Supplement of

Timescales of Secondary Organic Aerosols to Reach Equilibrium at Various Temperatures and Relative Humidities

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Contents of this file

Tables S1 to S2

Figures S1 to S5

Table S1. Temperature-dependent kinetic parameters used in the simulations.

Parameter (Unit)	Description	Equation [a]
ω (cm s^{-1})	mean thermal velocity	$\omega(T) = (8RT/(\pi M))^{1/2}$
D_g (cm^2 s^{-1})	gas-phase diffusion coefficient	$D_g(T, P) = (T/T_{standard})^{1.75} (P_{standard}/P) D_g(T_{standard}, P_{standard})$ [b]
D_b (cm^2 s^{-1})	bulk diffusion coefficient	$D_b(T, RH) = kT/(6\pi a\eta(T, RH))$
k_a (cm s^{-1})	first-order adsorption rate coefficient	$k_a(T) = \alpha_s\omega(T)/4$
k_d (s^{-1})	first-order desorption rate coefficient	$k_d(T) = Ae^{-\left(E_{des}/(RT)\right)}$
$k_{ss,s}$ (cm s^{-1})	first-order rate coefficient for quasi-static-to-sorption layer transport	$k_{ss,s}(T, RH) = 2D_b(T, RH)/(\delta_{ss} + \delta_Z)$
$k_{s,ss}$ (s^{-1})	first-order rate coefficient for sorption-to-quasi-static surface transport	$k_{s,ss}(T, RH) = k_{ss,s}(T, RH)k_d(T)[Z]_{ss,eq}/(k_a(T)[Z]_{g,eq})$
$k_{b1,ss}$ (cm s^{-1})	rate coefficient of bulk layer 1-to-quasi-static surface transport	$k_{b1,ss}(T, RH) = 2D_b(T, RH)/(\delta_{ss} + \delta(1))$
$k_{ss,b1}$ (cm s^{-1})	rate coefficient of surface-to-bulk layer 1 transport	$k_{ss,b1}(T, RH) = k_{b1,ss}(T, RH)$
$k_{b,b}$ (cm s^{-1})	rate coefficient of transport between bulk layers	$k_{b,b}(T, RH) = 2D_b(T, RH)/(\delta(k) + \delta(k + 1))$
τ_d (s)	desorption lifetime	$\tau_d = k_d^{-1}$

[a] Description and the values of the symbols shown in the equations are summarized in Table S2.

[b] $D_g(T_{standard}, P_{standard})$ is calculated by the EPA on-line tools: (https://www3.epa.gov/ceampubl/learn2model/part-two/onsite/estdiffusion-ext.html).

Table S2. Description and the values of the symbols in the equations of Table S1.

Variable (Unit)	Description	Value
R (J K^{-1} mol^{-1})	gas constant	8.314
T (K)	temperature	varied from 220 to 310
RH (%)	relative humidity	varied from 0 to 100
M (g mol^{-1})	molar mass of compound Z	200
T_0 (K)	room temperature	298
ρ (g cm^{-3})	density of organic particles	1.4
T_{standard} (K)	sea level standard temperature	288.15
P_{standard} (Pa)	sea level standard atmospheric pressure	101325
K (J K^{-1})	Boltzmann constant	1.38×10^{-23}
a (cm)	effective molecular radius	10^{-8}
δ_{Z} (cm)	effective molecular diameter	2×10^{-8}
$\alpha_{\mathrm{s,0}}$	surface accommodation coefficient on free-substrate	1
A (s^{-1})	pre-exponential factor	10^{12}
E_{des} (kJ mol^{-1})	desorption energy	40
$[\mathrm{Z}]_{\mathrm{g,eq}}$ (cm^{-3})	equilibrium (saturation) number concentrations of Z in the gas phase	variable
$[\mathrm{Z}]_{\mathrm{ss,eq}}$ (cm^{-2})	equilibrium (saturation) number concentrations of Z in the quasi-static surface layer	variable
δ_{ss} (cm)	thickness of the quasi-static surface layer	variable
$\delta(k)$ (cm)	thickness of the bulk layer k	variable

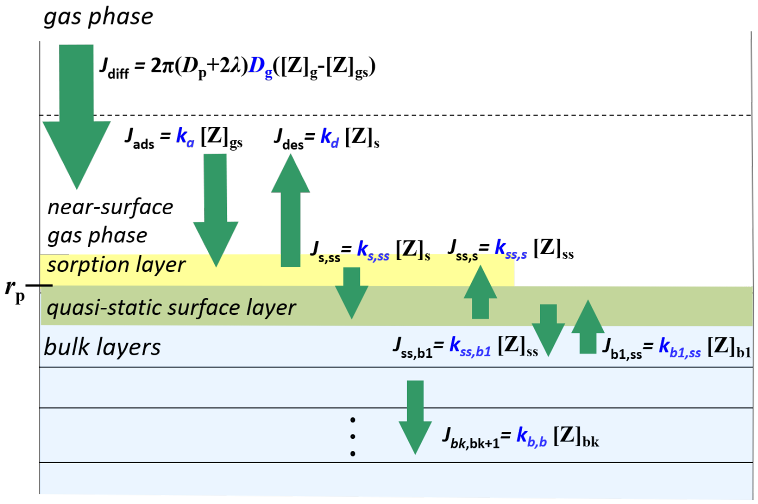

Figure S1. Temperature-dependent kinetic processes simulated in the KM-GAP model. Parameters in blue are treated as a function of temperature (Table S1). [Z] are concentrations of species Z in the gas (g) and near-surface gas phases (gs), at the sorption layer (s) and in the surface (ss) and in the bulk (b) layers. J are the transport fluxes between each layer, including the gas-phase diffusion flux (J_{diff}), the adsorption (J_{ads}) and desorption (J_{des}) fluxes, surface–bulk exchange fluxes ($J_{s,ss}$, $J_{ss,s}$, $J_{ss,b1}$, $J_{b1,ss}$), and bulk diffusion fluxes ($J_{b,b}$).

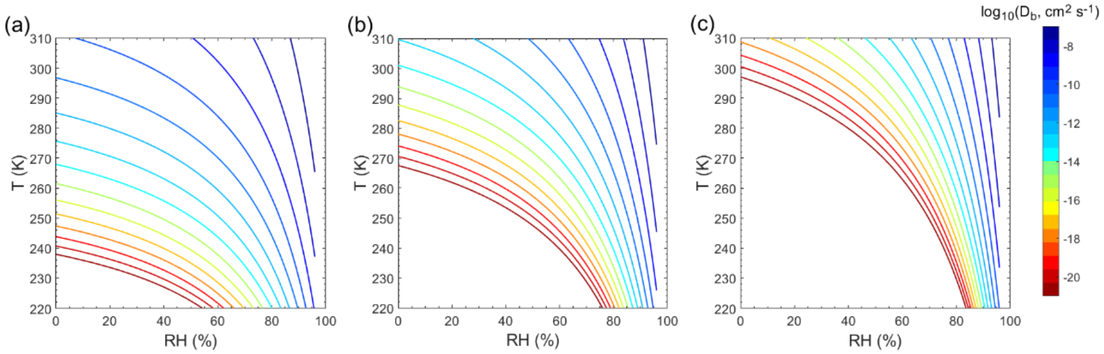

Figure S2. Bulk diffusion coefficient (D_b) in pre-existing particles as a function of temperature and relative humidity. The glass transition temperatures under dry conditions ($T_{g,org}$) are set to be (a) 240 K, (b) 270 K and (c) 300 K, respectively.

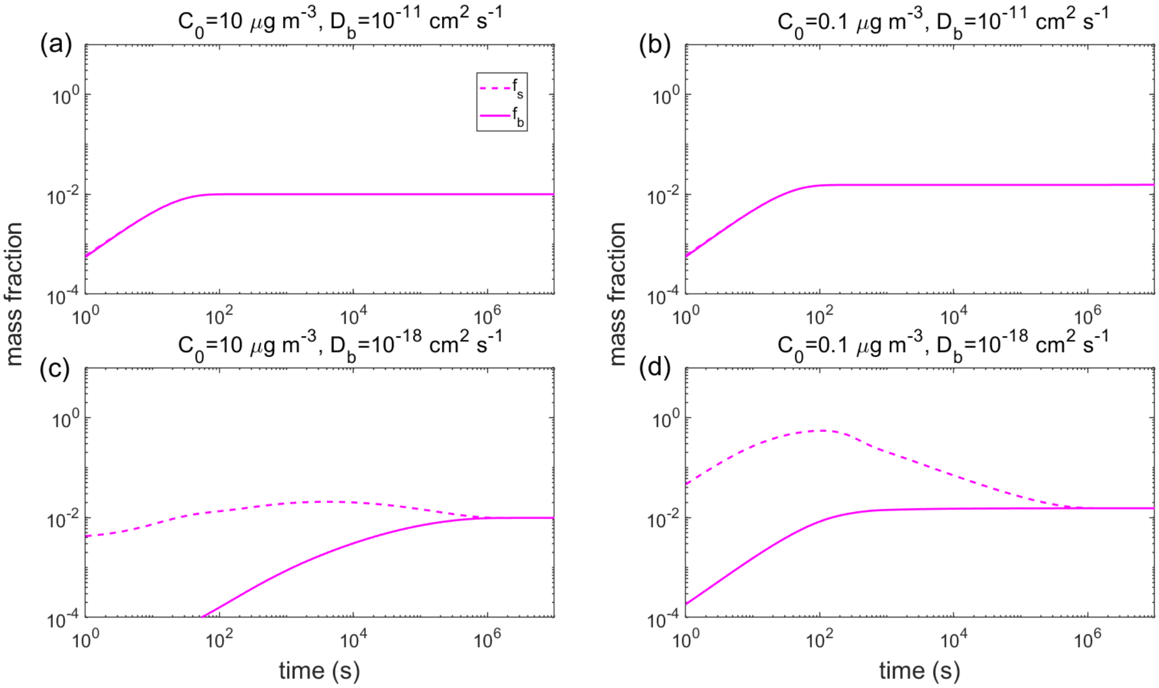

Figure S3. Temporal evolution of the mass fraction of Z in the near-surface bulk (f_s), and the average fraction of Z in the entire bulk (f_b). RH = 60% and T is (a, b) 298 K and (c, d) 250 K. The C_0 of Z is (a, c) 10 µg m^{-3} and (b, d) 0.1 µg m^{-3}. The glass transition temperature of pre-existing particles under dry conditions ($T_{g,org}$) is set to be 270 K, which leads to D_b of (a, b) 10^{-11} cm^2 s^{-1} and (c, d) 10^{-18} cm^2 s^{-1}. The initial mass concentration of pre-existing particles is assumed to be 20 µg m^{-3} with the number concentrations of 3 × 10^4 cm^{-3} and the initial particle diameter of 100 nm.

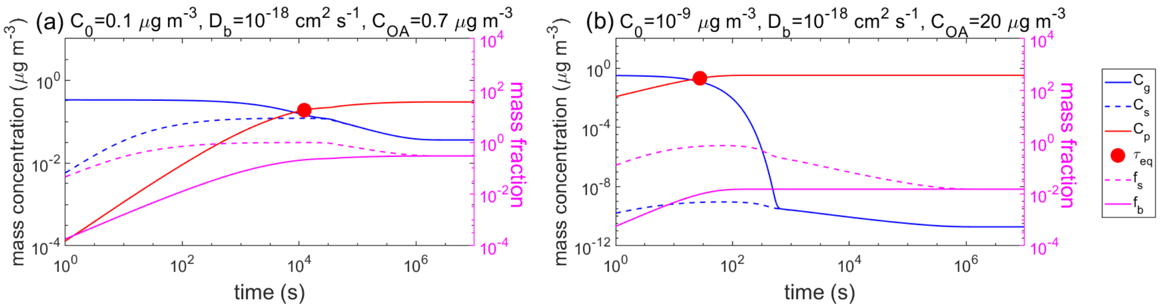

Figure S4. Temporal evolution of mass concentrations of the condensing compound Z in the gas phase (C_g), just above the particle surface (C_s), and in the particle phase (C_p). The mass fraction of Z in the near-surface bulk (f_s), and the average fraction of Z in the entire particle bulk (f_b) are also shown. D_b is 10^{-18} cm^2 s^{-1}. The C_0 of Z is (a) 0.1 µg m^{-3} and (b) 10^{-9} µg m^{-3}. The initial mass concentration of pre-existing particles is set to be (a) 0.7 µg m^{-3} and (b) 20 µg m^{-3}. τ_{eq} is marked with the red circle. τ_{eq} (~28 s) in (b) is consistent with the inverse of the condensation sink (29 s).

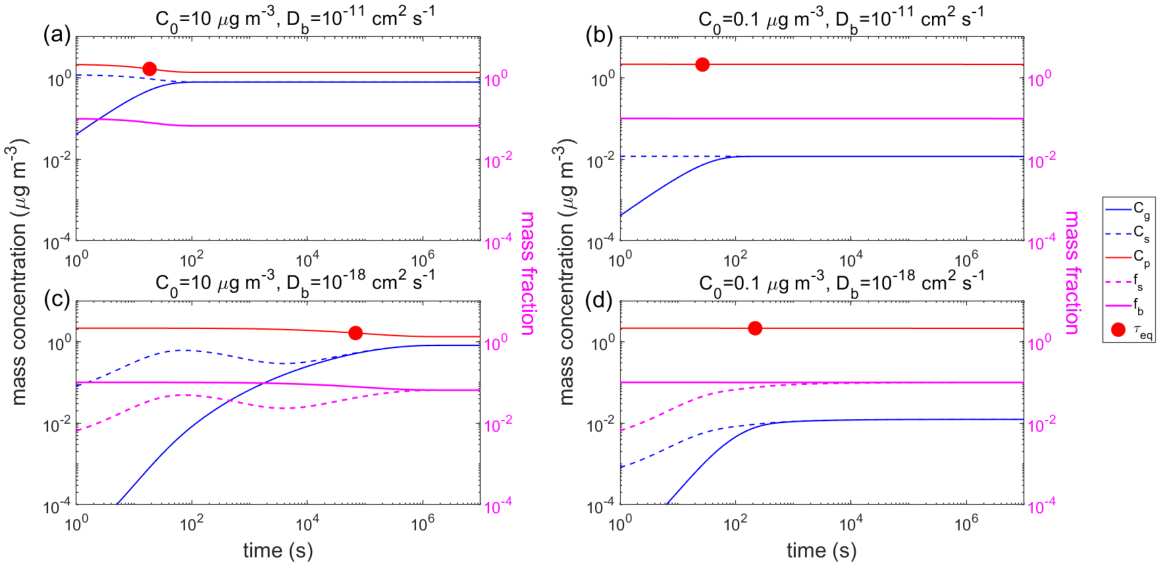

Figure S5. Temporal evolution of mass concentrations of the condensing compound Z in the gas phase (C_g), just above particle surface (C_s), in the particle phase (C_p), the mass fraction of Z in the near-surface bulk (f_s), and the average fraction of Z in the entire particle bulk (f_b). τ_eq are marked with red circles. RH = 60% and T is (a–b) 298 K and (c–d) 250 K. The C_0 of the condensing compound is (a, c) 10 µg m^{-3} and (b, d) 0.1 µg m^{-3}. The glass transition temperature of pre-existing particles under dry conditions ($T_{\mathrm{g,org}}$) is set to be 270 K, which leads to D_b of (a, b) 10^{-11} cm^2 s^{-1} and (c, d) 10^{-18} cm^2 s^{-1}. The initial mass concentration of pre-existing non-volatile particles (C_OA) is assumed to be 20 µg m^{-3} with the number concentrations of 3×10^4 cm^{-3} and the initial particle diameter of 100 nm.