi: <https://doi.org/10.1016/j.jes.2015.08.018>, 2016.
- Zhou, X., Lee, Y.-N., Newman, L., Chen, X., and Mopper, K.: Tropospheric formaldehyde concentration at the Mauna Loa 25 Observatory during the Mauna Loa Observatory Photochemistry Experiment 2, *Journal of Geophysical Research: Atmospheres*, 101, 14711-14719, doi:10.1029/95JD03226, 1996.
- 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, doi: 10.5194/acp-10-3855-2010, 2010.

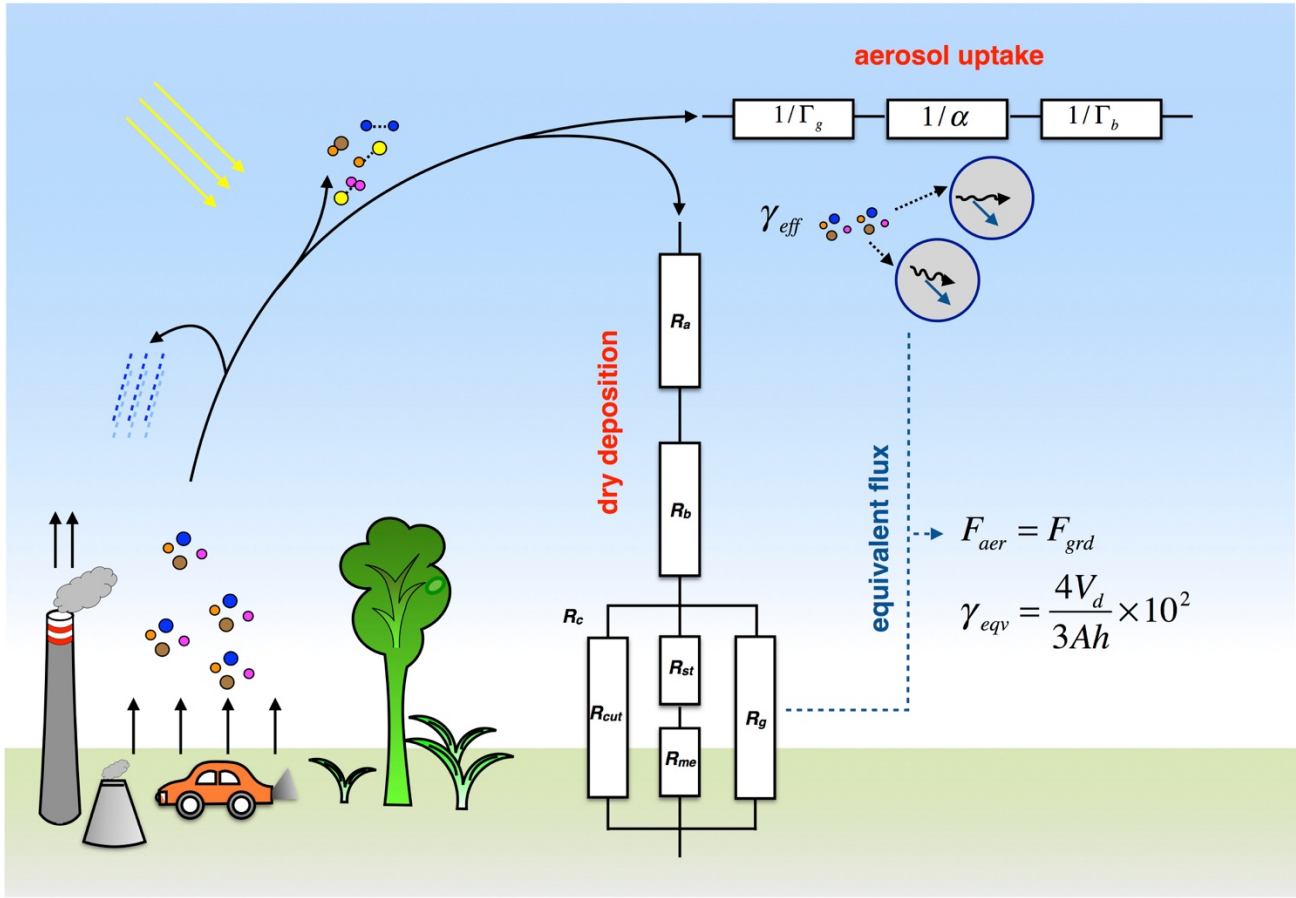


Figure 1. Schematic illustration of gas uptake on the ground and on aerosols in the planetary boundary layer as characterized by resistance models. The relative importance of aerosol uptake and dry deposition on the ground is characterized through comparing the aerosol uptake coefficient (γ_{eff}) with an equivalent uptake coefficient (γ_{eqv}) corresponding to the deposition velocity (V_d).

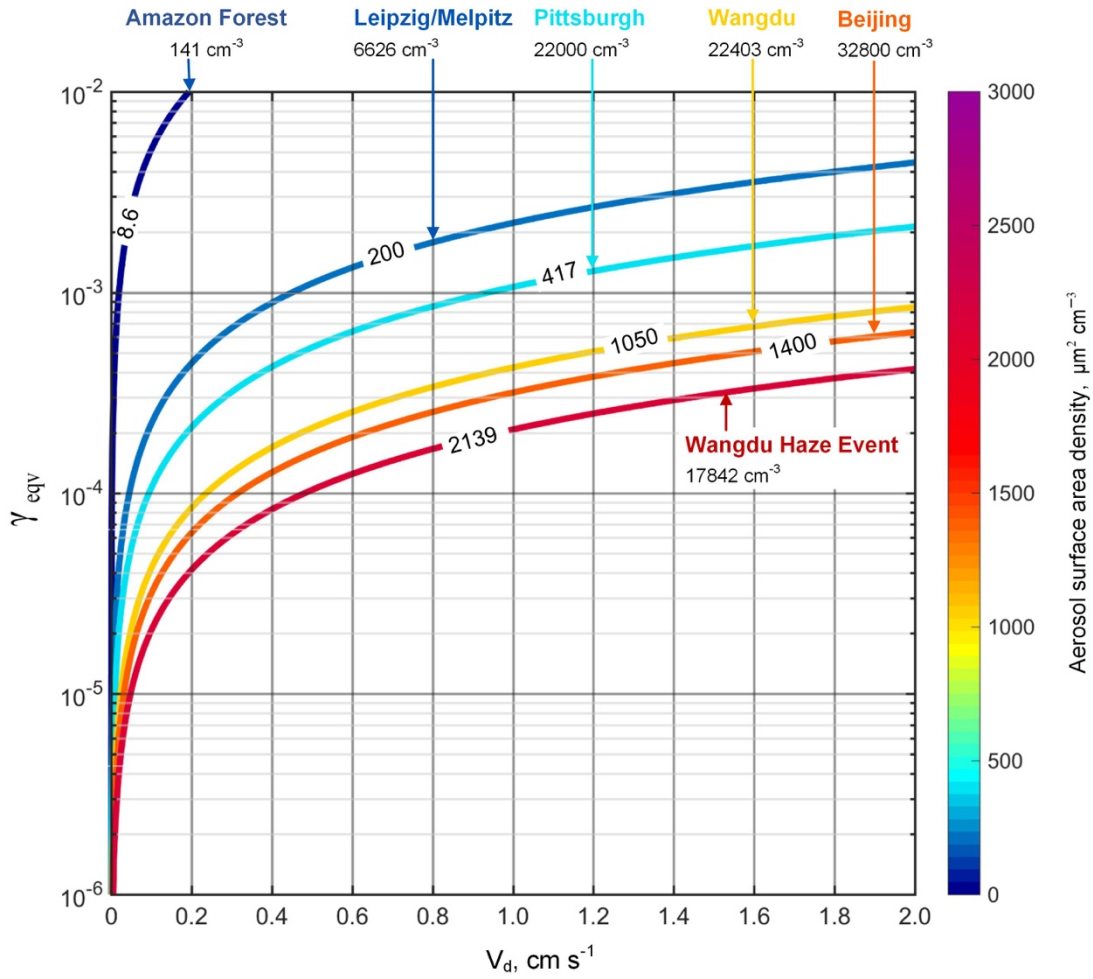


Figure 2. Relation between γ_{eqv} and V_d for a mixing height of 300 m and aerosol surface area densities (A) observed at locations and conditions: Amazon Forest (Pöschl et al., 2010), Leipzig/Melpitz (Ma et al., 2014; Ma and Birmili, 2015), Pittsburgh (Stanier et al., 2004), Wangdu with and without haze event (Wu et al., 2017), and Beijing (Wu et al., 2008). For each city/condition, the line is labelled with the corresponding aerosol surface area density. Aerosol particle number concentrations are also provided for orientation.

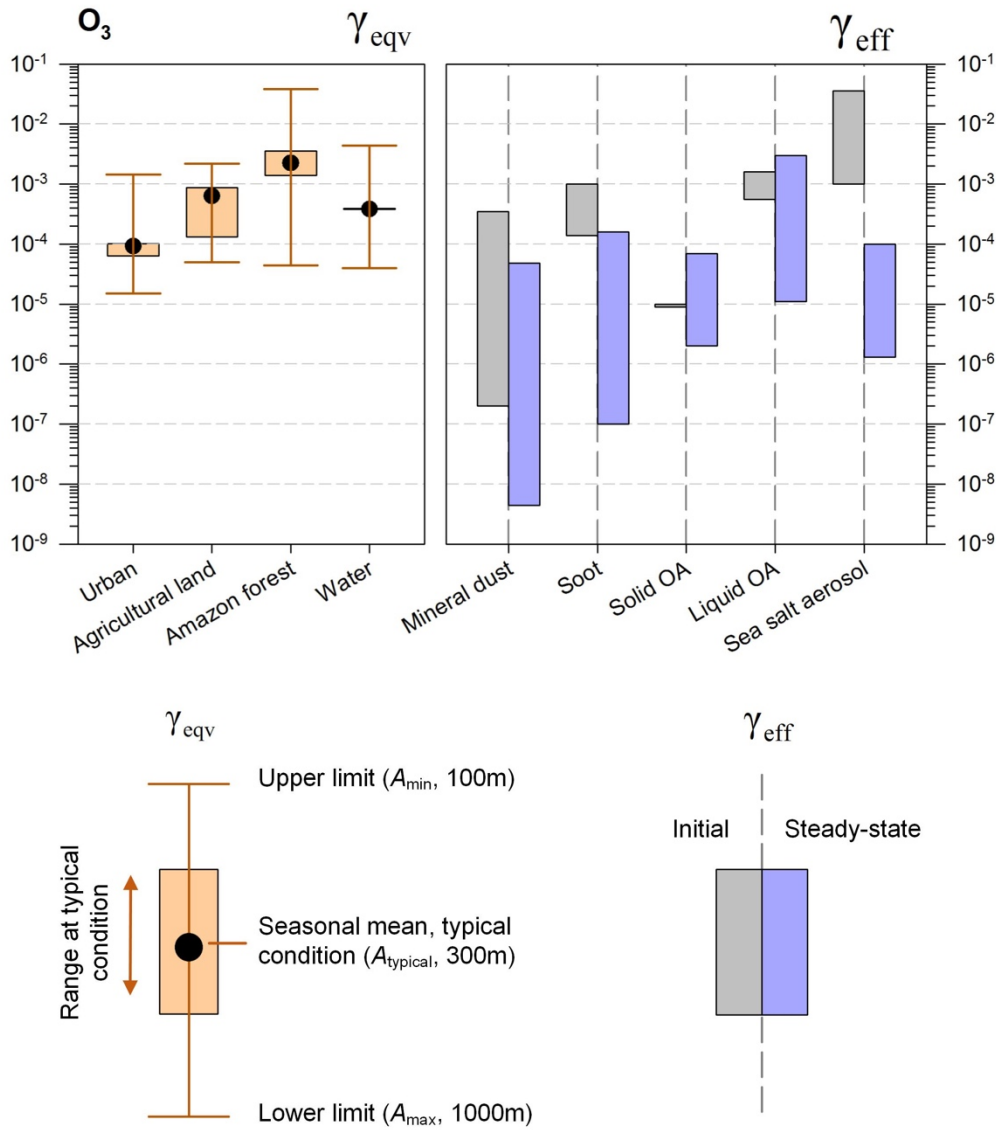


Figure 3. Equivalent uptake coefficients (γ_{eqv} , left) and laboratory measurement values (γ_{eff} , right) for O_3 on different ground types and aerosols.

For γ_{eqv} , upper whiskers represent maximum values calculated at lowest A and h ($h = 100$ m), lower whiskers represent minimum values calculated at highest A and h ($h = 1$ km), and boxes represent typical conditions (typical A as described in Sect. 3.1, $h = 300$ m). For γ_{eff} , the grey bar represents the range of initial values, and the blue bar represents the range of steady-state values observed in laboratory experiments.

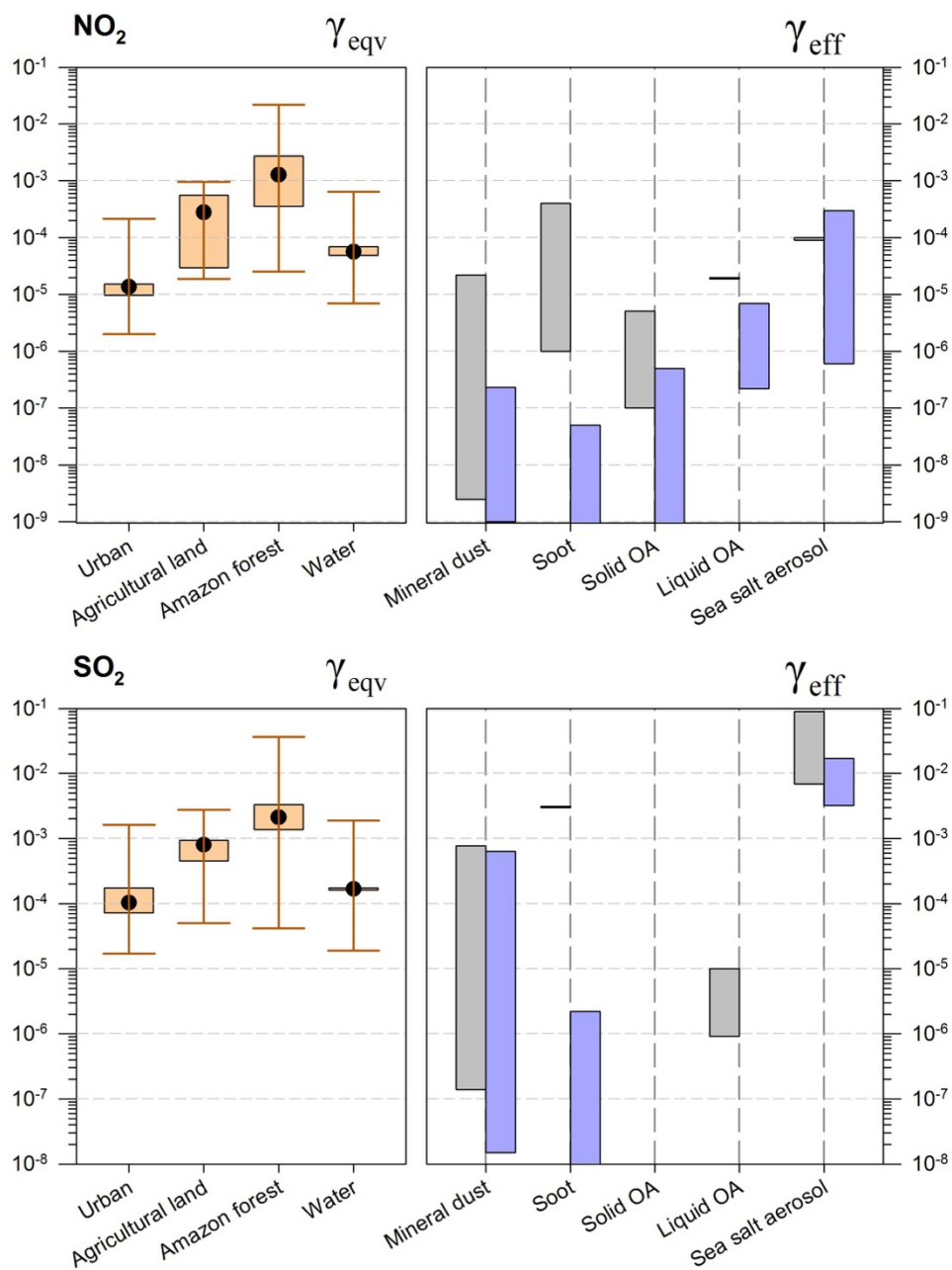


Figure 4. Uptake coefficients (γ_{eqv} , left; γ_{eff} , right) for NO_2 and SO_2 on different ground types and aerosols.

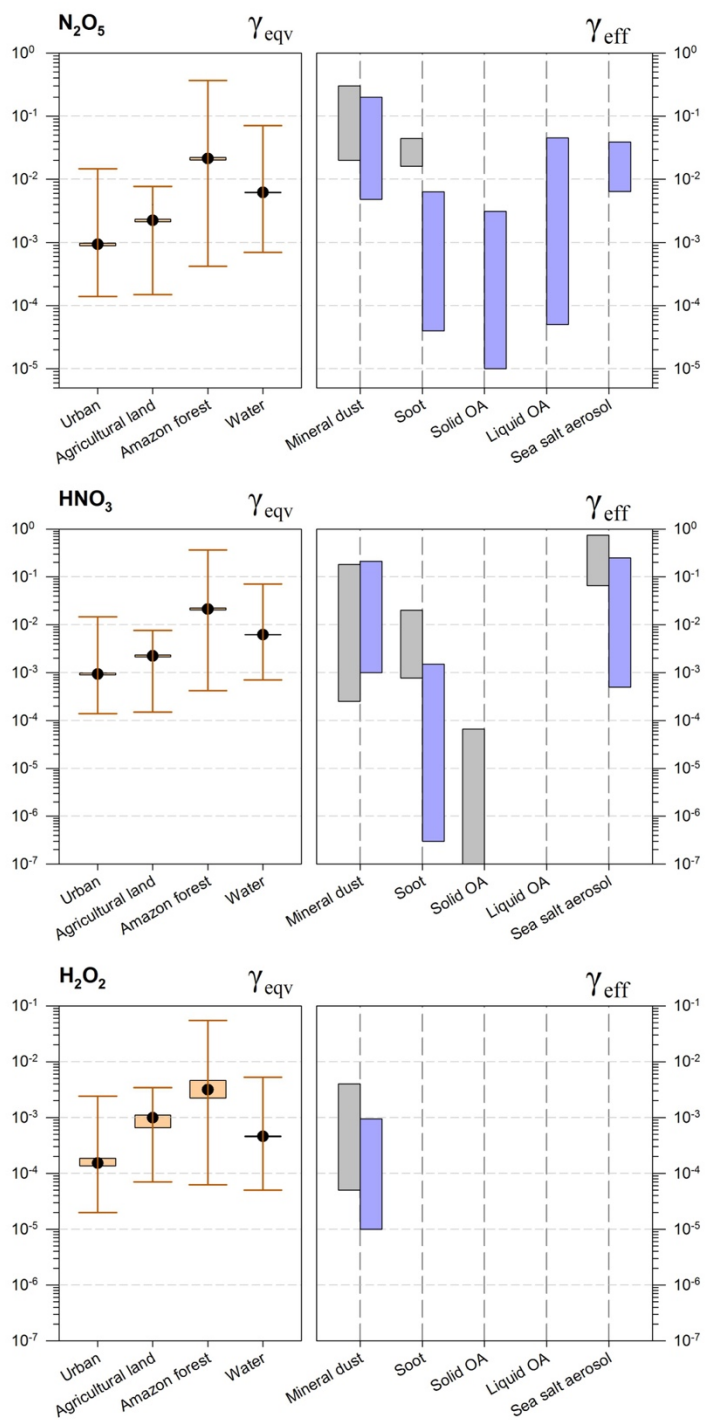


Figure 5. Uptake coefficients (γ_{eqv} , left; γ_{eff} , right) for N_2O_5 , HNO_3 and H_2O_2 on different ground types and aerosols.

Table 1. Aerosol uptake coefficients (γ_{eff}) observed in laboratory experiments ^a.

Gases	Mineral dust	Soot	Organic aerosol-solid	Organic aerosol-liquid	Sea salt aerosol
Steady state ^b					
O ₃	$4.4 \times 10^{-9} - 4.8 \times 10^{-5}$	$1.0 \times 10^{-7} - 1.6 \times 10^{-4}$	$2.0 \times 10^{-6} - 6.9 \times 10^{-5}$	$1.1 \times 10^{-5} - 3.0 \times 10^{-3}$	$1.3 \times 10^{-6} - 1.0 \times 10^{-4}$
NO ₂	$1.0 \times 10^{-9} - 2.3 \times 10^{-7}$	$< 5.0 \times 10^{-8}$	$< 5.0 \times 10^{-7}$	$2.2 \times 10^{-7} - 7.0 \times 10^{-6}$	$6.0 \times 10^{-7} - 3.0 \times 10^{-4}$
SO ₂	$1.5 \times 10^{-8} - 6.3 \times 10^{-4}$	$4.0 \times 10^{-9} - 2.2 \times 10^{-6}$	n/a	n/a	$3.2 \times 10^{-3} - 1.7 \times 10^{-2}$
N ₂ O ₅	$4.8 \times 10^{-3} - 2.0 \times 10^{-1}$	$4.0 \times 10^{-5} - 6.3 \times 10^{-3}$	$1.0 \times 10^{-5} - 3.1 \times 10^{-3}$	$5.0 \times 10^{-5} - 4.5 \times 10^{-2}$	$6.4 \times 10^{-3} - 3.9 \times 10^{-2}$
HNO ₃	$1.0 \times 10^{-3} - 2.1 \times 10^{-1}$	$3.0 \times 10^{-7} - 1.5 \times 10^{-3}$	n/a	n/a	$5.0 \times 10^{-4} - 2.5 \times 10^{-1}$
H ₂ O ₂	$1.0 \times 10^{-5} - 9.4 \times 10^{-4}$	n/a ^c	n/a	n/a	n/a
Initial state ^b					
O ₃	$2.0 \times 10^{-7} - 3.5 \times 10^{-4}$	$1.4 \times 10^{-4} - 1.0 \times 10^{-3}$	1.0×10^{-5}	$5.5 \times 10^{-4} - 1.6 \times 10^{-3}$	$1.0 \times 10^{-3} - 3.6 \times 10^{-2}$
NO ₂	$2.5 \times 10^{-9} - 2.2 \times 10^{-5}$	$1.0 \times 10^{-6} - 4.0 \times 10^{-4}$	$1.0 \times 10^{-7} - 5.1 \times 10^{-6}$	2.0×10^{-5}	1.0×10^{-4}
SO ₂	$1.4 \times 10^{-7} - 7.7 \times 10^{-4}$	3.0×10^{-3}	n/a	$9.2 \times 10^{-7} - 1.0 \times 10^{-5}$	$6.9 \times 10^{-3} - 9.0 \times 10^{-2}$
N ₂ O ₅	$2.0 \times 10^{-2} - 3.0 \times 10^{-1}$	$1.6 \times 10^{-2} - 4.4 \times 10^{-2}$	n/a	n/a	n/a
HNO ₃	$2.5 \times 10^{-4} - 1.8 \times 10^{-1}$	$7.7 \times 10^{-4} - 2.0 \times 10^{-2}$	$\leq 6.6 \times 10^{-5}$	n/a	$6.6 \times 10^{-2} - 7.5 \times 10^{-1}$
H ₂ O ₂	$5.0 \times 10^{-5} - 4.0 \times 10^{-3}$	n/a	n/a	n/a	n/a

^a The detailed review table with references of γ_{eff} are in Table A.1~A.4.

^b The feature (initial or steady state) of the reported uptake coefficients are mainly derived from the literature. If no specific description is found, we assign the measurements on a timescale of ms or s to initial state, and those with longer exposure time (~1h or longer) to steady state.

^c n/a: not available.

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 ₂ O ₅	✓	✓	✓	✓	✓
HNO ₃	✓				✓
H ₂ O ₂	✓				

5

5 **Table A.1. Aerosol uptake coefficients (γ_{eff}) for reactive gases on mineral dust observed in laboratory experiments (T=298±2K if not specified otherwise).**

Gases (Unit)	Aerosol type	Initial γ_{eff} , geometric surface	Initial γ_{eff} , BET	Steady-state γ_{eff} , geometric surface	Steady-state γ_{eff} , BET	References
O_3 ($\times 10^{-5}$)	TiO ₂ /SiO ₂	0.55 ~35		0.3~3	0.02~0.32	Nicolas et al., 2009
	Al ₂ O ₃ , Fe ₂ O ₃ , SiO ₂		5~18			Michel et al., 2002
	China loess		2.7			Michel et al., 2002
	Saharan sand		6			Michel et al., 2002
	Al ₂ O ₃ and others		0.27~20		0.6~2.2	Michel et al., 2003
	Saharan dust			0.22~4.8		Hanisch and Crowley, 2003
	Al ₂ O ₃		0.1~ 1			Sullivan et al., 2004
	Saharan dust		0.02~0.6			Chang et al., 2005
	Mineral dust				0.00044~ 0.01	Mogili et al., 2006
	Summary	0.02~35		0.00044~ 4.8		
NO_2 ($\times 10^{-7}$)	Al ₂ O ₃	0.656		9400, 1200	0.013~0.26	Börensén et al., 2000
	Al ₂ O ₃ and others		0.2~220			Underwood et al., 2001b
	Saharan dust		6.2			Ullerstam et al., 2003
	Illuminated TiO ₂					Gustafsson et al., 2006
	CaCO ₃		0.025~ 0.043			Li et al., 2010
	Kaolinite, pyrophyllite				0.07~0.81, 2.3	Angelini et al., 2007
	TiO ₂ /SiO ₂ , Saharan sand and others				0.01	Ndour et al., 2008
	Illuminated TiO ₂ /SiO ₂				1.2~19	Ndour et al., 2008
	Saharan sand				0.089, 1	Ndour et al., 2009
	Arizona test dust				0.06~0.24	Dupart et al., 2014
	Kaolin		0.31~1.44		0.0256~0.0456	Liu et al., 2015
	Hematite		0.186~1.58		0.0123~0.0150	Liu et al., 2015
	Summary	0.025~220		0.01~ 2.3		
SO_2 ($\times 10^{-5}$)	CaCO ₃ , O ₃	77	0.014	8.1	0.0015	Li et al., 2006
	Saharan dust, O ₃			390	0.05	Ullerstam et al., 2002
	Saharan dust		1.6			Ullerstam et al., 2003

	Al ₂ O ₃ , MgO		9.5, 26			Goodman et al., 2001
	Al ₂ O ₃ and others		7.0 ~ 51			Usher et al., 2002
	Chinese loess		3			Usher et al., 2002
	Saharan dust, O ₃		6.6			Adams et al., 2005
	Al ₂ O ₃ Fe ₂ O ₃ , MgO		40, 55, 100			Judeikis et al, 1978
	CaCO ₃ powder		0.1			Santschi and Rossi, 2006
	CaCO ₃ , O ₃	43.5~65.6	0.026~0.039	0.54~22.1	0.32~13.2	Zhang et al., 2018
	Asian mineral dust			10.1~21.4		Huang et al., 2015
	Tengger desert dust			22.9~39.0		Huang et al., 2015
	Arizona test dust			3.5~9.2		Huang et al., 2015
	Asian mineral dust, H ₂ O ₂			39.1~54.5		Huang et al., 2015
	Tengger desert dust, H ₂ O ₂			37.2~63.1		Huang et al., 2015
	Arizona test dust, H ₂ O ₂			4.6~13.1		Huang et al., 2015
	Summary	0.014~77		0.0015~63.1		
N ₂ O ₅ (×10 ⁻²)	Saharan sand	30		20		Karagulian et al., 2006
	Arizona test dust	20		11		Karagulian et al., 2006
	CaCO ₃	12		2.1		Karagulian et al., 2006
	Kaolinite	16~23		2.2~2.4		Karagulian et al., 2006
	Saharan sand	8		1.3		Seisel et al., 2005
	Arizona dust	0.5~1.3				Wagner et al., 2008
	Saharan sand	3.7		3.7		Wagner et al., 2008
	Arizona dust	2.2		2.2		Wagner et al., 2008
	CaCO ₃	5.0				Wagner et al., 2008
	CaCO ₃			0.48~0.53		Wagner et al., 2009
	CaCO ₃			1.13~1.94		Wagner et al., 2009
	Arizona dust			0.73~0.98		Wagner et al., 2009
	Quarz			0.86~0.45		Wagner et al., 2009
	Saharan sand				2	Tang et al., 2012
	Arizona dust			0.63		Tang et al., 2014
	Illite			9.1, 3.9		Tang et al., 2014
	Summary	2~30		0.48~20		
HNO ₃	Mineral dust			1.7~5.4		Umann et al., 2005

(×10 ⁻²)	Arizona dust, CaCO ₃ and SiO ₂	2~11.3				Vlasenko et al., 2006
	CaCO ₃			0.3~21		Liu et al., 2008
	CaCO ₃	6.0~15				Fenter et al., 1995
	CaCO ₃		0.025			Goodman et al., 2000
	Saharan dust, Arizona dust, CaCO ₃	11, 6, 10~18				Hanisch and Crowley, 2001a
	Saharan dust	13.6				Hanisch and Crowley, 2001b
	Chinese dust	17.1				Hanisch and Crowley, 2001b
	Al ₂ O ₃ and others		0.002~0.0097			Underwood et al., 2001a
	Al ₂ O ₃ and others		0.002~0.61			Underwood et al., 2001b
	Fe ₂ O ₃		0.0015			Frinak et al., 2004
	Al ₂ O ₃ , Saharan dust	13, 11	13, 11			Seisel et al., 2004
	CaCO ₃		0.2			Johnson et al., 2005
	CaCO ₃ powder		0.7~30		0.07~0.2	Santschi and Rossi, 2006
	Summary	0.025~18		0.1 ~ 21		
H ₂ O ₂ (×10 ⁻⁴)	Saharan sand			6.20~9.42		Pradhan et al., 2010b
	Gobi sand			3.33~6.03		Pradhan et al., 2010b
	TiO ₂			15, 5		Pradhan et al., 2010a
	Arizona test dust		1.47~2.71		0.557~ 0.995	Zhou et al., 2016 ^a
	Inner Mongolia desert dust		2.19~3.56		0.25~1.31	Zhou et al., 2016 ^a
	Xinjiang dust		0.446~0.734		0.377~0.431	Zhou et al., 2016 ^a
	Arizona test dust		3.2		0.095~0.185	El Zein et al., 2014 ^b
	TiO ₂ , dark		2.5~40			Romanias et al., 2012
	TiO ₂ , UV				35	Romanias et al., 2012
	Al ₂ O ₃		9.0			Romanias et al., 2013
	Fe ₂ O ₃		8.6			Romanias et al., 2013
	SiO ₂				0.000155, 0.000061	Zhao et al., 2011
	Al ₂ O ₃				0.00121, 0.00076	Zhao et al., 2011
	Ambient urban aerosol				0.81 ~ 4.63	Wu et al., 2015
	Summary	0.5~40		0.1~9.42		

^a T=253-313K

^b T= 268 - 320K

Table A.2. Aerosol uptake coefficients (γ_{eff}) for reactive gases on soot observed in laboratory experiments (T=298±2K if not specified otherwise).

Gases (Unit)	Aerosol type	Initial γ_{eff} , geometric surface	Initial γ_{eff} , BET	Steady-state γ_{eff} , geometric surface	Steady-state γ_{eff} , BET	References
O_3 ($\times 10^{-5}$)	BC	100				Rogaski et al., 1997
	Hydrocarbon soot			16	0.5	Longfellow et al., 2000
	Candle soot		13.9		0.628	Il'in, 1991
	Degussa carbon black		~100		~0.001	Disselkamp et al., 2000
	Spark generated		22~330			Fendel et al., 1995
	Spark generated		0.12		0.01	Kamm et al., 1999
	Kerosene, toluene soot		18 ~ 38			Lelièvre et al., 2004b
	Charcoal		22 ~ 413		2.7~11.3	Stephens et al., 1986
	Spark-generated soot coated with benzo[a]pyrene			0.6~2		Pöschl et al., 2001
	Summary	13.9~100		0.01~16		
NO_2 ($\times 10^{-5}$)	Hydrocarbon soot		2.9~5.0		<0.001	Lelièvre et al., 2004a
	Spark generated		0.15~170		0.0016~0.61	Kirchner et al., 2000
	Hexane soot	150~1840	2.5 ~ 4.72		0.48~1.17	Al-Abadleh and Grassian, 2000
	Spark generated				<= 0.004	Saathoff et al., 2001
	Hexane soot, BC				0.0015~0.0024	Prince et al., 2002
	Spark-generated, commercial soot	3~40				Kalberer et al., 1996
	Ambient soot	1100		33		Ammann et al. 1998
	Commercial soot		0.1		<0.001	Kleffmann et al., 1999
	Spark-generated		0.5~1.0			Arens et al., 2001
	Summary	0.1~40		<0.005		
SO_2 ($\times 10^{-6}$)	BC	3000				Rogaski et al., 1997
	Fresh BC, aged BC				0.00398, 0.32	Xu et al., 2015
	BC, O_3				2.17	Xu et al., 2015
	Summary	3000		0.004 ~ 2.17		
N_2O_5 ($\times 10^{-3}$)	Decane soot	44		5		Karagulian and Rossi, 2007
	Spark generated				0.04~0.2	Saathoff et al., 2001

	Hydrocarbon soot	16		6.3		Longfellow et al., 2000
	Summary	16~44		0.04~6.3		
HNO ₃ (×10 ⁻⁴)	Decane soot	200		4.6~5.2		Salgado-Muñoz and Rossi, 2002
	Spark generated				0.003	Saathoff et al., 2001
	Hydrocarbon soot			15	0.5	Longfellow et al., 2000
	Spark generated		0.052~7.7		0.00098~0.019	Kirchner et al., 2000
	Summary	7.7~200		0.003~15		

Table A.3. Aerosol uptake coefficients (γ_{eff}) for reactive gases on organic aerosols observed in laboratory experiments (T=298±2K if not specified otherwise).

Gases (Unit)	Aerosol type	Initial γ_{eff} , geometric surface	Initial γ_{eff} , BET	Steady-state γ_{eff} , geometric surface	Steady-state γ_{eff} , BET	References
O ₃ (×10 ⁻⁵)	Semi-solid protein aerosol	1.0		< 1.0		Shiraiwa et al., 2011
	Shikimic acid film			0.2~1.0		Berkemeier et al., 2016
	Solid 1-hexadecene			0.64~2.5		Moise and Rudich, 2000 ^a
	Monolayer organic film			17~27		Moise and Rudich, 2000 ^b
	Solid-liquid oleic acid			2~17		Knopf et al., 2005
	Solid-liquid oleic acid (meat-cooking aerosols)			1.6~6.9		Knopf et al., 2005
	Liquid oleic acid particle	150		5		Mendez et al., 2014
	Liquid organic compounds			1.0~100		de Gouw et al., 1998
	Aqueous α -pinene aerosol			300~750		King et al., 2008 ^c
	Aqueous fumarate aerosol			1.1		King et al., 2008 ^c
	Aqueous benzoate aerosol			1.5		King et al., 2008 ^c
	Liquid oleic acid aerosol	160				Morris et al., 2002
	Oleic acid aerosol	55~90		20~100		Sage et al., 2009
	Liquid 1-tridecene			52~55		Moise and Rudich, 2000 ^d
	Liquid 1-hexadecene			32~38		Moise and Rudich, 2000 ^e
	Liquid 1-hexadecane			2.0		Moise and Rudich, 2000
	Liquid oleic acid			88		Hearn et al., 2005
	Liquid oleic acid			40~72		Knopf et al., 2005
	Liquid oleic acid			730		Smith et al., 2002
	Summary	~1.0 for solid, 55~160 for liquid		0.2~6.9 for solid, 1.1~300 for liquid		
NO ₂ (×10 ⁻⁶)	Solid benzophenone, catechol, anthracene, anthracene, anthracene	0.07~1.26 (dark), 0.65~2.40 (light)				George et al., 2005 ^f
	Solid benzophenone, catechol, anthracene, anthracene	0.24 ~ 3.6 (dark), 1.3~5.1 (light)				George et al., 2005 ^f
	Solid 1,2,10-trihydroxyanthracene	0.7 ~ 2				Arens et al., 2002
	Solid 1,2,10-trihydroxyanthracene			< 0.5		Arens et al., 2002
	Nitroguaiacol, mixture of organics	52, 22				Knopf et al., 2011
	Solid levoglucosan, abietic acid	< 1.0				Knopf et al., 2011
	Solid pyrene, dark, near-UV			< 0.1, 3.5		Brigante et al., 2008
	Solid pyrene	<= 1.0				Gross et al., 2008 ^g

	SOA, pinene/O ₃ SOA, limonene/O ₃ , catechol/O ₃ , limonene/OH, toluene/OH Humic acid, light Humic acid, dark Humic acid, illuminated Catechol (surface-absorbed) + NaCl/NaBr/NaF Gentisic acid, tannic acid, UV/Vis light Solution of guaiacol, syringol, catechol	20		< 0.5 <1.5 < 0.1 2.6, 3.7 <0.1 for pH <7, 10 for pH > 10	3 ~ 7 0.22 ~0.88	Bröske et al., 2003 Bröske et al., 2003 Stemmler et al., 2006 Stemmler et al., 2007 Stemmler et al., 2007 Woodill and Hinrichs, 2010 Sosedova et al., 2011 Ammann et al., 2005
	Summary	0.1~5.1 for solid, 20 for liquid		<0.5 for solid, 0.22~7 for liquid		
SO ₂ (× 10 ⁻⁶)	Liquid oleic acid	0.92 ~ 6.44				Shang et al., 2016 ^e
	Liquid SOA by limonene and O ₃	10 ~50				Ye et al., 2018
	Summary	0.92~10 for liquid		Not available		
N ₂ O ₅ (×10 ⁻³)	Solid malonic acid		<1.0			Thornton et al., 2003
	Solid azelaic acid		0.5			Thornton et al., 2003
	Solid oxalic acid		<0.01			Griffiths et al., 2009
	Solid oxalic acid		3.1			Griffiths et al., 2009
	Solid succinic acid		<0.6, <0.3			Griffiths et al., 2009
	Aqueous malonic acid		2.0, 30			Thornton et al., 2003
	Aqueous aerosols coated with pinene ozonolysis products		0.45~3.4			Folkers et al., 2003
	Aqueous sulfate particles coated with monoterpene ozonolysis products		11~45			Anttila et al., 2006
	Humic acid		0.1, 0.3, 1.0			Badger et al., 2006
	Malonic acid		8 ~ 16			Griffiths et al., 2009
	Succinic acid		5.2 ~ 9			Griffiths et al., 2009
	Glutaric acid		0.6 ~ 8			Griffiths et al., 2009
	Liquid oleic acid		0.054			Gross et al., 2009
	Liquid diethyl sebacate		0.051			Gross et al., 2009
	Liquid glycerol		0.645~0.814			Gross et al., 2009
	Liquid conjugated linoleic acid		0.046			Gross et al., 2009
	Liquid linoleic acid		0.168			Gross et al., 2009
	Summary	0.01~3.1 for solid, 0.05 ~ 45 for liquid		Not available		
HNO ₃ (×10 ⁻³)	Solid pyrene	≤ 6.6				Gross et al., 2008 ^e
	Summary	≤ 6.6 for solid		Not available		

- 5 ^a T=272K
 ^b T=219-298K
 ^c T=293K
 ^d T=272-298K
 ^e T=283-298K
 ^f T=278-308K
 ^g T=293-297K

Table A.4. Aerosol uptake coefficients (γ_{eff}) for reactive gases on sea salt aerosols observed in laboratory experiments (T=298±2K if not specified otherwise).

Gases (Unit)	Aerosol type	Initial γ_{eff} , geometric surface	Initial γ_{eff} , BET	Steady-state γ_{eff} , geometric surface	Steady-state γ_{eff} , BET	References
O ₃ (×10 ⁻³)	Synthetic sea salt	1.0~10				Mochida et al., 2000
	Natural sea salt	0.97				Mochida et al., 2000
	NaCl				0.0013	Il'in et al., 1991 ^a
	Deliquesced NaCl			<0.1		Abbatt and Waschewsky, 1998
	NaCl/Fe ₂ O ₃		1.3, 33~36			Sadanaga et al., 2001
	Summary	1.0~ 36		0.0013~0.1		
NO ₂ (×10 ⁻⁴)	Deliquesced NaCl	<1.0				Abbatt and Waschewsky, 1998
	Deliquesced NaCl			2.8~3.7		Harrison and Collins, 1998 ^b
	Aqueous NaCl	1.0			0.00551	Yabushita et al., 2009
	Chinese seasalt				0.0126	Ye et al., 2010
	Chinese seasalt				0.6	Ye et al., 2010
	NaCl					Vogt et al., 1994
	Summary	1.0		0.006~3.0		
SO ₂ (×10 ⁻³)	Synthetic sea salt	6.0~90		3.2~17		Gebel et al., 2000
	Summary	6.0~90		3.2~17		
N ₂ O ₅ (×10 ⁻²)	NaCl				3.2	Behnke et al., 1997
	NaCl				0.64	Stewart et al., 2004
	NaCl				0.9	Stewart et al., 2004
	NaCl				1.04	Stewart et al., 2004
	NaCl				0.078	Stewart et al., 2004
	Sea salt				1.6	Stewart et al., 2004
	Sea salt				2.8	Stewart et al., 2004
	Sea salt				1.3	Stewart et al., 2004
	Sea salt				3.1	Stewart et al., 2004
	Synthetic sea salt				2.2	Thornton and Abbatt, 2005
	Synthetic sea salt				3.0	Thornton and Abbatt, 2005
	Synthetic sea salt				2.4	Thornton and Abbatt, 2005
	NaCl				1.8	Schweitzer et al., 1998 ^c
	NaCl				1.4~3.9	George et al., 1994
	Synthetic sea salt			0.29		Hoffman et al., 2003
	Summary	Not available		0.64~3.9		
HNO ₃ (×10 ⁻²)	Deliquesced sea salt	50				Guimbaud et al., 2002
	Synthetic sea salt	6.6~75		3.0~25		De Haan and Finlayson- Pitts, 1997

	deliquesced NaCl, 100nm size	0.49				Tolocka et al., 2004
	Deliquesced NaCl	15				Saul et al., 2006
	Deliquesced NaCl	21~11				Liu et al., 2007
	NaCl/MgCl2	25~12				Liu et al., 2007
	Sea salt	27~12				Liu et al., 2007
	Deliquesced NaCl	20				Abbatt and Waschewsky, 1998
	Seliquesced NaCl	50				Stemmler et al., 2008
	Synthetic sea salt			0.04~0.065		Hoffman et al., 2003
	Summary	6.6~75		0.05-25		

^a T=235-299K
^b T = 279K
^c T=262-278K

Supporting Information of

Relative importance of gas uptake on aerosol and ground surfaces characterized by equivalent uptake coefficients

Meng Li^{1,*}, Hang Su^{2,1}, Guo Li¹, Nan Ma², Ulrich Pöschl¹, and Yafang Cheng¹

¹ Max Planck Institute for Chemistry, Mainz, 55118, Germany

² Center for Air Pollution and Climate Change Research (APCC), Institute for Environmental and Climate Research (ECI), Jinan University, Guangzhou, 511443, China

*now at: Chemical Science Division, Earth System Research Laboratory, National Oceanic and Atmospheric Administration (NOAA), Boulder, Colorado 80305, United States

Correspondence to: Hang Su (h.su@mpic.de) and Yafang Cheng (yafang.cheng@mpic.de)

Description

Calculation of dry deposition velocities above the ground surface for the investigated gases.

Tables

Table S1. Friction velocity and canopy roughness length by land use type.

Table S2. Seasonal mean dry deposition velocities by gas species, unit: cm s^{-1} .

Table S3. Seasonal γ_{eqv} by gas species at typical condition (typical aerosol area density of A as described in the main text, and mixing height of 300m), $\times 10^{-4}$.

Table S4. Examples of aerosol uptake coefficients used in atmospheric models.

Figures

Figure S1. Resistance model for dry deposition, derived from the Figure 19.1 in Seinfeld and Pandis (2006).

Figure S2. Schematic model for canopy resistance, derived from Wesely (1989).

1. Calculation of dry deposition velocities above the ground surface for gases

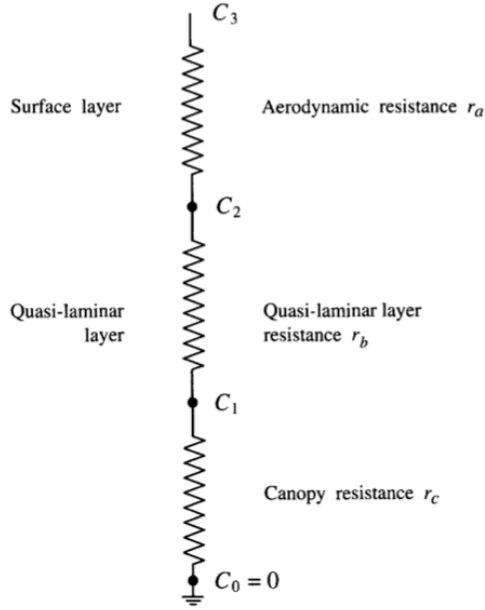


Figure S1. Resistance model for dry deposition, derived from the Figure 19.1 in Seinfeld and Pandis (2006).

$$V_d = \frac{1}{R_a + R_b + R_c}$$

Following Wesely (1989) and Zhang et al. (2003), we calculated R_a (aerodynamic resistance), R_b (quasi-laminar layer resistance) and R_c (canopy resistance) as below.

1.1 calculation of R_a

Under neutral atmospheric condition:

$$R_a = \int_{Z_0}^Z \frac{1}{\kappa u_* z} dz = \frac{1}{\kappa u_*} \ln\left(\frac{Z}{Z_0}\right)$$

where κ is the von Karman constant (about 0.41); u_* means the friction velocity (in unit of m s^{-1}); Z_0 is the roughness length (in unit of m); Z is the PBL mixing height (in unit of m), we use a typical value of Z as 300 m.

For different land use type, we assign different u_* following the parameterization scheme of Zhang et al. (2003), and Z_0 based on Seinfeld and Pandis (2006):

Table S1. Friction velocity and canopy roughness length by land use type.

land use index	land use type	u_{*_day} (m s ⁻¹)	u_{*_night} (m s ⁻¹)	Z_0 (m)
1	urban land	0.6	0.3	1
2	agricultural land	0.4	0.2	0.1
3	range land	0.4	0.2	0.1
4	deciduous forest	0.6	0.3	1
5	coniferous forest	0.6	0.3	0.9
6	mixed forest including wetland	0.6	0.3	0.9
7	water	0.3	0.25	0.1
8	barren land, desert	0.25	0.15	0.04
9	non-forested wetland	0.25	0.2	0.1
10	mixed agricultural and range land	0.4	0.2	0.1
11	rocky open areas with low-growing shrubs	0.4	0.2	0.1
12	amazon forest	0.6	0.3	1

1.2 calculation of R_b

$$R_b = \frac{5Sc^{2/3}}{u_*} \quad Sc = \frac{\nu}{D}$$

where Sc is the Schmidt number (unitless), ν means the kinematic viscosity of air, D is the molecular diffusivity for gases. We use $D=10^{-5}$ m² s⁻¹, and a temperature-dependent ν in the calculation.

1.3 calculation of R_c

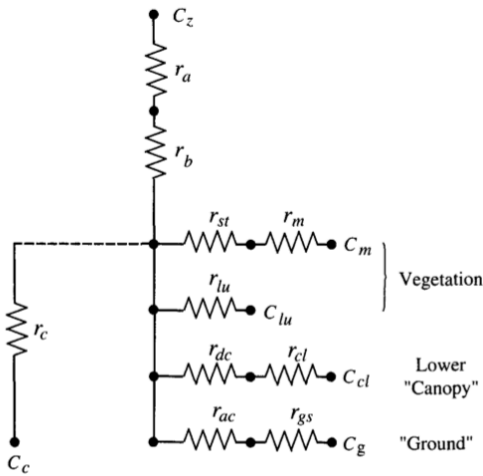


Figure S2. Schematic model for canopy resistance, derived from Wesely (1989).

$$R_c = \left(\frac{1}{R_{st} + R_m} + \frac{1}{R_{lu}} + \frac{1}{R_{dc} + R_{cl}} + \frac{1}{R_{ac} + R_{gs}} \right)^{-1}$$

As illustrated by Seinfeld and Pandis (2006), R_{st} represents the resistance of the leaf stomatal, R_m for mesophyll resistance, R_{lu} is the surface resistance in the upper canopy, R_{dc} means the resistance by buoyant convection, R_{cl} is the uptake resistance by leaves, twigs and etc., R_{ac} means the transfer resistance for processes at the ground, and R_{gs} is the uptake resistance by soil, leaf litter and others on the ground surface.

The equations to calculate each item of R_c are illustrated in Wesely (1989). The important input parameters for R_c calculation include: the input resistance by land use and season, the physical and chemical reactivity scales by gas species, and the meteorological parameters. We adopted the parameterization scheme of Wesely (1989) for the former two items of input, and a set of typical hourly temperature and radiation values for each season derived from the standard meteorological database for construction in China (Zhang, 2004).

The calculated dry deposition velocities by gas and land use type for each season are presented in Table S2. Furthermore, we show the seasonal equivalent uptake coefficients (γ_{eq}) at typical conditions based on the dry deposition velocities in Table S3.

Table S2. Seasonal mean dry deposition velocities by gas species, unit: cm s^{-1} .

Gases	Winter	Spring	Summer	Autumn with unharvested cropland	Late autumn after frost
Urban					
O ₃	0.15	0.24	0.24	0.24	0.24
NO ₂	0.02	0.04	0.04	0.04	0.04
SO ₂	0.41	0.17	0.20	0.20	0.20
N ₂ O ₅	2.10	2.26	2.31	2.14	2.14
HNO ₃	2.10	2.26	2.31	2.14	2.14
H ₂ O ₂	0.44	0.32	0.34	0.33	0.33
Agricultural land					
O ₃	0.07	0.45	0.44	0.29	0.41
NO ₂	0.02	0.20	0.29	0.07	0.07
SO ₂	0.49	0.43	0.42	0.24	0.40
N ₂ O ₅	1.10	1.18	1.21	1.12	1.12
HNO ₃	1.10	1.18	1.21	1.12	1.12
H ₂ O ₂	0.51	0.57	0.51	0.34	0.57
Amazon forest					
O ₃	0.15	0.27	0.37	0.14	0.14
NO ₂	0.04	0.17	0.28	0.04	0.04
SO ₂	0.14	0.25	0.34	0.14	0.14
N ₂ O ₅	2.10	2.26	2.31	2.14	2.14

HNO ₃	2.10	2.26	2.31	2.14	2.14
H ₂ O ₂	0.23	0.37	0.48	0.23	0.23
			Water		
O ₃	0.07	0.07	0.07	0.06	0.06
NO ₂	0.01	0.01	0.01	0.01	0.01
SO ₂	0.03	0.03	0.03	0.03	0.03
N ₂ O ₅	1.05	1.06	1.07	1.05	1.05
HNO ₃	1.05	1.06	1.07	1.05	1.05
H ₂ O ₂	0.08	0.08	0.08	0.08	0.08

Table S3. Seasonal γ_{eqv} by gas species at typical condition (typical aerosol area density of A as described in the main text, and mixing height of 300m), $\times 10^{-4}$.

Gases	Winter	Spring	Summer	Autumn with unharvested cropland	Late autumn after frost
Urban					
O ₃	0.64	1.01	1.01	1.00	1.00
NO ₂	0.10	0.15	0.15	0.15	0.15
SO ₂	1.73	0.72	0.84	0.83	0.83
N ₂ O ₅	8.91	9.59	9.78	9.07	9.07
HNO ₃	8.91	9.59	9.78	9.07	9.07
H ₂ O ₂	1.87	1.37	1.44	1.41	1.41
Agricultural land					
O ₃	1.31	8.76	8.55	5.63	7.93
NO ₂	0.30	3.88	5.57	1.38	1.36
SO ₂	9.45	8.39	8.12	4.55	7.69
N ₂ O ₅	21.21	22.88	23.37	21.65	21.65
HNO ₃	21.21	22.88	23.37	21.65	21.65
H ₂ O ₂	9.80	10.97	9.94	6.61	11.06
Amazon forest					
O ₃	14.01	25.72	35.56	13.78	13.78
NO ₂	3.88	16.54	27.18	3.56	3.56
SO ₂	13.96	24.22	33.03	13.76	13.76
N ₂ O ₅	203.33	218.79	223.13	207.08	207.08
HNO ₃	203.33	218.79	223.13	207.08	207.08
H ₂ O ₂	22.43	35.42	45.95	22.25	22.25
Water					
O ₃	3.92	3.81	3.82	3.76	3.76
NO ₂	0.70	0.53	0.53	0.49	0.49
SO ₂	1.76	1.66	1.67	1.61	1.61
N ₂ O ₅	61.37	62.24	62.36	61.33	61.33
HNO ₃	61.37	62.24	62.36	61.33	61.33
H ₂ O ₂	4.68	4.61	4.63	4.51	4.51

Table S4. Examples of aerosol uptake coefficients used in atmospheric models^a.

Gases	Aerosol type	γ_{eff} (Liao and Seinfeld, 2005)	References	γ_{eff} (Zhu et al., 2010)	References	γ_{eff} (Wang et al., 2012)	References
O ₃	Mineral dust	1.0×10^{-5}	Michel et al., 2002, 2003	2.7×10^{-5}	IUPAC ^c	$5.0 \times 10^{-5} \sim 1.0 \times 10^{-4}$	Dentener et al., 1996 ^b ; Zhang and Carmichael, 1999 ^b
NO ₂	Mineral dust			2.1×10^{-6}	IUPAC	$4.4 \times 10^{-5} \sim 2.0 \times 10^{-4}$	Underwood et al., 2001
	Wet aerosol	1.0×10^{-4}	Jacob, 2000				
SO ₂	Mineral dust	3.0×10^{-4} (RH<50%), 0.1 (RH≥50%)	Dentener et al., 1996	3.0×10^{-5}	IUPAC	$1.0 \times 10^{-4} \sim 2.6 \times 10^{-4}$	Zhang and Carmichael, 1999 ^b
	Sea salt aerosol	5.0×10^{-3} (RH<50%), 5.0×10^{-2} (RH≥50%)	Song and Carmichael, 2001 ^b				
N ₂ O ₅	Mineral dust	See footnote ^c	Bauer et al., 2004 ^b	3.0×10^{-2}	Seisel et al., 2005; Wagner et al., 2008; Karagulian et al., 2006	$1.0 \times 10^{-3} \sim 0.1$	Dentener et al., 1996; DeMore et al., 1997
	Organic carbon	$5.2 \times 10^{-4} \times$ RH (RH<50%), 0.03 (RH≥50%)	Thornton et al., 2003				
	Sea salt aerosol	5×10^{-3} (RH<50%), 0.03 (RH≥50%)	Atkinson et al., 2004				

	Sulfate/nitrate/ammonium	See footnote ^d	Kane et al., 2001, Hallquist et al., 2003				
HNO ₃	Mineral dust	0.1	Hanisch and Crowley, 2001	0.17	IUPAC	1.1×10 ⁻³ ~ 0.2	Dentener et al., 1996; DeMore et al., 1997; Underwood et al., 2001
H ₂ O ₂	Mineral dust			2.0×10 ⁻³	De Reus et al., 2005	1.0×10 ⁻⁴ ~ 2.0×10 ⁻³	Dentener et al., 1996

^a Here we present two parameterization schemes as examples: the full scheme of Liao and Seinfeld (2005), the scheme for mineral dust of Zhu et al. (2010) and Wang et al. (2012). The original references of the measurements regarding the uptake coefficients are listed. It should be addressed that these schemes are only examples of modelling studies.

^b Model parameterization. The specific references to laboratory measurements for uptake coefficients are not found.

$$^c \gamma = 4.25 \times 10^{-4} \times RH - 9.75 \times 10^{-3}$$

$$^d \gamma = 10^{\beta(T)} \times (C_1 + C_2 \times RH + C_3 \times RH^2 + C_4 \times RH^3)$$

$$\beta(T) = -4 \times 10^{-2} \times (T - 294), T \geq 282K$$

$$\beta(T) = 0.48, T < 282K$$

$$C_1 = 2.79 \times 10^{-4}; C_2 = 1.30 \times 10^{-4}; C_3 = -3.43 \times 10^{-6}; C_4 = 7.52 \times 10^{-8}$$

^e IUPAC: International Union of Pure and Applied Chemistry, available at <http://iupac.pole-ether.fr/>

Reference

- Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson Jr., R. F., Jenkin, M. E., Kerr, J. A., Rossi, M. J., and Troe, J.: Summary of evaluated kinetic and photochemical data for atmospheric chemistry, Cambridge Univ., Cambridge, UK., 2004 (Available at http://www.iupac-kinetic.ch.cam.ac.uk/summary/IUPACsumm_web_latest.pdf)
- Bauer, S. E., Balkanski, Y., Schulz, M., Hauglustaine, D. A., and Dentener, F.: Global modeling of heterogeneous chemistry on mineral aerosol surfaces: Influence on tropospheric ozone chemistry and comparison to observations, *Journal of Geophysical Research: Atmospheres*, 109, n/a-n/a, 10.1029/2003JD003868, 2004.
- de Reus, M., Fischer, H., Sander, R., Gros, V., Kormann, R., Salisbury, G., Van Dingenen, R., Williams, J., Zöllner, M., and Lelieveld, J.: Observations and model calculations of trace gas scavenging in a dense Saharan dust plume during MINATROC, *Atmos. Chem. Phys.*, 5, 1787-1803, 10.5194/acp-5-1787-2005, 2005.
- DeMore, W. B., Sander, S. P., Golden, D. M., Hampson, R. F., Kurylo, M. J., Howard, C. J., Ravishankara, A. R., Kolb, C. E., and Molina, M. J.: Chemical kinetics and photochemical data for use in stratospheric modeling, Eval. 12 97 - 4, Jet Propul. Lab., Pasadena, Calif., 1997.
- Dentener, F. J., Carmichael, G. R., Zhang, Y., Lelieveld, J., and Crutzen, P. J.: Role of mineral aerosol as a reactive surface in the global troposphere, *Journal of Geophysical Research: Atmospheres*, 101, 22869-22889, 10.1029/96JD01818, 1996.
- Hallquist, M., Stewart, D. J., Stephenson, S. K., and Anthony Cox, R.: Hydrolysis of N₂O₅ on sub-micron sulfate aerosols, *Physical Chemistry Chemical Physics*, 5, 3453-3463, 10.1039/B301827J, 2003.
- Hanisch, F., and Crowley, J. N.: Heterogeneous Reactivity of Gaseous Nitric Acid on Al₂O₃, CaCO₃, and Atmospheric Dust Samples: A Knudsen Cell Study, *The Journal of Physical Chemistry A*, 105, 3096-3106, 10.1021/jp001254+, 2001.
- Jacob, D. J.: Heterogeneous chemistry and tropospheric ozone, *Atmospheric Environment*, 34, 2131-2159, [https://doi.org/10.1016/S1352-2310\(99\)00462-8](https://doi.org/10.1016/S1352-2310(99)00462-8), 2000.
- Kane, S. M., Caloz, F., and Leu, M.-T.: Heterogeneous Uptake of Gaseous N₂O₅ by (NH₄)₂SO₄, NH₄HSO₄, and H₂SO₄ Aerosols, *The Journal of Physical Chemistry A*, 105, 6465-6470, 10.1021/jp010490x, 2001.
- Karagulian, F., Santschi, C., and Rossi, M. J.: The heterogeneous chemical kinetics of N₂O₅ on CaCO₃ and other atmospheric mineral dust surrogates, *Atmos. Chem. Phys.*, 6, 1373-1388, 10.5194/acp-6-1373-2006, 2006.

- Liao, H., and Seinfeld, J. H.: Global impacts of gas-phase chemistry-aerosol interactions on direct radiative forcing by anthropogenic aerosols and ozone, *Journal of Geophysical Research: Atmospheres*, 110, n/a-n/a, 10.1029/2005JD005907, 2005.
- Michel, A. E., Usher, C. R., and Grassian, V. H.: Heterogeneous and catalytic uptake of ozone on mineral oxides and dusts: A Knudsen cell investigation, *Geophys. Res. Lett.*, 29, 10-11-10-14, 10.1029/2002GL014896, 2002.
- Michel, A. E., Usher, C. R., and Grassian, V. H.: Reactive uptake of ozone on mineral oxides and mineral dusts, *Atmospheric Environment*, 37, 3201-3211, [https://doi.org/10.1016/S1352-2310\(03\)00319-4](https://doi.org/10.1016/S1352-2310(03)00319-4), 2003.
- Seinfeld, J. H., and Pandis, S. N.: *Atmospheric chemistry and physics: From air pollution to climate change*, John Wiley & Sons, New York, USA, 2006.
- Seisel, S., Börensén, C., Vogt, R., and Zellner, R.: Kinetics and mechanism of the uptake of N₂O₅ on mineral dust at 298 K, *Atmos. Chem. Phys.*, 5, 3423-3432, 10.5194/acp-5-3423-2005, 2005.
- Song, C. H., and Carmichael, G. R.: A three-dimensional modeling investigation of the evolution processes of dust and sea-salt particles in east Asia, *Journal of Geophysical Research: Atmospheres*, 106, 18131-18154, doi:10.1029/2000JD900352, 2001.
- Thornton, J. A., Braban, C. F., and Abbatt, J. P. D.: N₂O₅ hydrolysis on sub-micron organic aerosols: the effect of relative humidity, particle phase, and particle size, *Physical Chemistry Chemical Physics*, 5, 4593-4603, 10.1039/B307498F, 2003.
- Underwood, G. M., Li, P., Al-Abadleh, H., and Grassian, V. H.: A Knudsen Cell Study of the Heterogeneous Reactivity of Nitric Acid on Oxide and Mineral Dust Particles, *The Journal of Physical Chemistry A*, 105, 6609-6620, 10.1021/jp002223h, 2001a.
- Wagner, C., Hanisch, F., Holmes, N., de Coninck, H., Schuster, G., and Crowley, J. N.: The interaction of N₂O₅ with mineral dust: aerosol flow tube and Knudsen reactor studies, *Atmos. Chem. Phys.*, 8, 91-109, 10.5194/acp-8-91-2008, 2008.
- Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, *Atmos. Chem. Phys.*, 12, 10209-10237, 10.5194/acp-12-10209-2012, 2012.
- Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, *Atmospheric Environment* (1967), 23, 1293-1304, [https://doi.org/10.1016/0004-6981\(89\)90153-4](https://doi.org/10.1016/0004-6981(89)90153-4), 1989.
- Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, *Atmos. Chem. Phys.*, 3, 2067-2082, 10.5194/acp-3-2067-2003, 2003.

Zhang, Q.: China Standard Meteorological Database for Construction (CD-ROM), China Mechanical Industry Press, Beijing, 2004.

Zhang, Y., and Carmichael, G. R.: The Role of Mineral Aerosol in Tropospheric Chemistry in East Asia—A Model Study, *Journal of Applied Meteorology*, 38, 353-366, 10.1175/1520-0450(1999)038<0353:TROMAI>2.0.CO;2, 1999.

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.