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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 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 and H_2O_2), aerosol types (mineral dust, soot, organic aerosol and sea salt aerosol), environments (urban areas, agricultural land, the Amazon forest and water bodies), seasons and mixing heights.

For all investigated gases, γ_{eqv} ranges from magnitudes of 10^{-6}–10^{-4} in polluted urban environments to 10^{-4}–10^{-1} under pristine forest 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 the Amazon forest are minor compared with the dry deposition. The 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 are yet to be measured under laboratory conditions and evaluated.

Given the fact that most models have considered the uptakes of these species on the ground surface, we suggest also considering the following processes in atmospheric models: 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.

1 Introduction

Multiphase processes play an essential role in atmospheric chemistry and atmosphere–biosphere exchange (Ravishankara, 1997; Ammann et al., 1998, 2013; 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; Herrmann et al., 2015; Pöhlker et al., 2012; 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; Fröhlich-Nowoisky et al., 2016; Lappalainen et al., 2016; Tang et al., 2016; Meusel et al., 2018). They not only affect the atmospheric trace gas concentrations but also modify 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 (PBL), aerosols and the ground provide two kinds of surfaces for multiphase reactions. In previous gas uptake stud-

ies, different formulations have been used to describe and parameterize the gas uptake processes (Wesely, 1989, 2007; Ravishankara, 1997; Jacob, 2000; Wesely and Hicks, 2000; Zhang et al., 2003; Ammann and Pöschl, 2007; Pöschl et al., 2007).

A variety of ground surfaces, including vegetation, water, rock, roads and so on, can take up gaseous species via dry deposition, and, thus, have 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 units of cm s^{-1}) calculated as the reciprocal of the total resistance, is the key parameter to describe the uptake fluxes on the ground.

Since the 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 physiochemical properties of atmospheric aerosols (Kolb et al., 2010). Taking the multiphase chemistry into account has proven to be 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; X. Wang, et al., 2012; B. Zheng et al., 2015; Tang et al., 2017; Chen et al., 2018; Mu et al., 2018). Compared with 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 with an electrical circuit that decoupled the physiochemical 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 an effective uptake coefficient (γ_{eff}) has been widely used in current atmospheric models (Jacob, 2000; Liao and Seinfeld, 2005; K. Wang 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$_2$O$_5$, 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, the question still remains as to which surface types are more important for gas uptake in the PBL. The answer is not straightforward due to the following reasons:

1. First, although 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}, which is 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 200–1000 μm^2 cm^{-3} for rural environments (Ma et al., 2014; Ma and Birmili, 2015; Wu et al., 2017; Held et al., 2008).

2. Second, 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, whereas 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, especially those processes that have not yet been sufficiently considered in atmospheric models. Section 2 describes the methods of calculation and comparison. We present and discuss the main results in Sect. 3, which is followed by a summary of our major findings in Sect. 4.

2 Methods

In this work, we compared the relative importance of gas uptake by the ground and by 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$_2$O$_5$, HNO$_3$ and H$_2$O$_2$) were calculated and compared for four typical land use categories (urban areas, agricultural land, the Amazon forest and water) and five aerosol types (mineral dust, soot, solid organic aerosol, liquid organic aerosol and sea salt aerosol). These species were chosen considering their potential impor-
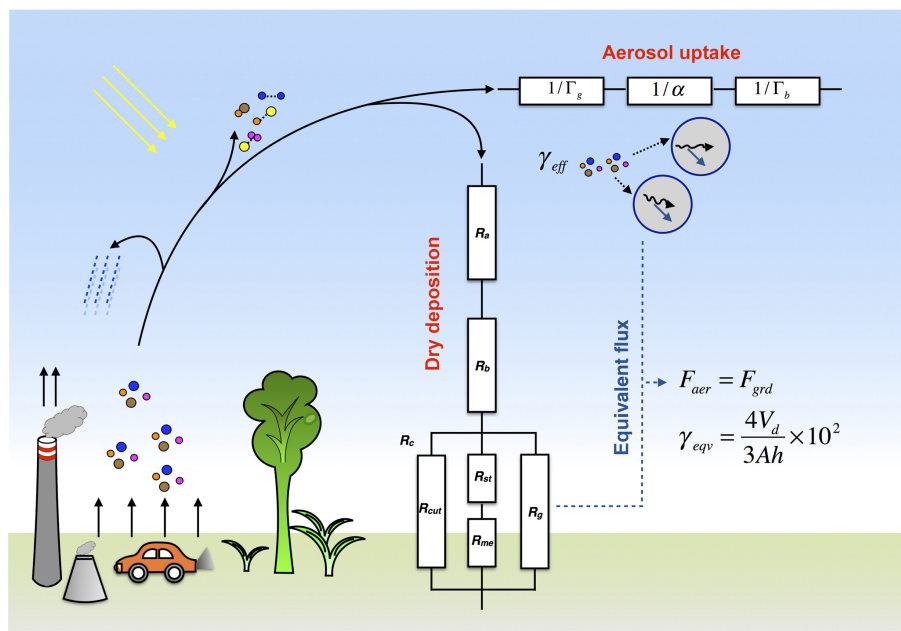
Figure 1. Schematic illustration of gas uptake on the ground and on aerosols in the planetary boundary layer as characterized by resistance models. The relative importance of aerosol uptake and dry deposition on the ground is characterized by comparing the aerosol uptake coefficient (γ_{eff}) with an equivalent uptake coefficient (γ_{eqv}) corresponding to the deposition velocity (V_{d}).

tance 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 as follows:

$$F_{\text{grd}} = -V_{\text{d}}\left[X_{\text{g}}\right] \times 10^{-2} \tag{1}$$

$$V_{\text{d}} = \frac{1}{R_{\text{grd}}} = \frac{1}{R_{\text{a}} + R_{\text{b}} + R_{\text{c}}}, \tag{2}$$

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}); $[X_{\text{g}}]$ is the averaged gas concentration (mol m^{-3}); and 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 the 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 Tables S1 and S2, respectively.

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

The net flux of gas X from the 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):

$$J_{\text{net}} = \frac{\omega \gamma_{\text{eff}}}{4}\left[X_{\text{g}}\right] \tag{3}$$

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; $[X_{\text{g}}]$ is the averaged gas concentration far away from the aerosol surface (mol m^{-3}).

$$\frac{1}{\gamma_{\text{eff}}} = \frac{1}{\Gamma_{\text{g}}} + \frac{1}{\alpha} + \frac{1}{\Gamma_{\text{b}}} \tag{4}$$

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 $1/\gamma_{\text{eff}}$ is composed of three resistor terms due to gas diffusion ($1/\Gamma_{\text{g}}$), interfacial mass transfer ($1/\alpha$) and bulk diffusion and reaction ($1/\Gamma_{\text{b}}$) (Pöschl et al., 2007). The conductance of gas diffusion is commonly calculated based on $\Gamma_{\text{g}} = 8D_{\text{g}}\omega^{-1}d_{\text{p}}^{-1}$, where D_{g} is the diffusion coefficient of

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

Gases	Mineral dust	Soot	Solid organic aerosol	Liquid organic aerosol	Sea salt aerosol
Steady state [b]					
O_3	4.4×10^{-9}–4.8×10^{-5}	1.0×10^{-7}–1.6×10^{-4}	2.0×10^{-6}–6.9×10^{-5}	1.1×10^{-5}–3.0×10^{-3}	1.3×10^{-6}–1.0×10^{-4}
NO_2	1.0×10^{-9}–2.3×10^{-7}	$< 5.0 \times 10^{-8}$	$< 5.0 \times 10^{-7}$	2.2×10^{-7}–7.0×10^{-6}	6.0×10^{-7}–3.0×10^{-4}
SO_2	1.5×10^{-8}–6.3×10^{-4}	4.0×10^{-9}–2.2×10^{-6}	n/a	n/a	3.2×10^{-3}–1.7×10^{-2}
N_2O_5	4.8×10^{-3}–2.0×10^{-1}	4.0×10^{-5}–6.3×10^{-3}	1.0×10^{-5}–3.1×10^{-3}	5.0×10^{-5}–4.5×10^{-2}	6.4×10^{-3}–3.9×10^{-2}
HNO_3	1.0×10^{-3}–2.1×10^{-1}	3.0×10^{-7}–1.5×10^{-3}	n/a	n/a	5.0×10^{-4}–2.5×10^{-1}
H_2O_2	1.0×10^{-5}–9.4×10^{-4}	n/a[c]	n/a	n/a	n/a
Initial state [b]					
O_3	2.0×10^{-7}–3.5×10^{-4}	1.4×10^{-4}–1.0×10^{-3}	1.0×10^{-5}	5.5×10^{-4}–1.6×10^{-3}	1.0×10^{-3}–3.6×10^{-2}
NO_2	2.5×10^{-9}–2.2×10^{-5}	1.0×10^{-6}–4.0×10^{-4}	1.0×10^{-7}–5.1×10^{-6}	2.0×10^{-5}	1.0×10^{-4}
SO_2	1.4×10^{-7}–7.7×10^{-4}	3.0×10^{-3}	n/a	9.2×10^{-7}–1.0×10^{-5}	6.9×10^{-3}–9.0×10^{-2}
N_2O_5	2.0×10^{-2}–3.0×10^{-1}	1.6×10^{-2}–4.4×10^{-2}	n/a	n/a	n/a
HNO_3	2.5×10^{-4}–1.8×10^{-1}	7.7×10^{-4}–2.0×10^{-2}	$\leq 6.6 \times 10^{-5}$	n/a	6.6×10^{-2}–7.5×10^{-1}
H_2O_2	5.0×10^{-5}–4.0×10^{-3}	n/a	n/a	n/a	n/a

[a] A detailed review with references for γ_{eff} values is given in Tables A1–A4. [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 milliseconds or seconds to the initial state, and those with a longer exposure time (~ 1 h or longer) to the steady state. [c] "n/a" denotes not available.

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., 2014a). 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 an aerosol surface area density of A (particle surface area per unit volume of air, μm^2 cm^{-3}), the total uptake flux of gas X by aerosols (F_{aer}, mol m^{-2} s^{-1}) is

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

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 the initial state and the steady state in Table 1 (details in Tables A1–A4). These coefficients are mainly derived from measured values in the literature, reviewed data from the 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/, last access: January 2019) and the NASA JPL (Jet Propulsion Laboratory, Burkholder et al., 2015) (see references in Tables A1–A4). As we focus on the PBL, γ_{eff} values measured at room temperatures (~ 298 K) are mainly presented. Gas uptakes at very low temperatures (e.g., in the polar region or stratosphere) are outside the scope of this study and should be explored in future work.

Although the initial and steady-state uptake coefficients are listed, it should be noted that the values at the 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 values recommended by the IUPAC group with relatively high reliability. As shown in Table A1, variances of more than 3 orders of magnitude 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.

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 for γ_{eqv} as follows:

$$\gamma_{eqv} = \frac{4}{3} \frac{V_d}{Ah} \times 10^2 \quad (6)$$
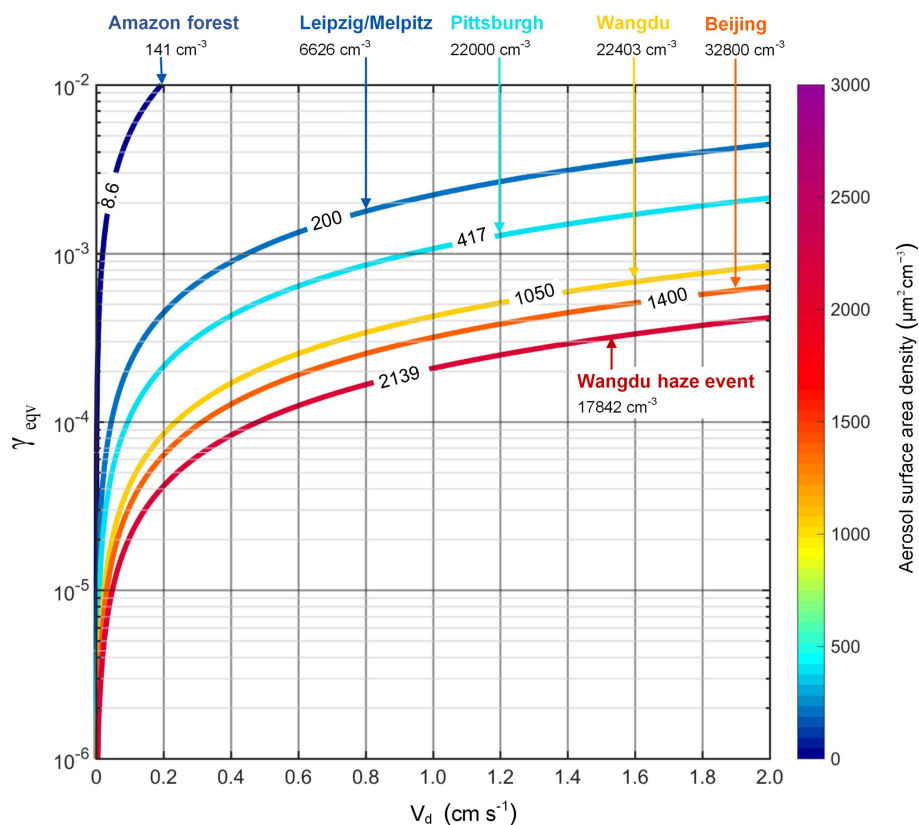
Figure 2. Relation between γ_{eqv} and V_d for a mixing height of 300 m and aerosol surface area densities (A) observed at different locations and under different conditions: Amazon forest (Pöschl et al., 2010), Leipzig/Melpitz (Ma et al., 2014; Ma and Birmili, 2015), Pittsburgh (Stanier et al., 2004), Wangdu with and without haze event (Wu et al., 2017) and Beijing (Wu et al., 2008). For each city/condition, the line is labeled with the corresponding aerosol surface area density. Aerosol particle number concentrations are also provided for orientation.

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

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

According to Eq. (6), γ_{eqv} is proportional to V_d, and is inversely proportional to the aerosol surface area density and the mixing height. We calculated a series of γ_{eqv} values 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 a 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} values 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 the 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 from the literature. A values are in the range of 200–2000 $\mu m^2\,cm^{-3}$ for aerosols in urban areas (Woo et al., 2001; Stanier et al., 2004; Wu et al., 2008, 2017; Ma and Birmili, 2015), 200–1000 $\mu m^2\,cm^{-3}$ in agricultural regions (suburban and rural, Held et al., 2008; Su et al., 2008b; Ma et al., 2014; Ma and Birmili, 2015; Wu et al., 2017), 8–700 $\mu m^2\,cm^{-3}$ in the Amazon forest (Zhou et al., 2002; Rissler et al., 2006; Pöschl et al., 2010; Andreae et al., 2015) and 20–200 $\mu m^2\,cm^{-3}$ 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 Figs. 3–5): we use 1050 $\mu m^2\,cm^{-3}$ for the urban environment (Wu et al., 2017), 230 $\mu m^2\,cm^{-3}$ for agricultural regions (Held et al., 2008), 46 $\mu m^2\,cm^{-3}$ for the Amazon forest (Rissler et al., 2006) and 76 $\mu m^2\,cm^{-3}$ for SSA (canonical distribution at a 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 velocities and aerosol surface area densities at a mixing height of 300m. V_d values for different scenarios were calculated based on the resistance scheme illustrated above, showing a range of 0.01–$2.3\,\mathrm{cm\,s^{-1}}$, with the lowest for NO_2 and the highest for N_2O_5 and HNO_3 (details in Table S2). Aerosol surface area densities covered a range of 8.6 to $2139\,\mu m^2\,cm^{-3}$, from pristine rainforest to polluted megacities, respectively. We show the calculated γ_{eqv} under typical conditions (typical A as described above, $h = 300\,m$) by season in Table S3 and a detailed illustrated γ_{eqv} for each gas species in the sections below. As shown in Fig. 2, γ_{eqv} decreases with increasing A, which is closely related to the air pollution level, and increases with increasing V_d.

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

With the increase of V_d ($> 0.1\,\mathrm{cm\,s^{-1}}$), γ_{eqv} increases to 10^{-5}–10^{-2} accordingly. In a pristine region of the 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 ($A = 2139\,\mu m^2\,cm^{-3}$). 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 region, NO_2 on agricultural region/Amazon forest, and N_2O_5 and HNO_3 over all land use types (see Table S2). Thus, we can derive a general conservative criterion of $\gamma_{eff} > 10^{-5}$ 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 and H_2O_2). The uptake coefficients at the initial state are generally 1–3 magnitudes higher than those at steady state (see Table 1 and Figs. 3–5). Considering the timescale of gas uptake by aerosols in the real world and applications in models, we mainly focus on the comparisons of γ_{eqv} and the steady-state γ_{eff} in the following discussions.
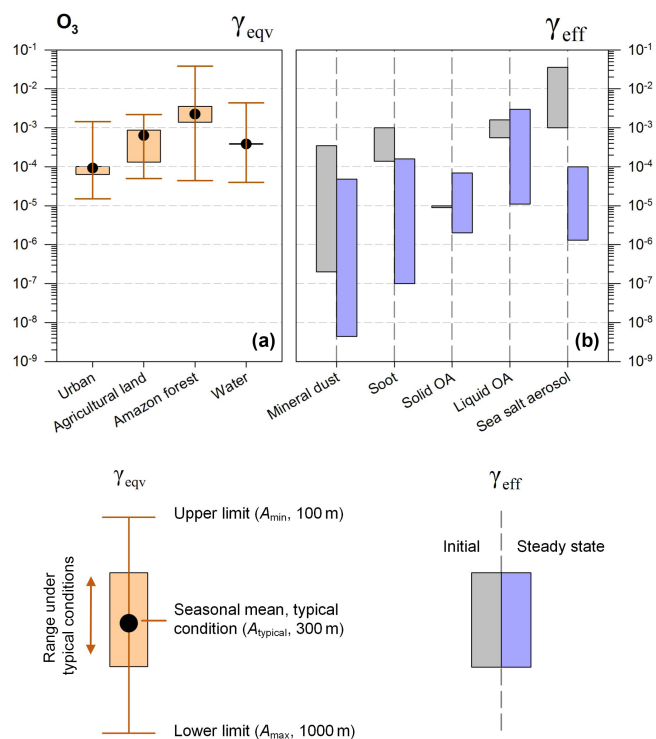

Figure 3. Equivalent uptake coefficients (γ_{eqv}, **a**) and laboratory measurement values (γ_{eff}, **b**) for O_3 on different ground types and aerosols. For γ_{eqv}, the upper whiskers represent maximum values calculated at the lowest A and h ($h = 100\,m$), the lower whiskers represent minimum values calculated at the 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 gray bar represents the range of initial values, and the blue bar represents the range of steady-state values observed in laboratory experiments.

3.1 O_3

Under typical conditions (typical A by land use, $h = 300\,m$, as illustrated above), γ_{eqv} values for O_3 between 9.2×10^{-5} and 2.2×10^{-3} are determined, with the lowest value in urban areas and the highest value in the Amazon forest. The extended range is 1.4×10^{-5}–3.8×10^{-2}, which varies with particle area densities and mixing heights (Fig. 3). There are overlaps between γ_{eqv} and γ_{eff} for liquid organic aerosols (OAs) among all investigated typical environments, and other types of aerosols under favorable circumstances for aerosol uptake in urban areas. γ_{eff} values lie below γ_{eqv} values 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 areas (e.g., $A = 1400\,\mu m^2\,m^{-3}$, 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 liquid organic aerosols and 1.3×10^{-6} to 1.0×10^{-4} for SSA, we can expect high
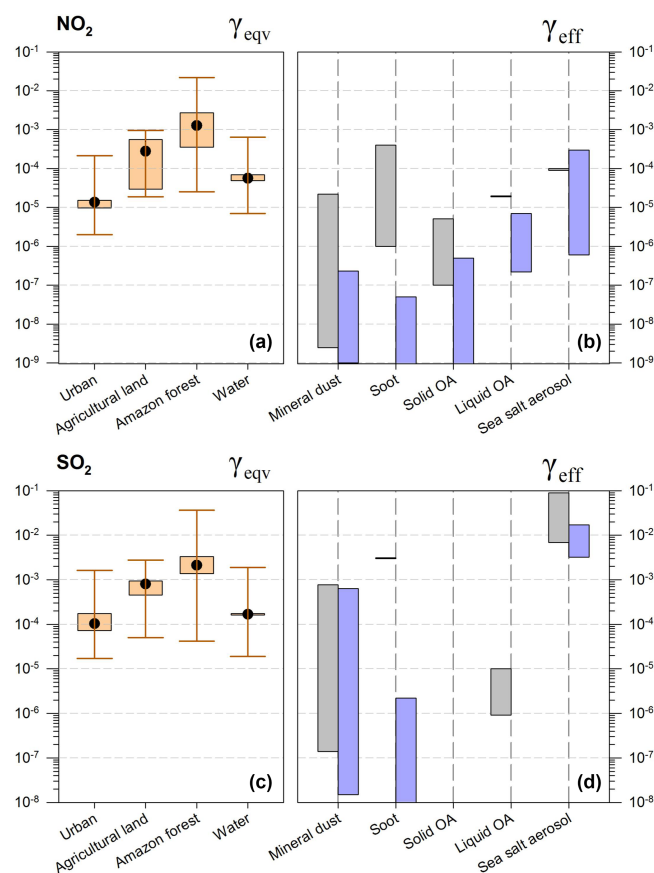
Figure 4. Uptake coefficients (γ_{eqv}, **a, c**; γ_{eff}, **b, d**) for NO_2 and SO_2 on different ground types and aerosols.

uptake fluxes of O_3 on these three kinds of aerosols when the corresponding γ_{eff} value is larger than 10^{-4} for ground surfaces other than urban.

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} values show large variances from 10^{-5} to 10^{-3} and corresponding γ_{eqv} values 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 demonstrate the importance of the phase state of aerosols in multiphase reactions and the transport of polycyclic aromatic hydrocarbons to improve the model performances at both regional and global scales.

Shiraiwa et al. (2017) show the global map of the SOA (secondary organic aerosol) phase state at the Earth's surface. SOA in southern China, the Amazon forest and South Africa are mainly in the liquid phase within the PBL. For these regions, the comparable uptake fluxes for O_3 on liquid organic aerosols compared with 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 to 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 and NO_x through chemical reactions in the gas phase.

3.2 NO_2

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

NO_2 tends to deposit on the ground surface instead of on mineral dust particles, soot and solid organic aerosols. As reviewed in Tables A1–A3, 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 values ranging from $46\,\mu m^2\,cm^{-3}$ (Amazon) to $1050\,\mu m^2\,cm^{-3}$ (Wangdu) and at a mixing height of $300\,m$, γ_{eqv} values of NO_2 lie between 1.4×10^{-5} and 1.3×10^{-3}, and are 1–3 orders of magnitude larger than γ_{eff} on these three kinds of aerosols. Increasing the PBL mixing height and aerosol surface area may reduce γ_{eqv} values by $\sim$ 1–2 orders of magnitude, but they are still above the measured γ_{eff} values 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 (6.0×10^{-7}–3.0×10^{-4}) are probably attributed to the reactions of Cl^- with dissolved NO_2 in the 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 the land surface or water bodies in coastal areas; therefore, it 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 2.2×10^{-7}–7.0×10^{-6}) in urban areas of high A. As shown in Fig. 4, the lower limit of γ_{eqv} in urban is $\sim 2.2 \times 10^{-6}$, which lies within the range of γ_{eff}. The uptake coefficients of NO_2 on pure water are estimated to be around 10^{-7}–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 5.4–5.8×10^{-4} for NO_2 uptake on ammonium sulfate aerosols at high relative humidity (RH; RH $= 50\%$, 85%). The presence of reactants such as inorganics of HSO_3^- or phenolic compounds in aqueous aerosols can promote the uptake significantly via chemical reactions with dissolved NO_2 to 10^{-5}–

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., 2008a; Cheng et al., 2016). Multiple measurements and modeling work have also pointed out that the 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 addressed in detail in models. In the Amazon forest, where A is too low ($46\,\mu m^2\,cm^{-3}$), corresponding to a γ_{eqv} value of the order of 10^{-3}, even a high γ_{eff} value 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 3 orders of magnitude with aerosol composition (10^{-7}–10^{-4}). On liquid organic aerosols and sea salt aerosols, the uptake can reach up to 10^{-6}–10^{-4} via chemical reactions (Abbatt and Waschewsky, 1998; Ammann et al., 2005; Yabushita et al., 2009), which is significantly larger than the uptake on pure water of 10^{-7}–10^{-6} (Lee and Tang, 1988; Kleffmann et al., 1998; Gutzwiller et al., 2002). For liquid ammonium sulfate aerosols, discrepancies with 2 orders of magnitude (10^{-6}–10^{-4}) in γ_{eff} values are found, although the reasons for this are currently unknown (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} values of SO_2 vary between 1.0×10^{-4} and 2.1×10^{-3} for land surfaces and 1.7×10^{-4} above water bodies under typical conditions. As shown in Fig. 4, the SO_2 uptake by mineral dust is comparable to the ground uptake in urban areas, and under favorable conditions over agricultural land and water bodies. For soot, aerosol uptake is magnitudes lower than those on the ground ($\gamma_{eqv} \geq \gamma_{eff}$); thus, this process is unimportant for SO_2. For SSA, γ_{eff} values of 3.2×10^{-3}–1.7×10^{-2} have been reported for SO_2 at an aerosol pH of 5.4–6.6, which is high enough to compete with dry deposition 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 1.6×10^{-5}–1.6×10^{-3}, 5.5×10^{-5}–2.8×10^{-3} and 1.9×10^{-5}–1.9×10^{-3} for urban areas, agricultural land and water bodies, respectively. The γ_{eff} of mineral dust falls in this range under high aerosol loadings or high mixing heights. The wide range of γ_{eff} values for mineral dust (1.5×10^{-8} to 6.3×10^{-4}) is a big challenge re-

garding its application in models, because it can be affected by the presence of oxidants, the phase state, the 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 under different conditions in the following.

Under dry conditions (as reviewed in Table A1), γ_{eff} values of the order of 10^{-7}–10^{-4} are measured (Goodman et al., 2001; Usher et al., 2002; Ullerstam et al., 2003; Adams et al., 2005; Li et al., 2006). IUPAC recommends 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 a high RH, water can enhance or inhibit the uptake by affecting reactive sites, and this effect varies with experimental conditions (Huang et al., 2015; Zhang et al., 2018). Conversely, the uptake rate can be improved by several factors and/or aqueous chemical reactions, such as the presence of O_3, H_2O_2 and transition metal ions (TMIs), which strongly depend on the aerosol pH (Jayne et al., 1990; Li et al., 2006; Cheng et al., 2016; Zhang et al., 2018). The initial γ_{eff} value of SO_2 on pure water can reach as high as 10^{-3}–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 a TMI- or H_2O_2-dominated regime to a NO_2- or O_3-dominated regime (Cheng et al., 2016). In this case, the SO_2 uptake on aqueous aerosols is expected to play a dominant role over dry deposition under specific circumstances, such as haze events (He et al., 2014; Cheng et al., 2016), which should be quantified by combining in situ measurements and atmospheric modeling.

As shown in the examples in Table S4, several model schemes adopt a γ_{eff} value of $\sim 10^{-4}$ (Liao and Seinfeld, 2005; K. Wang et al., 2012), around 1 order of magnitude higher than the measured values under 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). Under low RH conditions, the uptake coefficient commonly used in models is based on the dry deposition measurement of SO_2 on calcareous soils, cements and Fe_2O_3, rather than on laboratory measured γ_{eff} values 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 models under high RH conditions 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} value should be applied with the caveat that these prerequisites have been fulfilled, especially when extending it for other type of aerosols (Zheng et al., 2015).
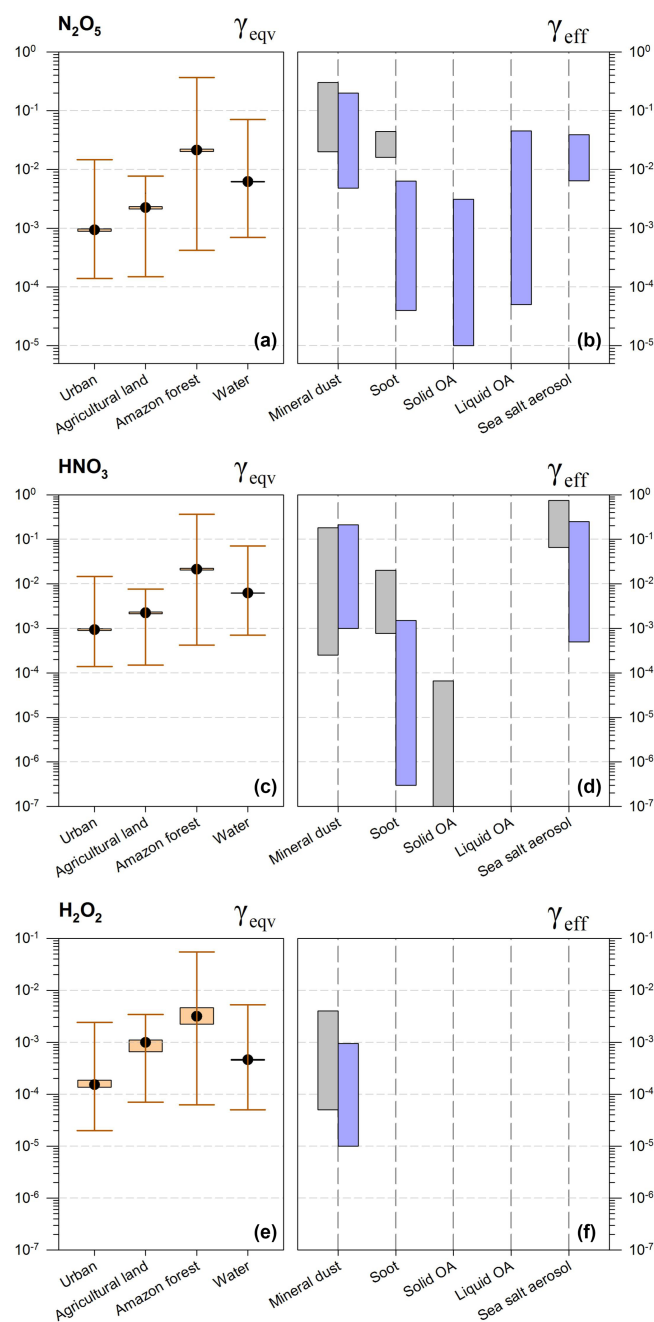
Figure 5. Uptake coefficients (γ_{eqv}, **a, c, e**; γ_{eff}, **b, d, f**) for N_2O_5, HNO_3 and H_2O_2 on different ground types and aerosols.

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 is 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; K. Wang 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, this uptake should also be characterized in atmospheric models in detail.

For N_2O_5, the measured uptake coefficients are 4.8×10^{-3}–0.20 for mineral dust, 4.0×10^{-5}–6.3×10^{-3} for soot and 6.4×10^{-3}–3.9×10^{-2} for SSA, which are comparable to or 1–2 orders of magnitude higher than the calculated γ_{eqv} values of 9.3×10^{-4}–2.1×10^{-2} under typical conditions (details in Tables A1–A4). 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} values of 10^{-3}–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, for which the relation was determined from laboratory experiments (Kane et al., 2001; Bauer et al., 2004; Liao and Seinfeld, 2005).

The extended ranges of γ_{eqv} for HNO_3 is 1.5×10^{-4}–1.5×10^{-2} (urban), 1.5×10^{-4}–7.7×10^{-3} (agricultural land), 4.2×10^{-4}–3.7×10^{-1} (Amazon forest) and 7.0×10^{-4}–7.0×10^{-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 the more important role of aerosol uptake than of the ground surfaces. The uptake of HNO_3 on soot and solid organic aerosols appears to be less important. The HNO_3 uptake on mineral dust has been implemented in current models with an uptake coefficient of 0.1, or between 1.1×10^{-3} and 0.2, which is consistent with the range of experimentally determined γ_{eff} values reviewed in this study (Liao and Seinfeld, 2005; K. Wang et al., 2012).

The study of the uptake of H_2O_2 by aerosols is rather limited compared with the other aforementioned trace gases. The reported γ_{eff} values on dust and ambient aerosol samples suggest that 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 1.0×10^{-5}–9.4×10^{-4} and overlap with the calculated γ_{eqv} of 1.5×10^{-4}–3.0×10^{-3} under typical conditions. Ambient aerosols collected in urban areas show similar γ_{eff} values of H_2O_2 (8.1×10^{-5}–4.6×10^{-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 has been 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; K. Wang et al., 2012). The uptake by other aerosol types has not been considered due to limited experimental data. Thus, more laboratory kinetic measurements are needed. As 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. The large variability found in the measured γ_{eff} for SO_2 and NO_2 is 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 the limitations 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, differences of more than 3 orders of magnitude are found for its uptake by mineral dust (10^{-8}–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 and NO_2), RH, measurement techniques and the 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 1 order of magnitude to 1.6–6.6×10^{-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 the other different investigations shown in Table A1, 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 for use in models under low RH conditions. For high RH, we suggest determining γ_{eff} using information on aerosol pH due to the high correlation between these factors, as illustrated in Sect. 3.3.

For NO_2 uptake on liquid aerosol droplets, differences of 3 orders of magnitude are found (10^{-7}–10^{-4}), which vary significantly with aerosol composition. On pure water, the uptake is measured at 10^{-7}–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}–10^{-4} via multiphase reactions (Abbatt and Waschewsky, 1998; Ammann et al., 2005; Yabushita et al., 2009). For ammonium sulfate aerosols, large discrepancies of 10^{-6}–10^{-4} for the initial γ_{eff} are found, although the reasons for this are currently unknown (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 and geometric vs. BET

Measurements of the effective uptake coefficients revealed the instantly fast uptake at the initial state and the gradual decline 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 orders of magnitude faster than that at the steady state for aerosols (see Tables 1 and A1–A4). The timescale for reaching surface saturation/equilibrium is dependent on the reaction system. For a gas–aqueous particle surface, the timescale to establish equilibrium for the investigated species is less than 1 s (Seinfeld and Pandis, 2006, 554–557). For dust particles, it can take hours for complete saturation (Judeikis et al., 1978; Goodman et al., 2001). Fine particles with diameters $< 10 \mu m$ have lifetimes of several days in the atmosphere (Prospero, 1999; Lee et al., 2009). 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 that the surface passivation is negligible compared with 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 emission sources. Using a uniform uptake coefficient in atmospheric models may not be enough to reflect the deactivation process of the multiphase gas uptake during aerosol aging, considering the large range of γ_{eff} values varying with time.

In addition, the γ_{eff} values are measured and reported based on the geometric surface and/or the BET surface. Differences of more than 3 orders of magnitude are derived depending on whether or not the pores within the microstructure of solid aerosol surface are considered (see Table A1). Using the same method to calculate the available surface area may reconcile these differences (Tang et al., 2017). In this study, γ_{eff} values with a revised BET surface are generally used as the lower limit, and those using the geometric surface are used as the upper limit. Whether or not the BET area is used as a correction in the calculation of γ_{eff} or not remains a discrepancy when applied in models (Hanisch and Crowley, 2001a, b; 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

We can conclude that the 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 magnitude higher than on solid/semi-solid surfaces. In regions with high RH conditions and sufficient sources of organic compounds (e.g., the Amazon forest and southern China), the gas uptake is anticipated to have a considerable effect on concentrations. The effect is yet to be evaluated in combination with further model simulations.

Measurement of uptake by ambient aerosols is crucial to reconcile laboratory experiments and modeling results, especially for gases that have undergone limited investigation (e.g., H_2O_2). Currently limited work has been undertaken to address the uptake of H_2O_2 by aerosol particles other than mineral dust (Liao and Seinfeld, 2005; Pradhan et al., 2010a, b; K. Wang et al., 2012; Zhou et al., 2016). Because ambient aerosol samples show a γ_{eff} value comparable to that of dust particles, we recommend similar γ_{eff} values of 1.0×10^{-5}–9.4×10^{-4} for H_2O_2 uptake by other types of aerosol, which will lead to 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 the available factors mentioned above should be a feasible way to implement the gas uptake processes in atmospheric models (Mu et al., 2018).

There are limitations to the comparisons conducted in this study. We use a unified thermal velocity ($300 \, \text{m s}^{-1}$) for all gases, which will introduce positive biases of $+4\%$ to $+30\%$ for O_3, NO_2, SO_2, HNO_3 and 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 and 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 in 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 temperatures (e.g., in the polar region and stratosphere) are outside the 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 online measurements of γ_{eff} will favor the model parameterization and improve our understanding of the multiphase processes within the 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, 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 the ground and aerosols for six reactive trace gases (O_3, NO_2, SO_2, N_2O_5, HNO_3 and H_2O_2), various environments, aerosol types and mixing heights. The purpose of the study was to identify the aerosol uptake process, which is equally or more important than dry deposition on ground surfaces but has not been adequately addressed in models.

For efficient comparison, we derived a criterion, γ_{eqv}, to identify the dominant surface with respect to gas uptake. For investigated gas species, γ_{eqv} values generally lie in the magnitude of 10^{-4}, and can be extended to lower values in polluted areas and/or at low dry deposition velocities: γ_{eqv} values 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}) values 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, which is favored by the high particle surface densities. On the contrary, the contribution of aerosol uptake is minor compared with dry deposition for gases in the Amazon forest.

The aerosol uptake of the following gases 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 have unfortunately not received enough attention. For other combinations of gas species and aerosols, the ground tends to be the dominant surface rather than aerosols with respect to taking up trace gases within the 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 conditions and the existence of organic aerosols in the liquid state such as southern China, the Amazon forest and South Africa, the multiphase uptakes of O_3 by aerosols are expected to contribute comparable sinking fluxes to dry deposition. Compared with the relatively low uptakes on (semi-) solid organic aerosols, we can conclude that the phase state is a crucial factor influencing the uptake rates.

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

Gases	Mineral dust	Soot	Solid organic aerosol	Liquid organic aerosol	Sea salt aerosol
O_3				√	
NO_2					√
SO_2	√				√
N_2O_5	√	√	√	√	√
HNO_3	√				√
H_2O_2	√				

b. Large uncertainties should be addressed for the comparison results of SO_2 and NO_2. There are more than 3 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 under high RH condition, the contributions of aerosols should be cautiously determined with full consideration of the aerosol component, aerosol pH and so on.

c. H_2O_2 uptake on a variety of aerosols needs to be measured and evaluated. It is 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 a similar magnitude.

Appendix A

Table A1. Aerosol uptake coefficients (γ_{eff}) for reactive gases on mineral dust observed in laboratory experiments ($T = 298 \pm 2$ K 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
	TiO$_2$/SiO$_2$			0.3–3	0.02–0.32	Nicolas et al. (2009)
	Al$_2$O$_3$, Fe$_2$O$_3$ and SiO$_2$		5–18			Michel et al. (2002)
	China loess		2.7			Michel et al. (2002)
	Saharan sand		6			Michel et al. (2002)
O$_3$	Al$_2$O$_3$ and others		0.27–20		0.6–2.2	Michel et al. (2003)
($\times 10^{-5}$)	Saharan dust	0.55–35		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		
	Al$_2$O$_3$				0.013–0.26	Börensen 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$_2$			9400, 1200		Gustafsson et al. (2006)
	CaCO$_3$	0.656	0.025–0.043			Li et al. (2010)
	Kaolinite and pyrophyllite				0.07–0.81, 2.3	Angelini et al. (2007)
NO$_2$	TiO$_2$/SiO$_2$, Saharan sand and others				0.01	Ndour et al. (2008)
($\times 10^{-7}$)	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		

Table A1. Continued.

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
	$CaCO_3$, O_3	77	0.014	8.1	0.0015	Li et al. (2006)
	Saharan dust and O_3			390	0.05	Ullerstam et al. (2002)
	Saharan dust		1.6			Ullerstam et al. (2003)
	Al_2O_3 and MgO		9.5, 26			Goodman et al. (2001)
	Al_2O_3 and others		7.0–51			Usher et al. (2002)
	Chinese loess		3			Usher et al. (2002)
	Saharan dust and O_3		6.6			Adams et al. (2005)
	Al_2O_3 Fe_2O_3 and MgO		40, 55, 100			Judeikis et al. (1978)
SO_2	$CaCO_3$ powder		0.1			Santschi and Rossi (2006)
$(\times 10^{-5})$	$CaCO_3$ and 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		
	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)
N_2O_5	$CaCO_3$	5.0				Wagner et al. (2008)
$(\times 10^{-2})$	$CaCO_3$			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)
	Quartz			0.86–0.45		Wagner et al. (2009)
	Saharan sand				2	Tang et al. (2012)
	Arizona dust			0.63		Tang et al. (2014b)
	Illite			9.1, 3.9		Tang et al. (2014b)
	Summary	2–30		0.48–20		

Table A1. Continued.

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
	Mineral dust			1.7–5.4		Umann et al. (2005)
	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)
HNO_3	Saharan dust	13.6				Hanisch and Crowley (2001b)
($\times 10^{-2}$)	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 and 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		
	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]
H_2O_2	TiO_2, dark		2.5–40			Romanias et al. (2012)
($\times 10^{-4}$)	TiO_2, 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] $T = 253$–313 K; [b] $T = 268$–320 K.

Table A2. Aerosol uptake coefficients (γ_{eff}) for reactive gases on soot observed in laboratory experiments ($T = 298 \pm 2$ K 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
	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)
O_3	Spark generated		22–330			Fendel et al. (1995)
($\times 10^{-5}$)	Spark generated		0.12		0.01	Kamm et al. (1999)
	Kerosene and 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		
	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)
NO_2	Hexane soot, BC				0.0015–0.0024	Prince et al. (2002)
($\times 10^{-5}$)	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		
	BC	3000				Rogaski et al. (1997)
SO_2	Fresh BC and aged BC				0.00398, 0.32	Xu et al. (2015)
($\times 10^{-6}$)	BC and O_3				2.17	Xu et al. (2015)
	Summary	3000		0.004–2.17		
	Decane soot	44		5		Karagulian and Rossi (2007)
N_2O_5	Spark generated				0.04–0.2	Saathoff et al. (2001)
($\times 10^{-3}$)	Hydrocarbon soot	16		6.3		Longfellow et al. (2000)
	Summary	16–44		0.04–6.3		
	Decane soot	200		4.6–5.2		Salgado-Muñoz and Rossi (2002)
	Spark generated				0.003	Saathoff et al. (2001)
HNO_3	Hydrocarbon soot			15	0.5	Longfellow et al. (2000)
($\times 10^{-4}$)	Spark generated		0.052–7.7		0.00098–0.019	Kirchner et al. (2000)
	Summary	7.7–200		0.003–15		

Table A3. Aerosol uptake coefficients (γ_{eff}) for reactive gases on organic aerosols observed in laboratory experiments ($T = 298 \pm 2$ K 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 (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		

Table A3. Continued.

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
	Solid benzophenone, catechol, anthracene and anthrarobin	0.07–1.26 (dark), 0.65–2.40 (light)				George et al. (2005)[f]
	Solid benzophenone, catechol, anthracene and 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 and a mixture of organics	52, 22				Knopf et al. (2011)
	Solid levoglucosan and 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]
NO$_2$ ($\times 10^{-6}$)	SOA, pinene / O$_3$			< 0.5		Bröske et al. (2003)
	SOA, limonene / O$_3$, catechol / O$_3$, 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 and 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		
	Liquid oleic acid	0.92–6.44				Shang et al. (2016)[e]
SO$_2$ ($\times 10^{-6}$)	Liquid SOA by limonene and O$_3$	10–50				Ye et al. (2018)
	Summary	0.92–10 for liquid		Not available		

Table A3. Continued.

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
	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)
N_2O_5	Humic acid		0.1, 0.3, 1.0			Badger et al. (2006)
$(\times 10^{-3})$	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	Solid pyrene	≤ 6.6				Gross et al. (2008)[g]
$(\times 10^{-5})$	Summary	≤ 6.6 for solid		Not available		

[a] $T = 272$ K; [b] $T = 219$–298 K; [c] $T = 293$ K; [d] $T = 272$–298 K; [e] $T = 283$–298 K; [f] $T = 278$–308 K; [g] $T = 293$–297 K.

Table A4. Aerosol uptake coefficients (γ_{eff}) for reactive gases on sea salt aerosols observed in laboratory experiments ($T = 298 \pm 2$ K 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$_3$ ($\times 10^{-3}$)	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$_2$ ($\times 10^{-4}$)	Deliquesced NaCl	< 1.0				Abbatt and Waschewsky (1998)
	Deliquesced NaCl			2.8–3.7		Harrison and Collins (1998)[b]
	Aqueous NaCl	1.0				Yabushita et al. (2009)
	Chinese sea salt				0.00551	Ye et al. (2010)
	Chinese sea salt				0.0126	Ye et al. (2010)
	NaCl				0.6	Vogt et al. (1994)
	Summary	1.0		0.006–3.0		
SO$_2$ ($\times 10^{-3}$)	Synthetic sea salt	6.0–90		3.2–17		Gebel et al. (2000)
	Summary	6.0–90		3.2–17		
N$_2$O$_5$ ($\times 10^{-2}$)	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		

Table A4. Continued.

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
	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, 100 nm size	0.49				Tolocka et al. (2004)
	Deliquesced NaCl	15				Saul et al. (2006)
	Deliquesced NaCl	21–11				Liu et al. (2007)
HNO$_3$	NaCl / MgCl2	25–12				Liu et al. (2007)
($\times 10^{-2}$)	Sea salt	27–12				Liu et al. (2007)
	Deliquesced NaCl	20				Abbatt and Waschewsky (1998)
	Deliquesced 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] $T = 235$–299 K; [b] $T = 279$ K; [c] $T = 262$–278 K.

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