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

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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
15 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
20 conditions. In urban areas, aerosol uptake is relevant for all species ($\gamma_{eff} \geq \gamma_{eqv}$) 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 $\gamma_{eff} \geq \gamma_{eqv}$. 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

5 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

10 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 μm^2 cm^{-3}, comparable to aerosol surface area concentrations of 200 ~ 2000 μm^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 ~

15 1000 μm^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.

20 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

25 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

30 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:

$$F_{grd} = -V_d [X_g] \times 10^{-2} \tag{1}$$

$$V_d = \frac{1}{R_{grd}} = \frac{1}{R_a + R_b + R_c} \tag{2}$$

where F_{grd} represents the gas deposition fluxes on various ground surfaces (mol m^{-2} 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}, mol m^{-2} s^{-1}) for one aerosol particle can be expressed as Eq. (3) under (quasi-) steady-state conditions (Pöschl et al., 2007):

$$J_{net} = \frac{\omega \gamma_{eff}}{4} [X_g] \tag{3}$$

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}).

$$\frac{1}{\gamma_{eff}} = \frac{1}{\Gamma_g} + \frac{1}{\alpha} + \frac{1}{\Gamma_b} \tag{4}$$

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 $\Gamma_g = 8 D_g \omega^{-1} d_p^{-1}$, where D_g is the diffusion coefficient of gas X in the gas phase (m^2 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 ~0.2 μm, 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, $\mu m^2\ cm^{-3}$), the total uptake flux of gas X by aerosols (F_{aer}, $mol\ m^{-2}\ s^{-1}$) is:

$$F_{aer} = J_{net}Ah = \frac{\omega\gamma_{eff}}{4}Ah[X_g]\times10^{-6} \tag{5}$$

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 (~298K) 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 $\gamma_{eff} > \gamma_{eqv}$, 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.

$$\gamma_{eqv} = \frac{4}{3}\frac{V_d}{Ah}\times10^2 \tag{6}$$

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

$$\gamma_{eqv} = \frac{V_d}{2.25A} \tag{7}$$

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 ~ 2000 μm^2 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 ~ 1000 μm^2 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 ~ 700 μm^2 cm^{-3} in Amazon forest (Zhou et al., 2002; Rissler et al., 2006; Pöschl et al., 2010; Andreae et al. 2015), and 20 ~ 200 μm^2 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 μm^2 cm^{-3} for the urban environment (Wang et al., 2017), 230 μm^2 cm^{-3} for the agricultural land (Held et al., 2008), 46 μm^2 cm^{-3} for the Amazon forest (Rissler et al., 2006), and 76 μm^2 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 ~ 2.3 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 μm^2 cm^{-3} to 2139 μm^2 cm^{-3}, from pristine rainforest to polluted megacities. We show the calculated γ_{eqv} at typical conditions (typical A as described above, h=300m) 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 (≤ 0.1 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

5   increasing importance of gas uptake in polluted areas than clean areas.

With the increase of V_d (> 0.1 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 µm^2 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

10   use types (see Table S2). Thus we can derive a general criterion of $\gamma_{eff} > 10^{-5}$ 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,

15   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=300m, 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

20   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 µm^2 m^{-3}, Beijing) and/or high mixing layers (e.g., h=1.0 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

25   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

30   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 μm^2 cm^{-3} (Amazon) to 1050 μm^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

5   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 μm^2 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.

10   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

15   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

20   under favorable conditions over agricultural land and water body. For soot, aerosol uptake is magnitudes lower than those on the ground ($\gamma_{eqv} \geq \gamma_{eff}$), 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

25   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

30   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 SO_2 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 O_3, H_2O_2, 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 SO_2 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 SO_2 uptake and sulfate formation may transit from TMI- or H_2O_2-dominated regime to NO_2- or O_3-dominated regime (Cheng et al., 2016). In this case, the SO_2 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 SO_2 on calcareous soils, cements and Fe_2O_3, 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 SO_2 by O_3 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 N_2O_5, HNO_3, and H_2O_2

N_2O_5, HNO_3, and H_2O_2 demonstrate their high uptake ability on atmospheric aerosols, as shown in Fig. 5. For N_2O_5, 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 N_2O_5 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 μm 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 1.0×10^{-5}~9.4×10^{-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 (~298K). 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} ~ 10^{-4} in polluted urban environments and 10^{-4} ~ 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 ($\gamma_{eff} \geq \gamma_{eqv}$, 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 ($\gamma_{eff} \geq \gamma_{eqv}$). 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.

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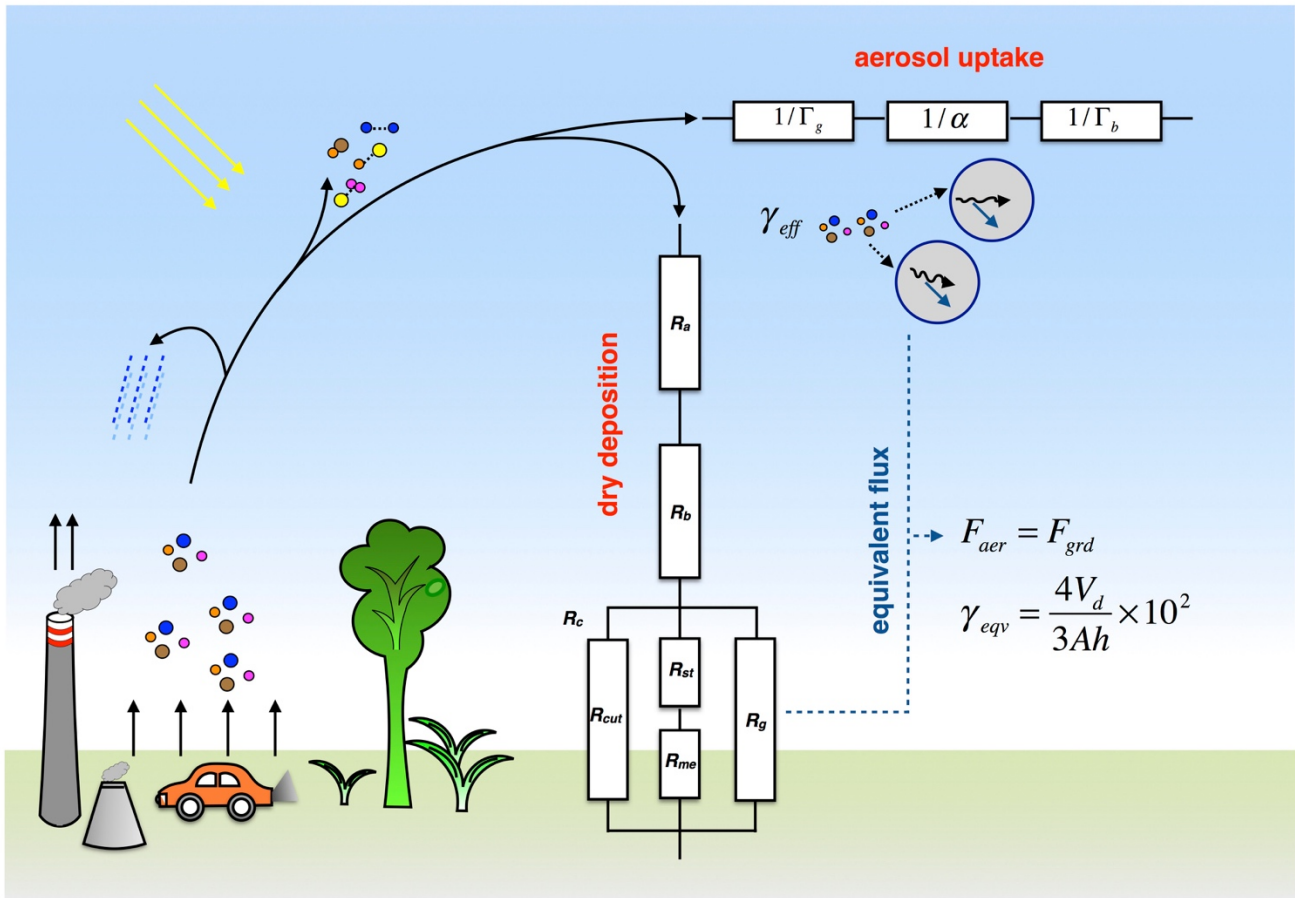
Figure 1. Schematic illustration of gas uptake on the ground and on aerosols in the planetary boundary layer as characterized by resistance models. The relative importance of aerosol uptake and dry deposition on the ground is characterized through comparing the aerosol uptake coefficient (γ_{eff}) with an equivalent uptake coefficient (γ_{eqv}) corresponding to the deposition velocity (V_d).
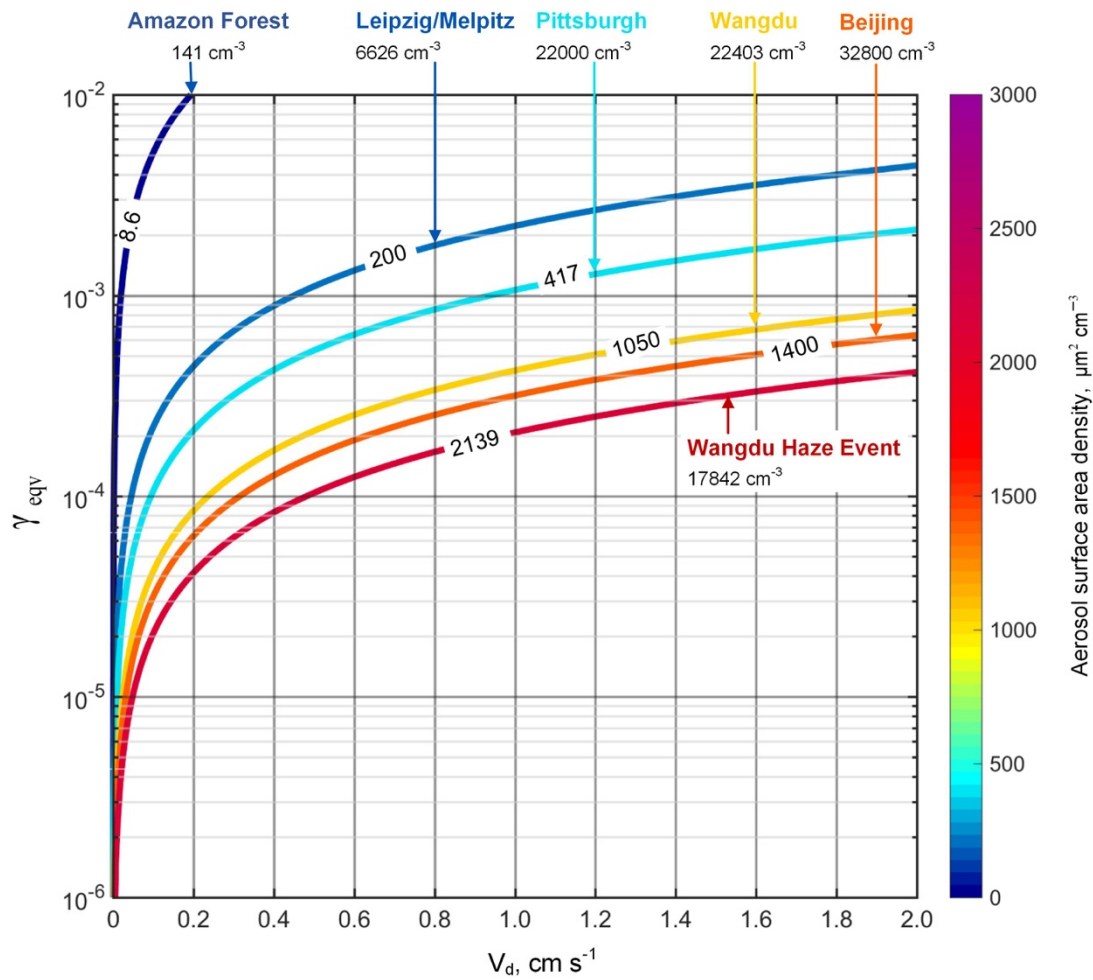
Figure 2. Relation between γ_{eqv} and V_d for a mixing height of 300 m and aerosol surface area densities (*A*) observed at locations and conditions: Amazon Forest (Pöschl et al., 2010), Leipzig/Melpitz (Ma et al., 2014; Ma and Birmili, 2015), Pittsburgh (Stanier et al., 2004), Wangdu with and without 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.
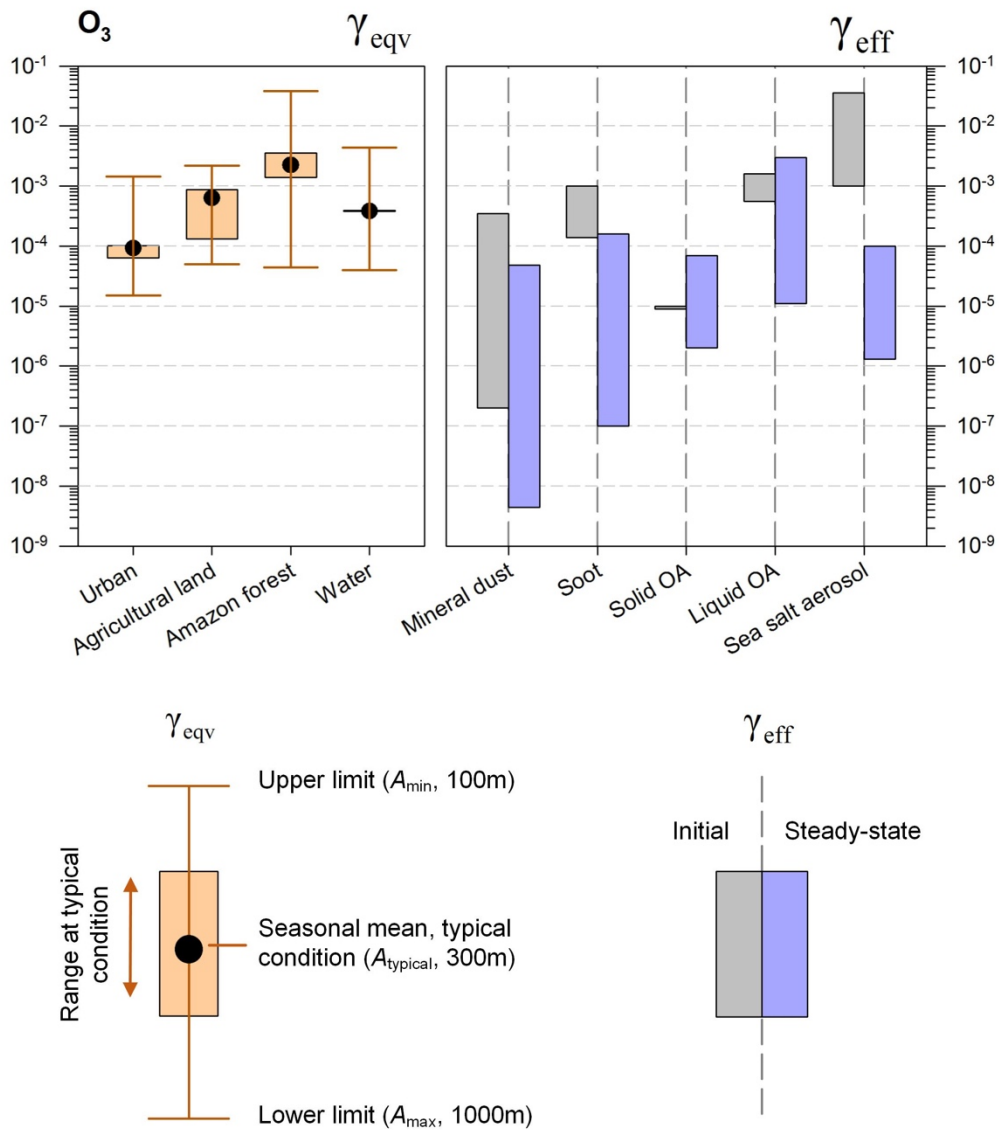
Figure 3. Equivalent uptake coefficients (γ_{eqv}, left) and laboratory measurement values (γ_{eff}, right) for O_3 on different ground types and aerosols.

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.
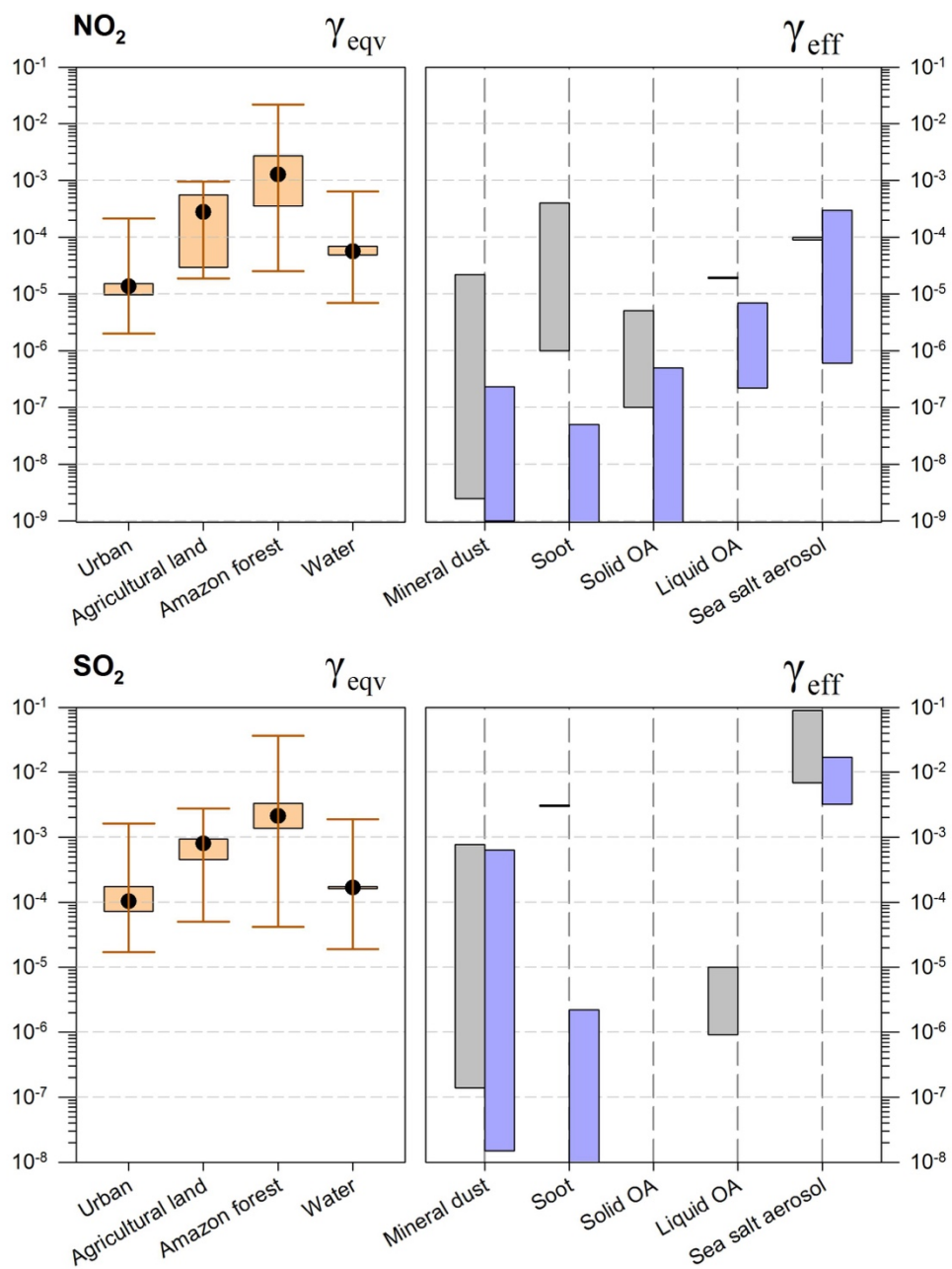
Figure 4. Uptake coefficients (γ_{eqv}, left; γ_{eff}, right) for NO$_2$ and SO$_2$ on different ground types and aerosols.
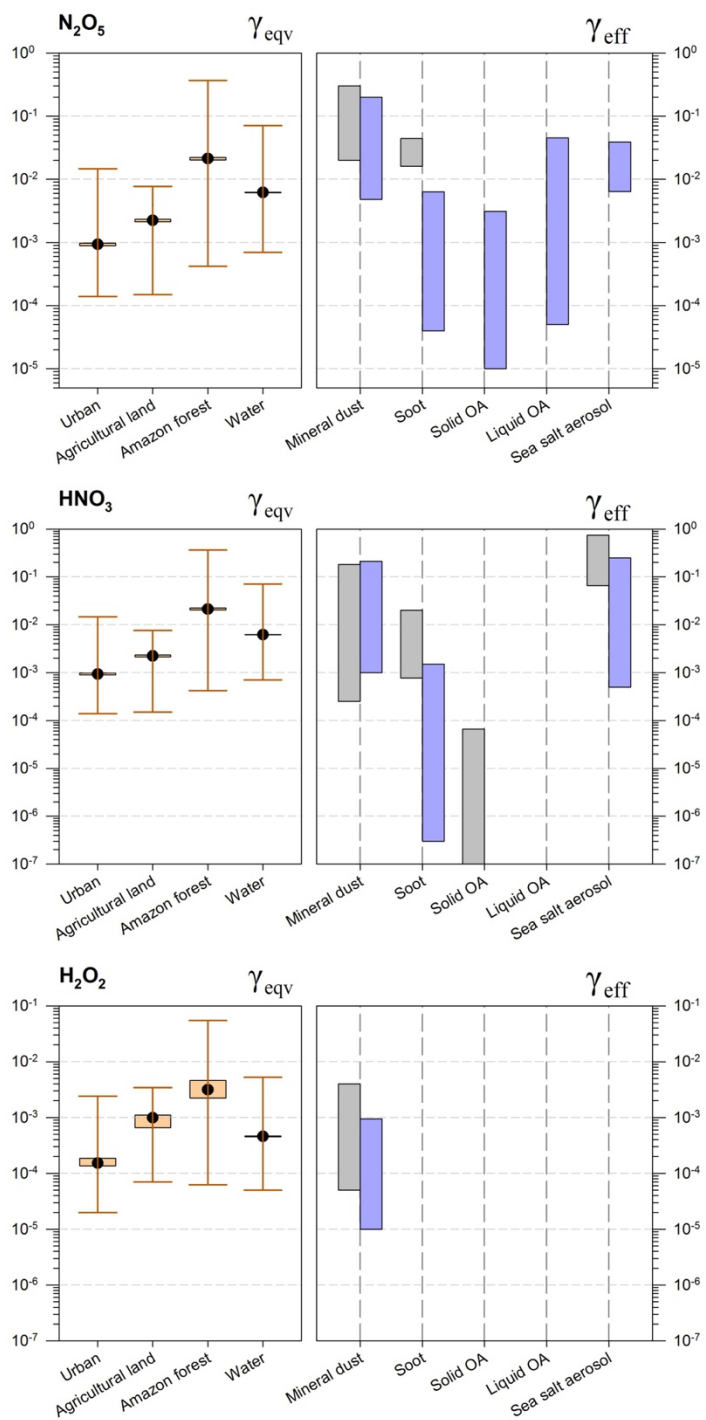
Figure 5. Uptake coefficients (γ_{eqv}, left; γ_{eff}, right) for N_2O_5, HNO_3 and H_2O_2 on different ground types and aerosols.

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_2O_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_2O_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	$9.2 \times 10^{-7} - 1.0 \times 0^{-5}$	$6.9 \times 10^{-3} - 9.0 \times 10^{-2}$
N_2O_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_2O_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_2O_5	√	√	√	√	√
HNO_3	√				√
H_2O_2	√				

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			0.3~3	0.02~0.32	Nicolas et al., 2009
	Al_2O_3, Fe_2O_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_2O_3 and others		0.27~20		0.6~2.2	Michel et al., 2003
	Saharan dust	0.55 ~35		0.22~4.8		Hanisch and Crowley, 2003
	Al_2O_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_2O_3				0.013~0.26	Börensen et al., 2000
	Al_2O_3 and others		0.2~220			Underwood et al., 2001b
	Saharan dust		6.2			Ullerstam et al., 2003
	Illuminated TiO_2			9400, 1200		Gustafsson et al., 2006
	$CaCO_3$	0.656	0.025~ 0.043			Li et al., 2010
	Kaolinite, pyrophylite				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

Species	Material					Reference
	Al₂O₃, MgO		9.5, 26			Goodman et al., 2001
	Al₂O₃ 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
	Al₂O₃ Fe₂O₃, MgO		40, 55, 100			Judeikis et al, 1978
	CaCO₃ powder		0.1			Santschi and Rossi, 2006
	CaCO₃, 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_2O_2			39.1~54.5		Huang et al., 2015
	Tengger desert dust, H_2O_2			37.2~63.1		Huang et al., 2015
	Arizona test dust, H_2O_2			4.6~13.1		Huang et al., 2015
	Summary	0.014~77		0.0015~63.1		
N_2O_5 ($\times10^{-2}$)	Saharan sand	30		20		Karagulian et al., 2006
	Arizona test dust	20		11		Karagulian et al., 2006
	CaCO₃	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
	CaCO₃	5.0				Wagner et al., 2008
	CaCO₃			0.48~0.53		Wagner et al., 2009
	CaCO₃			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		
HNO₃	Mineral dust			1.7~5.4		Umann et al., 2005

(×10⁻²)	Material					Reference
	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_2O_3 and others		0.002~0.0097			Underwood et al., 2001a
	Al_2O_3 and others		0.002~0.61			Underwood et al., 2001b
	Fe_2O_3		0.0015			Frinak et al., 2004
	Al_2O_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_2O_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]
	$TiO2$, dark		2.5~40			Romanias et al., 2012
	$TiO2$, UV				35	Romanias et al., 2012
	Al_2O_3		9.0			Romanias et al., 2013
	Fe_2O_3		8.6			Romanias et al., 2013
	SiO_2				0.000155, 0.000061	Zhao et al., 2011
	Al_2O_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$_3$($\times10^{-4}$)	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 ($\times 10^{-5}$)	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 (meet-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 ($\times 10^{-6}$)	Soild benzophenone, catechol, anthracene, anthrarobin	0.07~1.26 (dark), 0.65~2.40 (light)				George et al., 2005[f]
	Soild benzophenone, catechol, anthracene, anthrarobin	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/O₃			< 0.5		Bröske et al., 2003
	SOA, limonene/O₃, catechol/O₃, limonene/OH, toluene/OH			<1.5		Bröske et al., 2003
	Humic acid, light	20				Stemmler et al., 2006
	Humic acid, dark			< 0.1		Stemmler et al., 2007
	Humic acid, illuminated			2.6, 3.7		Stemmler et al., 2007
	Catechol (surface-absorbed) + NaCl/NaBr/NaF				3 ~ 7	Woodill and Hinrichs, 2010
	Gentisic acid, tannic acid, UV/Vis light				0.22 ~0.88	Sosedova et al., 2011
	Solution of guaiacol, syringol, catechol			<0.1 for pH <7 , 10 for pH > 10		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[c]
	Liquid SOA by limonene and O₃	10 ~50				Ye et al., 2018
	Summary	0.92~10 for liquid		Not available		
N_2O_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 (×10⁻⁵)	Solid pyrene	≤ 6.6				Gross et al., 2008[g]
	Summary	≤ 6.6 for solid		Not available		

[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 ($\times10^{-3}$)	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 ($\times10^{-4}$)	Deliquesced NaCl	<1.0				Abbatt and Waschewsky, 1998
	Deliquesced NaCl			2.8~3.7		Harrison and Collins, 1998[b]
	Aqueous NaCl	1.0				Yabushita et al.,2009
	Chinese seasalt				0.00551	Ye et al., 2010
	Chinese seasalt				0.0126	Ye et al., 2010
	NaCl				0.6	Vogt et al., 1994
	Summary	1.0		0.006~3.0		
SO_2 ($\times10^{-3}$)	Synthetic sea salt	6.0~90		3.2~17		Gebel et al., 2000
	Summary	6.0~90		3.2~17		
N_2O_5 ($\times10^{-2}$)	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 ($\times10^{-2}$)	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