



## *Supplement of*

## The impact of aerosol on cloud water: a heuristic perspective

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Introduction. Please refer to the main text for any definitions and references that are not stated here.

<span id="page-1-0"></span>Text S1. As argued in the main text, the charge/discharge to the thermodynamic carrying capacity can be represented as

<span id="page-1-1"></span>
$$
\frac{f(L_{\infty,h}) - f(L)}{\tau_{t}},
$$

where *L* is the liquid water path,  $L_{\infty,h}$  the thermodynamic carrying capacity, and  $\tau_t$  the timescale of this process. However, the form of the function *f* is not known. In the main text,  $f(x) = x$  is used, to which we refer to as linear thermodynamic charge/discharge. In this supplement, a logarithmic version in the form of  $f(x) = \ln(x)$  is explored.

Using the same setup as for Fig. 2, Fig. S2 shows the sensitivity of the model with logarithmic thermodynamic charge/discharge to the model parameters (a)  $\tau_t$ , (b)  $c_1$ , (c)  $L_0$ , and (d)  $m_{\infty,h}$ . Without addressing the details of each panel, one sees clearly that the model assumes the prescribed slope  $m_{\infty,h}$  for high *N* as expected (thin red lines). For small *N*, however, the slope  $m_1 = 0.43$  (thin black line) is larger than  $m_{\infty}$  (thin blue lines). The reason for this is the faster logarithmic thermodynamic recharge of precipitation losses. While larger *L* at low *N* could be remedied with an increased precipitation constant  $c_1$  (panel b),  $m_1$  cannot be tuned to match  $m_{\infty,1}$  and hence the ensemble LES results of [Glassmeier et al.](#page-1-0) [\(2021\)](#page-1-0). In fact,  $m_1$  does not vary for  $m_{\infty,h}$  < 2.0 when logarithmic thermodynamic charge/discharge is used (panel d), indicating fundamental differences between the linear and logarithmic formulations. Based on this analysis, we find that linear thermodynamic charge/discharge agrees better with our reference [\(Glassmeier et al., 2021\)](#page-1-0), and is thus used in this study.

Text S2. As stated in (3) of the main text, the temporal change of *L* is given by

$$
\frac{\mathrm{d}L}{\mathrm{d}t} = \frac{L_{\infty, h} - L}{\tau_{\text{t}}} - c_1 \frac{L^{3/2}}{N}.
$$

From the steady state solution of (3), we determine the derivative with respect to *N* as

*.*

$$
0 = \frac{\mathrm{d}}{\mathrm{d}N} \left[ \frac{L_{\infty, h} - L_{\infty}}{\tau_{t}} - c_{1} \frac{L_{\infty}^{3/2}}{N} \right].
$$

Solving for  $dL_{\infty}/dN$  gives

$$
\frac{\mathrm{d}L_{\infty}}{\mathrm{d}N} = \left(\frac{1}{\tau_{\mathrm{t}}} \frac{\mathrm{d}L_{\infty,\mathrm{h}}}{\mathrm{d}N} + c_1 \frac{L_{\infty}^{3/2}}{N^2}\right) \left(\frac{1}{\tau_{\mathrm{t}}} + \frac{3}{2}c_1 \frac{L_{\infty}^{1/2}}{N}\right)^{-1}
$$

The slope *m* relates to d*L/*d*N* as

$$
m = \frac{\mathrm{d}\ln(L)}{\mathrm{d}\ln(N)} = \frac{N}{L}\frac{\mathrm{d}L}{\mathrm{d}N}.
$$

This yields for the steady state

$$
m_{\infty} = \left(\frac{1}{\tau_{\rm t}}\frac{N}{L_{\infty}}\frac{\mathrm{d}L_{\infty,h}}{\mathrm{d}N} + c_1 \frac{L_{\infty}^{1/2}}{N}\right) \left(\frac{1}{\tau_{\rm t}} + \frac{3}{2}c_1 \frac{L_{\infty}^{1/2}}{N}\right)^{-1}.
$$

Using definition (5), we determine that

$$
\frac{N}{L_{\infty}} \frac{dL_{\infty,h}}{dN} = \frac{N}{L_{\infty}} \frac{d}{dN} \left[ L_0 \left( \frac{N}{N_0} \right)^{m_{\infty,h}} \right]
$$

$$
= m_{\infty,h} \frac{L_{\infty,h}}{L_{\infty}}
$$

$$
= m_{\infty,h} \left[ 1 + \frac{2}{3} \frac{\tau_t}{\tau_p} \right],
$$

where we used (7) for the last equality. With the definition of the precipitation timescale (4),

$$
\tau_{\rm p} = \frac{2}{3} \frac{1}{c_1} \frac{N}{L^{1/2}},
$$

we find

$$
m_{\infty} = \left(\frac{1}{\tau_{\rm t}} m_{\infty, {\rm h}} + \frac{1}{\tau_{\rm p}} \frac{2}{3} (m_{\infty, {\rm h}} + 1)\right) \left(\frac{1}{\tau_{\rm t}} + \frac{1}{\tau_{\rm p}}\right)^{-1}.
$$

This expression can be rearranged to yield

$$
m_{\infty} = \frac{m_{\infty, h}}{1 + \frac{\tau_{\text{t}}}{\tau_{\text{p}}}} + \frac{\frac{2}{3}(m_{\infty, h} + 1)}{1 + \frac{\tau_{\text{p}}}{\tau_{\text{t}}}},
$$

which is  $(8)$  in the main text.



Figure S1. The temporal change of *L* as a function of *N* for thermodynamics [radiation (blue), surface fluxes (orange), and entrainment (green)] and precipitation (red) is shown for  $L = 60 \text{ g m}^{-2}$ . Note that this plot is based on the data shown in Fig. 3 of [Hoffmann et al.](#page-1-1) [\(2020\)](#page-1-1), but presented to fit the arguments of this study. Please refer to [Hoffmann et al.](#page-1-1) [\(2020\)](#page-1-1) for details on how this graph has been created.



**Figure S2.** For a system with logarithmic thermodynamic charge/discharge, we show *L* after 7 days as a function of *N* for variations in (a)  $\tau_t$ , (b)  $c_1$ , (c)  $L_0$ , and (d)  $m_{\infty, h}$  (colored dots). The default configuration is differentiated by gray dots. Plots are overlayed with  $m_{\infty, 1} = 0.24$ ,  $m_{\infty,h} = -0.64$ , and  $m_l = 0.43$  (thin blue, red, and black lines, respectively), and the 14*µ*m cloud top effective droplet radius (black dashed line).



**Figure S3.** Joint *L*-*N* histograms (opaque colors) and mean  $\ln(L)$  (thick black line) for perturbations in *L* and *N* for  $\tau_{\text{prt}} = 10 \text{h}$ ,  $\sigma_{\text{prt}} = 1.0$ with (a)  $m_{\text{prt}} = -1$ , (b) 0.0, and (c) 1.0. Plots are overlayed with  $m_{\infty,1} = 0.24$  and  $m_{\infty,h} = -0.64$  (blue and red lines), and the 14 $\mu$ m cloud top effective droplet radius (black dashed line). Note that the histograms are normalized such that the integral over each *N* column yields 1 (cf. Gryspeerdt et al., 2019). Panel (d) shows the fitted slopes  $m_1$  (blue lines) and  $m_h$  (red lines) for  $\sigma_{\text{prt}} = 0.5$  (thin lines), 1*.*0 (medium lines), 2.0 (thick lines), and  $m_{\text{prt}} = -1.0$  (dashed lines), 0.0 (continuous lines), 1.0 (dashdotted lines).