Supplemental Material

Table S1. Kinetic input parameter sets of achetypal cases to showcase idealized limiting behavior. Values of "1" are used when a process is very fast and "0" may indicate that a process is either very slow or unimportant.

Parameter \ Limiting Case	S_{rx}	S_{bd}	S_{α}	S_{gd}	B_{rx}	B_{bd}	B_{α}	B_{gd}	B^{rd}_{trad},[a]
$k_{BR}\left(\frac{cm^3}{mol \cdot s}\right)$	0	0	0	0	5.00×10^{-17}	1	1	1	5.00×10^{-16}
$k_{SLR}\left(\frac{cm^3}{mol \cdot s}\right)$	7.00×10^{-15}	1	1	1	0	0	0	0	0
$D_{b,X}\left(\frac{cm^2}{s}\right)$	0	0	0	0	1	2.00×10^{-07}	1	1	2.00×10^{-08}
$D_{b,Y}\left(\frac{cm^2}{s}\right)$	1	5.00×10^{-13}	1	1	1	3.00×10^{-14}	1	1	1
$H_X\left(\frac{mol}{cm^3 \cdot atm}\right)$	0	0	0	0	4.80×10^{-04}	4.80×10^{-04}	4.80×10^{-04}	4.80×10^{-04}	4.80×10^{-04}
$\tau_{d,X}$ (s)	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}	1.00×10^{-02}
$\alpha_{s,0,X}$ (-)	1	1	1.00×10^{-04}	1	1	1	1	1	1
$D_{g,X}\left(\frac{cm^2}{s}\right)$	1.40×10^{-01}	1.40×10^{-01}	1.40×10^{-01}	8.00×10^{-06}	1.40×10^{-01}	1.40×10^{-01}	1.40×10^{-01}	8.00×10^{-06}	1.40×10^{-01}

[a]: Note that B^{rd}_{trad} is not a limiting case, but a distinct scenario in the reaction-diffusion regime.

Table S2. Environmental input parameters based on the oleic acid - ozone reaction system for calculation of archetypal limiting cases in Tab. S1.

Parameter	Value	Unit
r_{p}	1×10^{-5}	cm
L	100	(-)
T	298	K
$[\mathrm{X}]_{\mathrm{gs}}$	1×10^{14}	cm^{-3}
$[\mathrm{Y}]_{\mathrm{b}}(t=0)$	1.9×10^{21}	cm^{-3}
ρ_{X}	1.35	$\frac{\mathrm{g}}{\mathrm{cm}^3}$
ρ_{Y}	0.895	$\frac{\mathrm{g}}{\mathrm{cm}^3}$
M_{X}	48	$\frac{\mathrm{g}}{\mathrm{mol}}$
M_{Y}	282.46	$\frac{\mathrm{g}}{\mathrm{mol}}$
δ_{X}	3.89×10^{-8}	cm
δ_{Y}	8.06×10^{-8}	cm
σ_{X}	1.52×10^{-15}	cm^2
σ_{Y}	6.50×10^{-15}	cm^2

Table S3. Normalized sensitivity parameters for the archetypal limiting cases when the reaction has proceeded by 50 %. All values are unitless. Within a limiting case (column), the relative influence of each kinetic parameter is found by comparing the normalized sensitivity coefficients.

Norm. Sens. (S_i^n) \ Limiting Case	S_{rx}	S_{bd}	S_{α}	S_{gd}	B_{rx}	B_{bd}	B_{α}	B_{gd}	B_{trad}^{rd}, [a]
k_{BR}	0.00	0.00	0.00	0.00	1.00	0.00	0.00	0.00	0.48
k_{SLR}	1.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
$D_{b,X}$	0.00	0.00	0.00	0.00	0.00	0.80	0.00	0.00	0.52
$D_{b,Y}$	0.00	1.00	0.00	0.00	0.00	0.17	0.00	0.00	0.00
H_X	0.00	0.00	0.00	0.00	1.00	0.83	0.00	0.00	1.00
$\tau_{d,X}$	0.07	0.00	0.00	0.00	0.00	-0.01	0.01	0.00	0.00
$\alpha_{s,0,X}$	0.07	0.00	0.99	0.00	0.00	0.00	0.98	0.00	0.00
$D_{g,X}$	0.00	0.00	0.00	0.96	0.00	0.00	0.00	0.98	0.00

[a]: Note that B_{trad}^{rd} is not a limiting case, but a distinct scenario in the reaction-diffusion regime.

Table S4. Bulk limiting cases by SSR and BSR. Note that BSR≈1 is not possible while SSR≈0 when X is an externally applied reactive gas. Generally bulk reactions are classified only by their BSR, but SSR can be used to separate the two B_{α} subcases.

SSR	BSR	Possible regimes / cases
≈1	≈1	B^{rd}, B^{rd}_{trad}, B_{rx}, B_{bd}
≈1	≈0	$B_{\alpha,s\rightarrow b}$
≈0	≈0	B^{mt}, $B_{\alpha,g\rightarrow s}$, B_{gd}

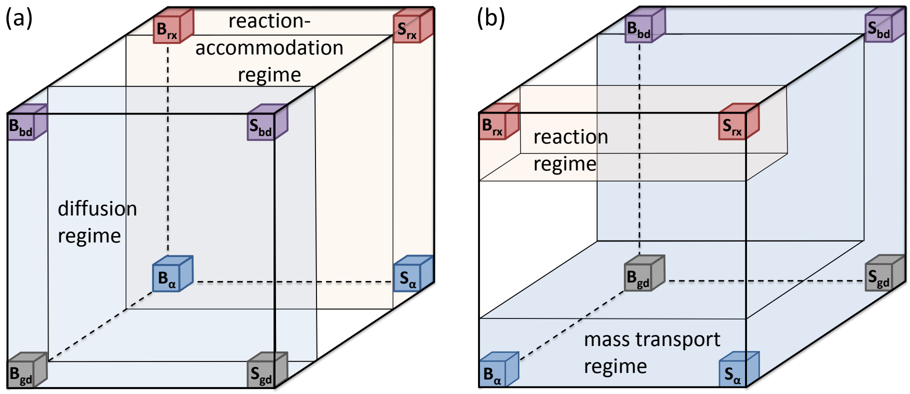
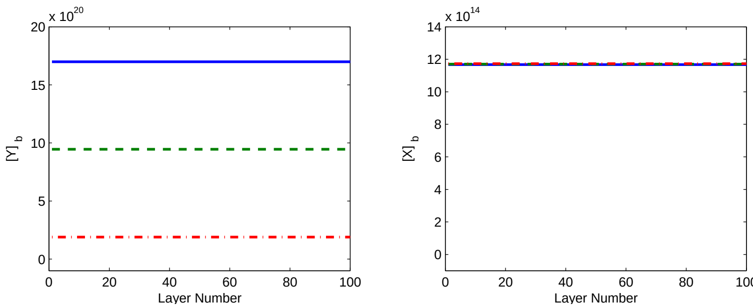
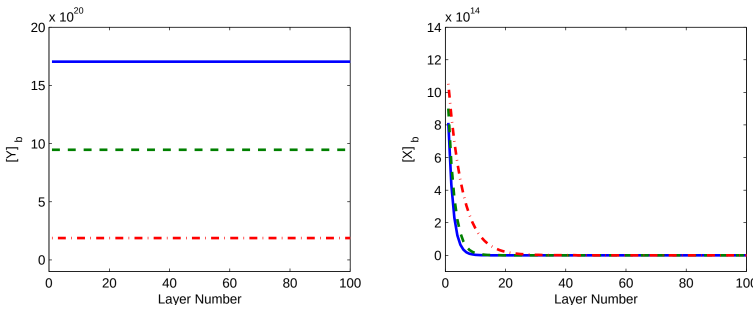


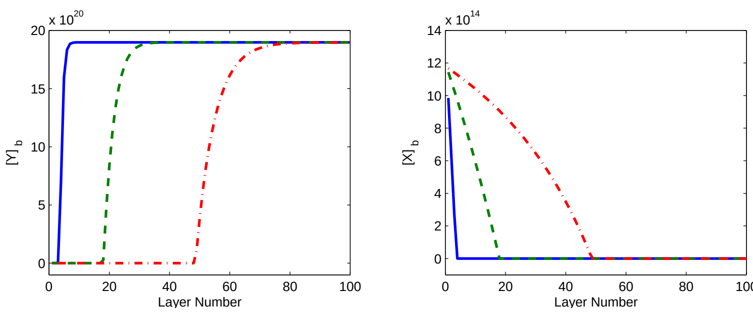
Fig. S1. Visualization of regimes as volumes of the cube in Fig. 2. In panel **(a)**, bulk and gas-phase diffusion limited cases are connected to form a diffusion regime, leaving a reaction-accommodation regime from the other limiting cases. In panel **(b)**, regimes are separated into reaction limitation and mass-transport limitation, illustrating the many possible physical (as opposed to chemical) limitations on aerosol reactive uptake. Note that the shading colors of the regimes in this figure do not correspond to the color-coding in Figs. 2 and 4.



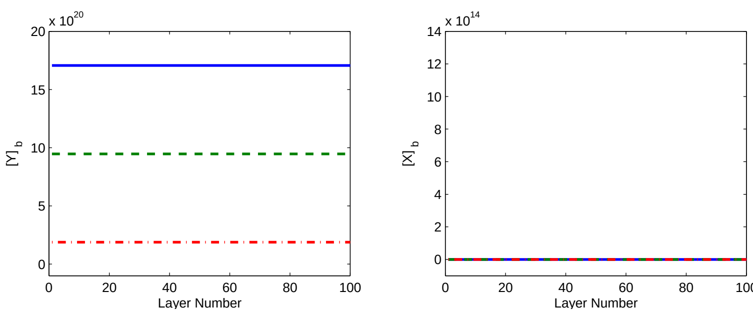
(a) B_{rx}



(b) B_{trad}^{rd}



(c) B_{bd}



(d) B_{α}, B_{gd}

Fig. S2. Concentration profiles for limiting cases with primarily bulk reaction. Colored lines show the evolution of the system as reaction progresses: 10 % reaction, blue; 50 % reaction, green; 90 % reaction, red. Not shown: the S_{bd} limiting case has a diffusional gradient in Y, which becomes depleted toward the surface.