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

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

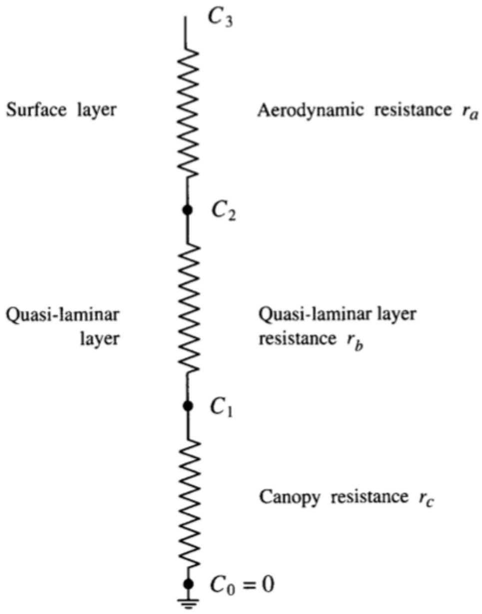

Figure S1. Resistance model for dry deposition, derived from the Figure 19.1 in Seinfeld and Pandis (2006).

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

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

1.1 calculation of R_a

Under neutral atmospheric condition:

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

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

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

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

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

1.2 calculation of *R*$_b$

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

where Sc is the Schmidt number (unitless), v means the kinematic viscosity of air, D is the molecular diffusivity for gases. We use D=10^{-5} m^2 s^{-1}, and a temperature-dependent v in the calculation.

1.3 calculation of *R*$_c$

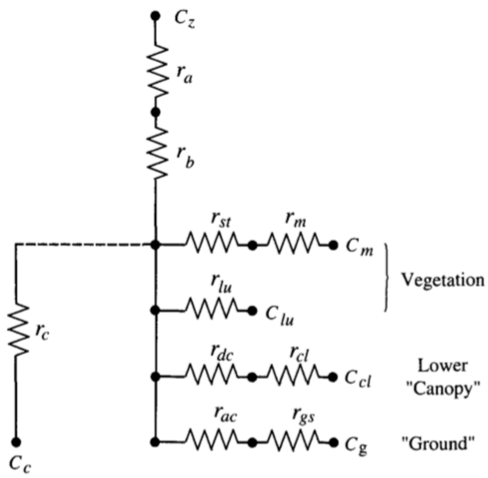

Figure S2. Schematic model for canopy resistance, derived from Wesely (1989).

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

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

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

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

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

Gases	Winter	Spring	Summer	Autumn with unharvested cropland	Late autumn after frost
			Urban		
O_3	0.15	0.24	0.24	0.24	0.24
NO_2	0.02	0.04	0.04	0.04	0.04
SO_2	0.41	0.17	0.20	0.20	0.20
N_2O_5	2.10	2.26	2.31	2.14	2.14
HNO_3	2.10	2.26	2.31	2.14	2.14
H_2O_2	0.44	0.32	0.34	0.33	0.33
			Agricultural land		
O_3	0.07	0.45	0.44	0.29	0.41
NO_2	0.02	0.20	0.29	0.07	0.07
SO_2	0.49	0.43	0.42	0.24	0.40
N_2O_5	1.10	1.18	1.21	1.12	1.12
HNO_3	1.10	1.18	1.21	1.12	1.12
H_2O_2	0.51	0.57	0.51	0.34	0.57
			Amazon forest		
O_3	0.15	0.27	0.37	0.14	0.14
NO_2	0.04	0.17	0.28	0.04	0.04
SO_2	0.14	0.25	0.34	0.14	0.14
N_2O_5	2.10	2.26	2.31	2.14	2.14

HNO_3	2.10	2.26	2.31	2.14	2.14
H_2O_2	0.23	0.37	0.48	0.23	0.23
			Water		
O_3	0.07	0.07	0.07	0.06	0.06
NO_2	0.01	0.01	0.01	0.01	0.01
SO_2	0.03	0.03	0.03	0.03	0.03
N_2O_5	1.05	1.06	1.07	1.05	1.05
HNO_3	1.05	1.06	1.07	1.05	1.05
H_2O_2	0.08	0.08	0.08	0.08	0.08

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

Gases	Winter	Spring	Summer	Autumn with unharvested cropland	Late autumn after frost
Urban					
O_3	0.64	1.01	1.01	1.00	1.00
NO_2	0.10	0.15	0.15	0.15	0.15
SO_2	1.73	0.72	0.84	0.83	0.83
N_2O_5	8.91	9.59	9.78	9.07	9.07
HNO_3	8.91	9.59	9.78	9.07	9.07
H_2O_2	1.87	1.37	1.44	1.41	1.41
Agricultural land					
O_3	1.31	8.76	8.55	5.63	7.93
NO_2	0.30	3.88	5.57	1.38	1.36
SO_2	9.45	8.39	8.12	4.55	7.69
N_2O_5	21.21	22.88	23.37	21.65	21.65
HNO_3	21.21	22.88	23.37	21.65	21.65
H_2O_2	9.80	10.97	9.94	6.61	11.06
Amazon forest					
O_3	14.01	25.72	35.56	13.78	13.78
NO_2	3.88	16.54	27.18	3.56	3.56
SO_2	13.96	24.22	33.03	13.76	13.76
N_2O_5	203.33	218.79	223.13	207.08	207.08
HNO_3	203.33	218.79	223.13	207.08	207.08
H_2O_2	22.43	35.42	45.95	22.25	22.25
Water					
O_3	3.92	3.81	3.82	3.76	3.76
NO_2	0.70	0.53	0.53	0.49	0.49
SO_2	1.76	1.66	1.67	1.61	1.61
N_2O_5	61.37	62.24	62.36	61.33	61.33
HNO_3	61.37	62.24	62.36	61.33	61.33
H_2O_2	4.68	4.61	4.63	4.51	4.51

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

Gases	Aerosol type	γ_{eff} (Liao and Seinfeld, 2005)	References	γ_{eff} (Zhu et al., 2010)	References	γ_{eff} (Wang et al., 2012)	References
O_3	Mineral dust	1.0×10^{-5}	Michel et al., 2002, 2003	2.7×10^{-5}	IUPAC[e]	5.0×10^{-5} ~ 1.0×10^{-4}	Dentener et al., 1996[b]; Zhang and Carmichael, 1999[b]
NO_2	Mineral dust			2.1×10^{-6}	IUPAC	4.4×10^{-5} ~ 2.0×10^{-4}	Underwood et al., 2001
	Wet aerosol	1.0×10^{-4}	Jacob, 2000				
SO_2	Mineral dust	3.0×10^{-4}(RH<50%), 0.1(RH≥50%)	Dentener et al., 1996	3.0×10^{-5}	IUPAC	1.0×10^{-4} ~ 2.6×10^{-4}	Zhang and Carmichael, 1999[b]
	Sea salt aerosol	5.0×10^{-3}(RH<50%), 5.0×10^{-2} (RH≥50%)	Song and Carmichael, 2001[b]				
N_2O_5	Mineral dust	See footnote[c]	Bauer et al., 2004[b]	3.0×10^{-2}	Seisel et al., 2005; Wagner et al., 2008; Karagulian et al., 2006	1.0×10^{-3} ~ 0.1	Dentener et al., 1996; DeMore et al., 1997
	Organic carbon	$5.2\times10^{-4}\times$ RH(RH<50%), 0.03(RH≥50%)	Thornton et al., 2003				
	Sea salt aerosol	5×10^{-3} (RH<50%), 0.03 (RH≥50%)	Atkinson et al., 2004				

	Sulfate/nitrate/ammonium	See footnote [d]	Kane et al., 2001, Hallquist et al., 2003				
HNO_3	Mineral dust	0.1	Hanisch and Crowley, 2001	0.17	IUPAC	1.1×10^{-3} ~ 0.2	Dentener et al., 1996; DeMore et al., 1997; Underwood et al., 2001
H_2O_2	Mineral dust			2.0×10^{-3}	De Reus et al., 2005	1.0×10^{-4} ~ 2.0×10^{-3}	Dentener et al., 1996

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

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

[c] $\gamma = 4.25\times10^{-4}\times RH - 9.75\times10^{-3}$

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

$\beta(T) = -4\times10^{-2}\times(T - 294), T \geq 282K$

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

$C_1=2.79\times10^{-4}$; $C_2=1.30\times10^{-4}$; $C_3=-3.43\times10^{-6}$; $C_4=7.52\times10^{-8}$

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

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