Reversed Twomey effect: fact or fiction?

Wojciech W. Grabowski[1] and Subin Jose[2]

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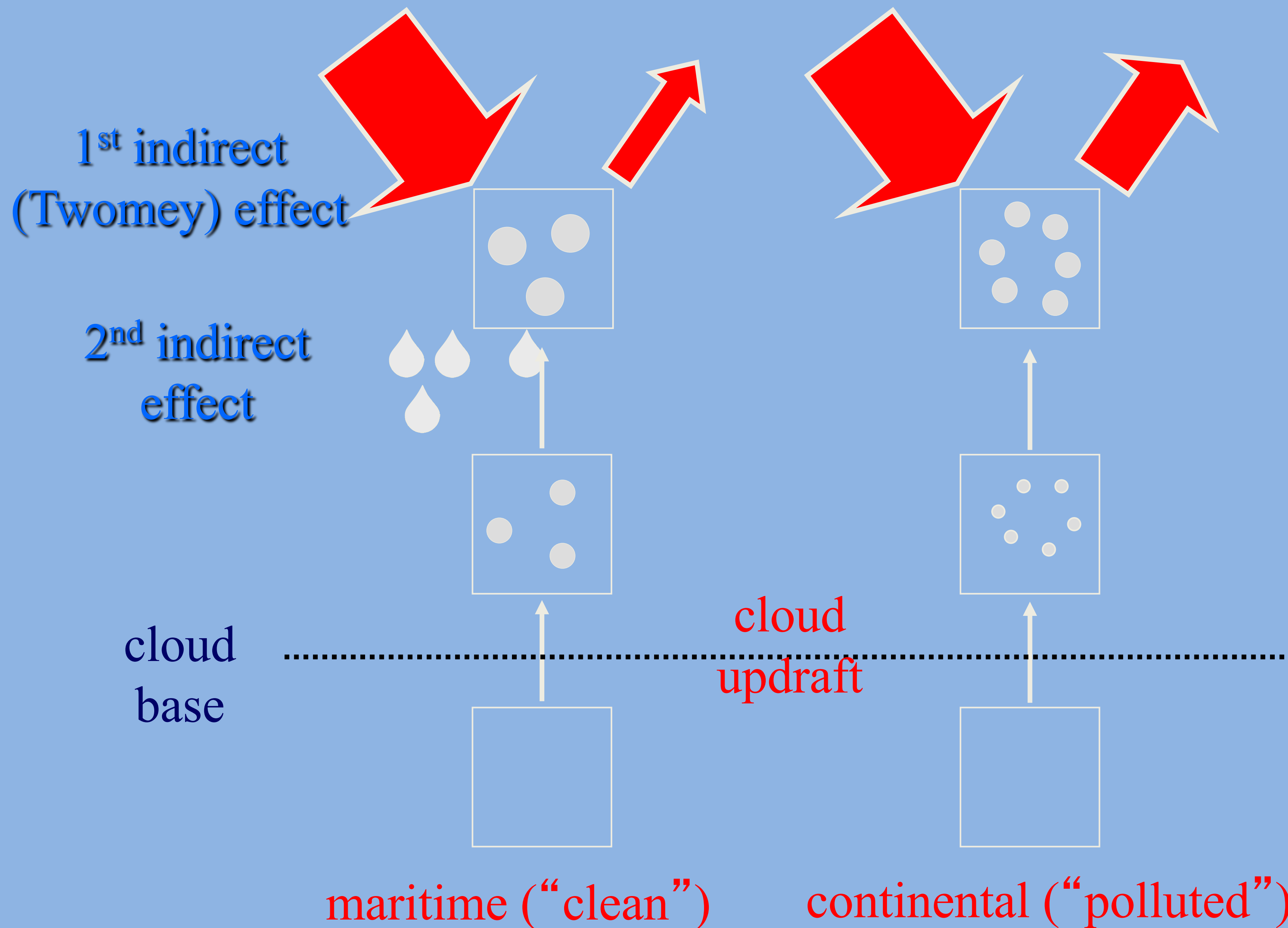
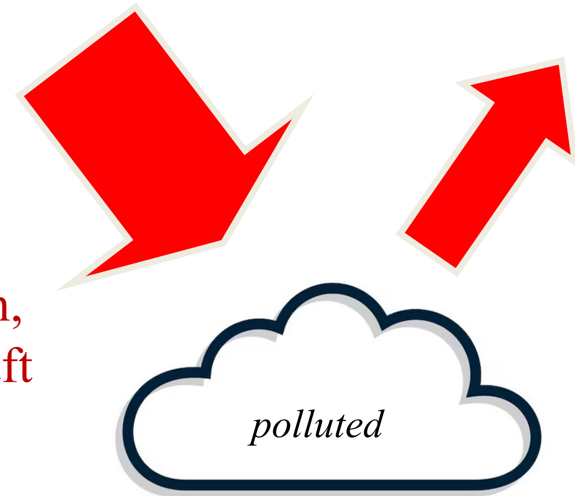
This material is based upon work supported by the National Center for Atmospheric Research, which is a major facility sponsored by the National Science Foundation under Cooperative Agreement No. 1852977.

Indirect aerosol effects (warm rain only)



the same liquid water path,
the same cloud base updraft



complications:
different liquid water path,
different cloud base updraft

Reversed Twomey effect:

www.nature.com/scientificreports

scientific reports

2020

Check for updates

OPEN

Anthropogenic emissions from South Asia reverses the aerosol indirect effect over the northern Indian Ocean

Subin Jose✉, Vijayakumar S. Nair & S. Suresh Babu

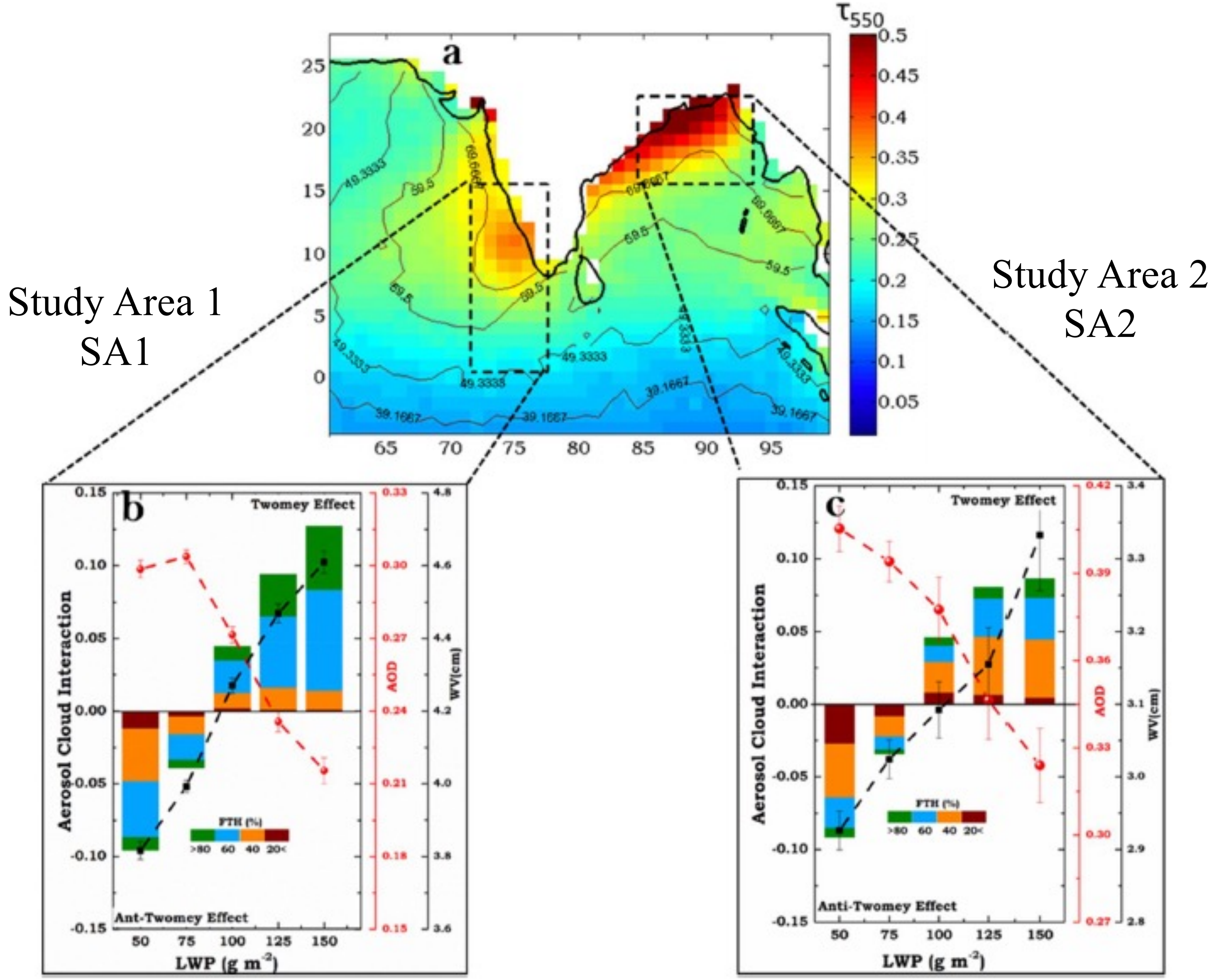
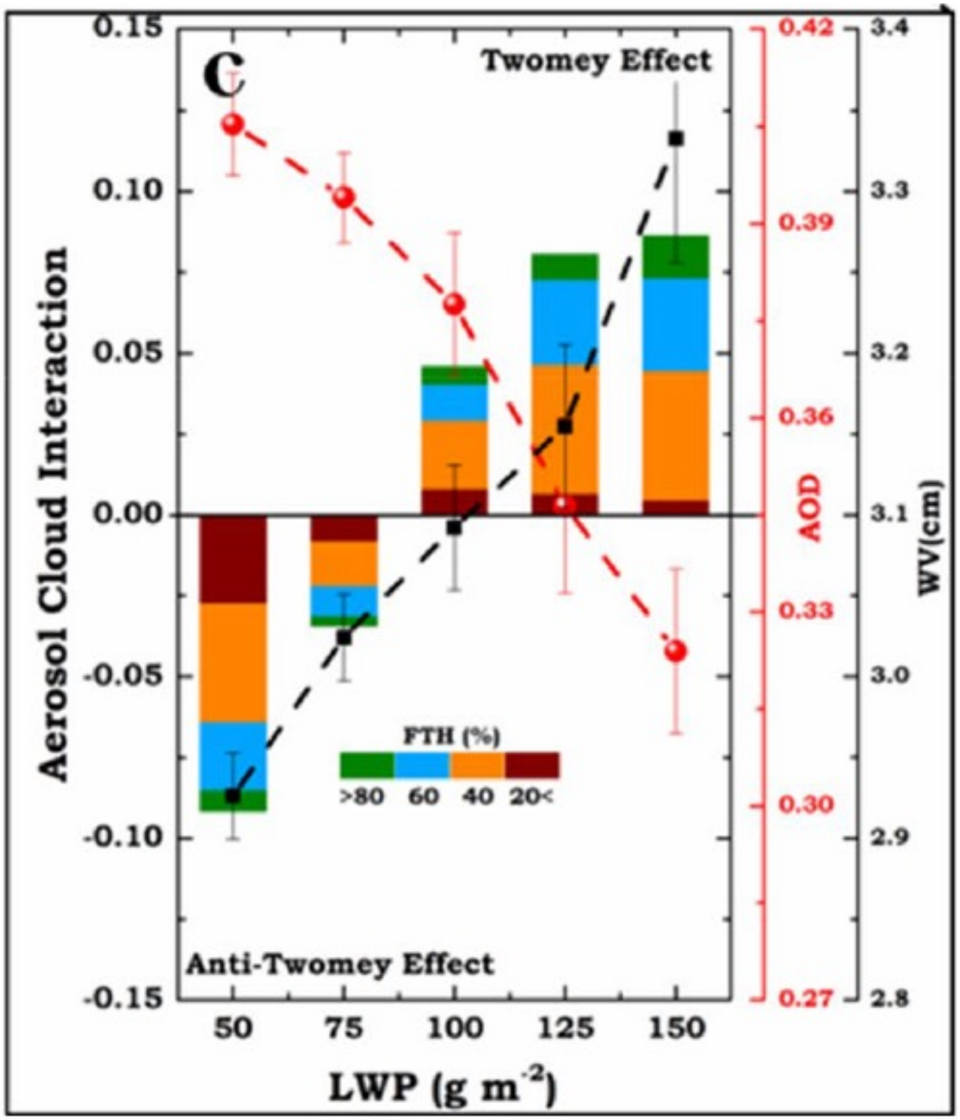


Figure 1. (**a**) Long-term (2003–2017) aerosol loading (AOD_{550}) over South Asia derived from MODIS sensor onboard Aqua satellite. Contour indicates the anthropogenic fraction to total AOD (%). Rectangular boxes represent the study area SA1 and SA2 respectively. (**b**,**c**) Aerosol cloud interaction over SA1 and SA2 respectively for different cloud liquid water path (LWP) bins. Red and black curve in (**b**,**c**) represent AOD_{550} and precipitable water vapour (PWV, cm) corresponding to respective cloud LWP bin used in the estimation of ACI. Colorbars in (**b**,**c**) represent the percentage occurrence of free tropospheric humidity (FTH_{700hPa}, %).



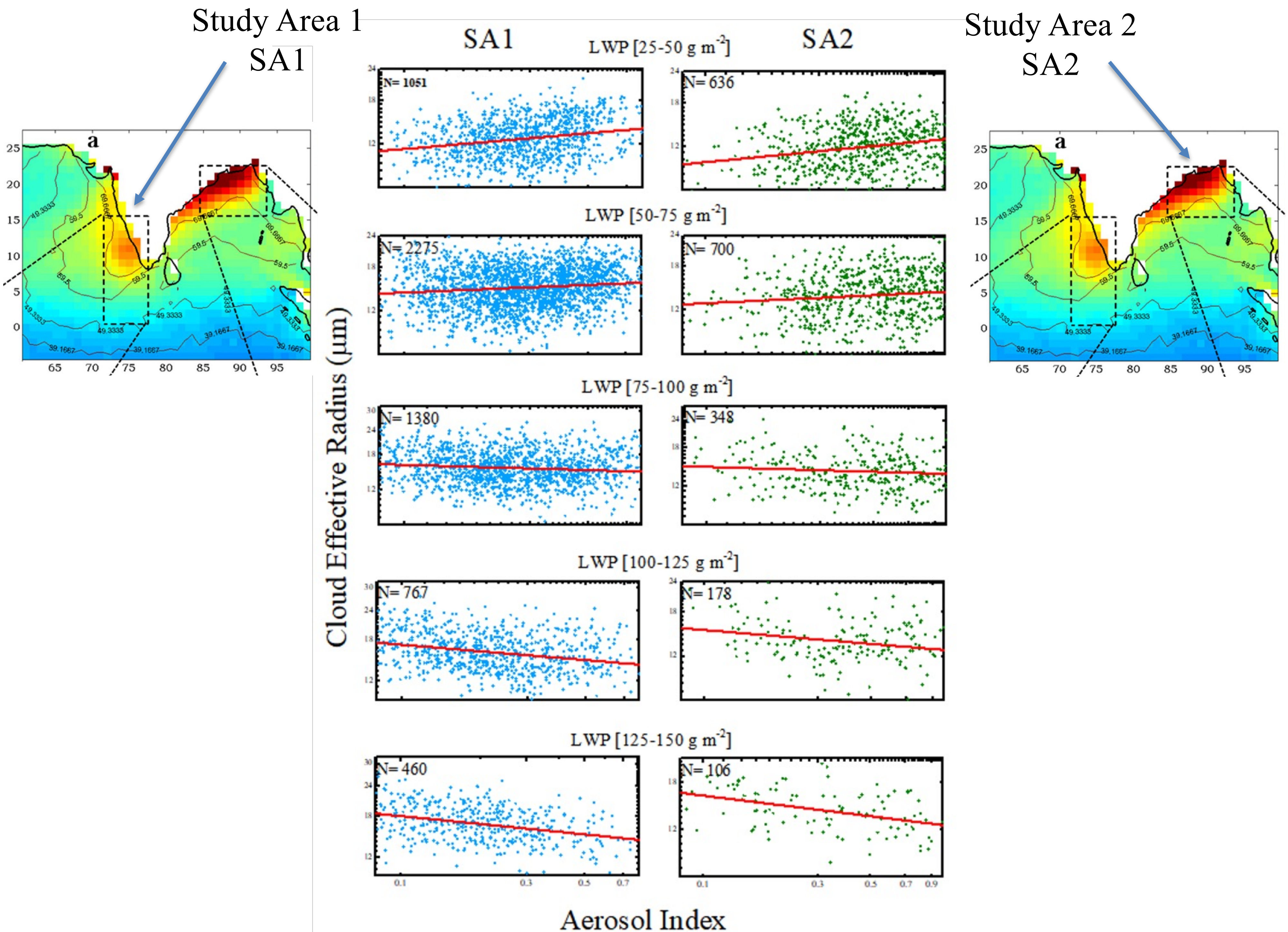
liquid water path (LWP) of 100 g m^{-2} corresponds to a 100 m deep cloud with water content of 1 g m^{-3} (about 1 g kg^{-1} at low levels as the air density is around1 kg m^{-3})…

Aerosol Cloud Interaction (ACI) is quantified for a fixed LWP is estimated as[54]:

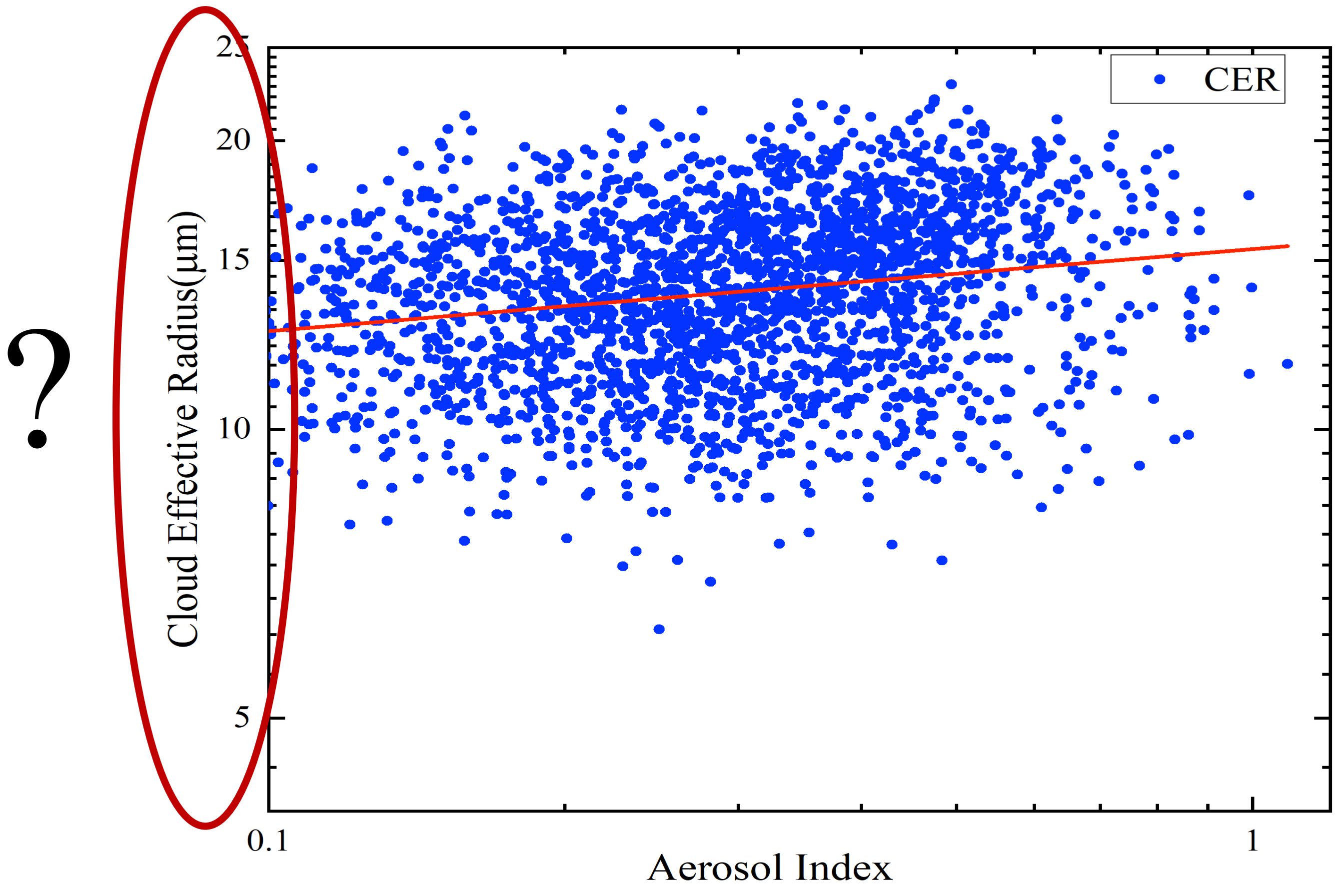
$$ACI = \frac{-\partial lnCER}{\partial lnAI}|_{LWP}$$

where, Aerosol Index (AI) is the product of AOD and AE is widely used as a proxy for CCN concentration

CER – cloud effective radius; AI – aerosol index, AI ~ AOD x AE – aerosol optical depth x Angstrom exponent



Subin Jose (personal communication)



Effective radius is different from the mean volume radius, but…

Mean volume radius in highly polluted shallow (LWP < 100 g m^{-2}) warm clouds is likely just a few microns. Can the spectral width increase it so much?

GEOPHYSICAL RESEARCH LETTERS, VOL. 27, NO. 13, PAGES 1903-1906, JULY 1, 2000

Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius

Yangang Liu* and Peter H. Daum

$$r_e = \alpha \left(\frac{L}{N} \right)^{1/3}$$

L – liquid water content
N – droplet concentration
$(L/N)^{1/3}$ ~ mean volume radius

r_e – effective radius

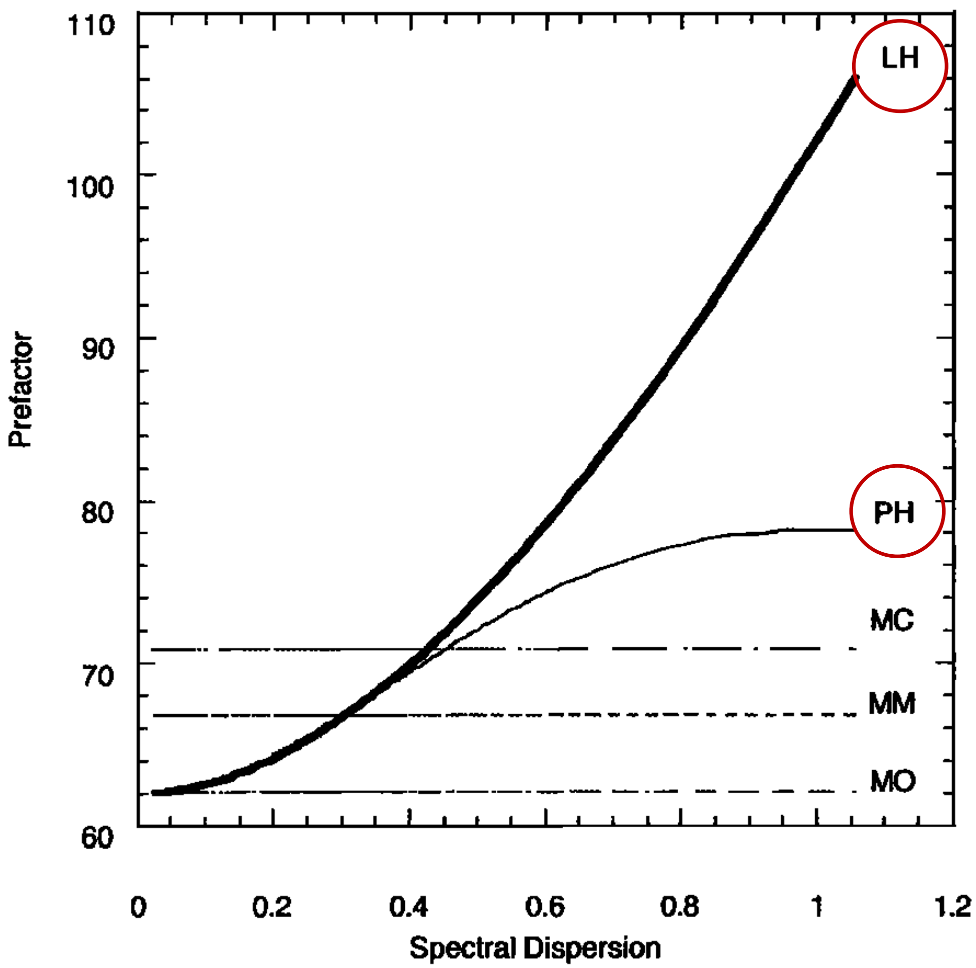
$$\alpha_{PH}(d) = 62.04 \frac{\left(1+3d^2\right)^{2/3}}{\left(1+d^2\right)},$$

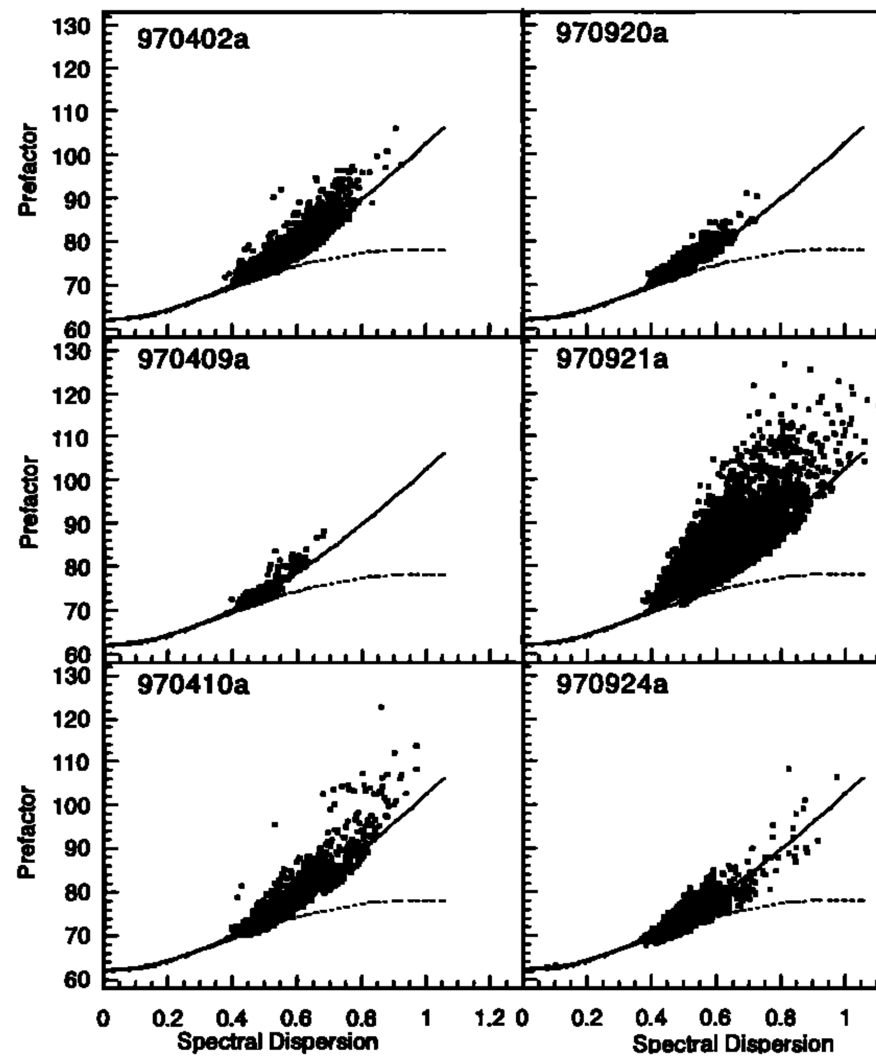
Pontikis and Hicks *GRL* 1992

where d is defined as the ratio of the standard deviation to the mean radius of the corresponding droplet size distribution.

$$\alpha_{LH}(b) = 64.52 \frac{\Gamma^{2/3}(3/b)}{\Gamma(2/b)} b^{1/3} , \quad (3)$$

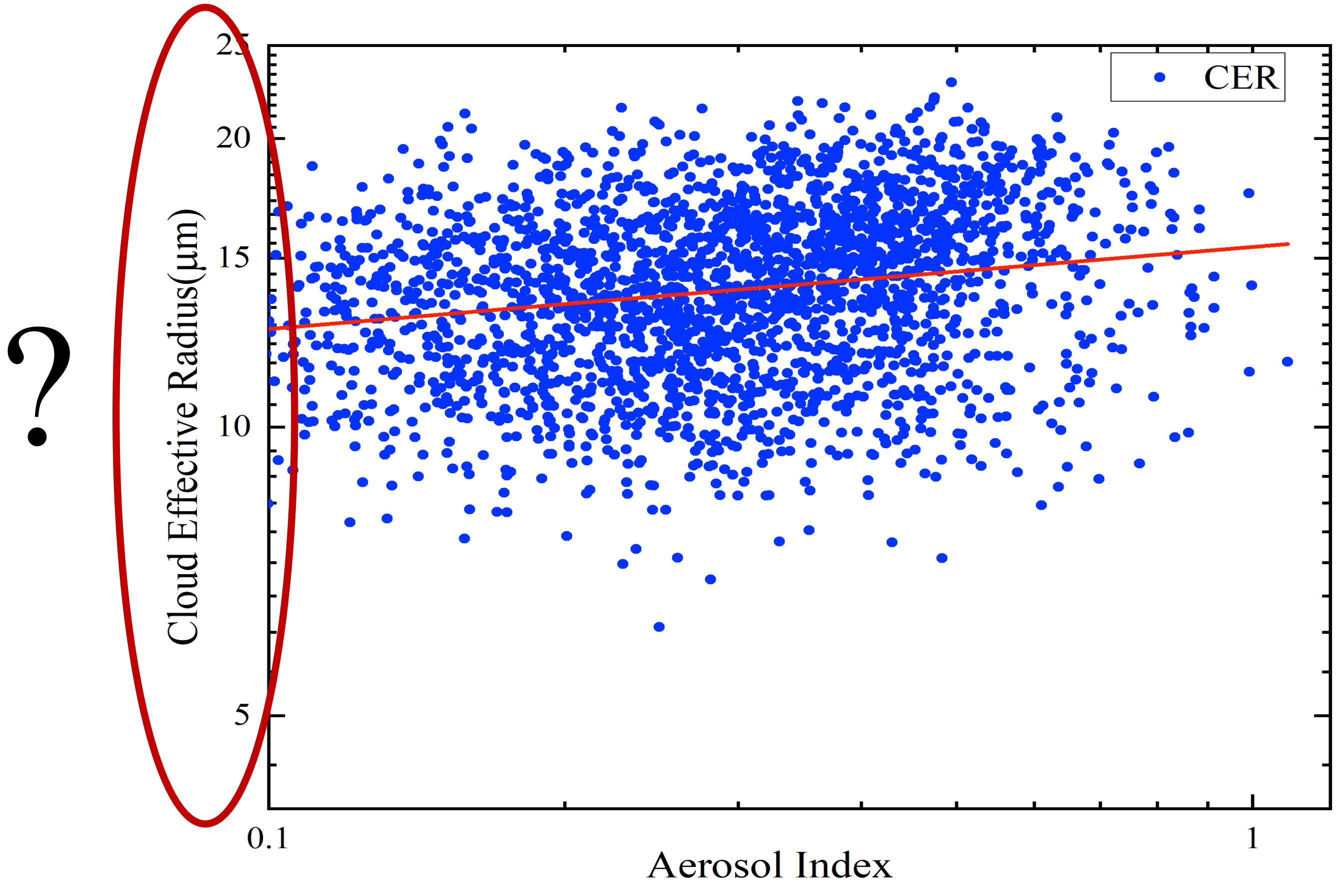
where $\Gamma(t) = \int_0^\infty z^{t-1} \exp(-z) dz$, and b is a parameter that depends on physical processes such as entrainment and mixing.





For a monodisperse droplet spectrum $r_e = r_{mv}$: effective radius = mean volume radius
For a very wide spectrum, r_e unlikely exceeds $2\ r_{mv}$

Conclusion: r_e unlikely to be over 10 microns for extremely polluted clouds…



Effective radius is different from the mean volume radius, but…

Mean volume radius in highly polluted shallow (LWP < 100 g m^{-2}) warm clouds is likely just a few microns. Can the spectral width increase it so much?

Not likely…

Geophysical Research Letters

2018

RESEARCH LETTER
10.1029/2018GL077562

Key Points:
- Cloud effective radius is positively correlated to aerosol index over three industrial regions, but negatively over

Opposite Aerosol Index-Cloud Droplet Effective Radius Correlations Over Major Industrial Regions and Their Adjacent Oceans

X. Ma[1], H. Jia[1], F. Yu[2], and J. Quaas[3]

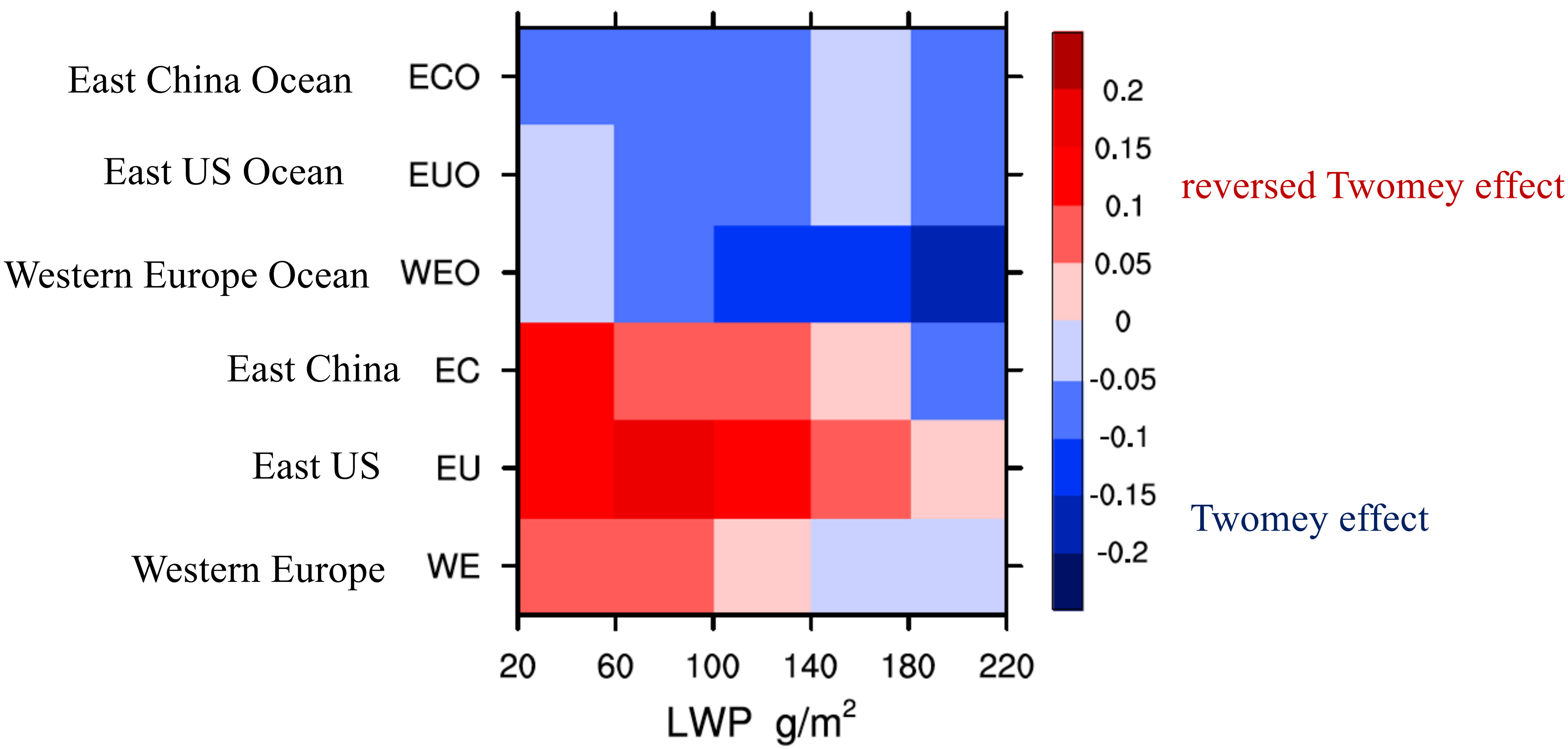


Figure 1. The computed slopes of CER versus AI on log-log scale, in which both CER and AI are stratified according to LWP.

How is it possible to have larger droplets for more polluted clouds?

How is that possible?

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 106, NO. D19, PAGES 22,907–22,922, OCTOBER 16, 2001

Analysis of smoke impact on clouds in Brazilian biomass burning regions: An extension of Twomey's approach

Graham Feingold
NOAA Environmental Technology Laboratory, Boulder, Colorado, USA

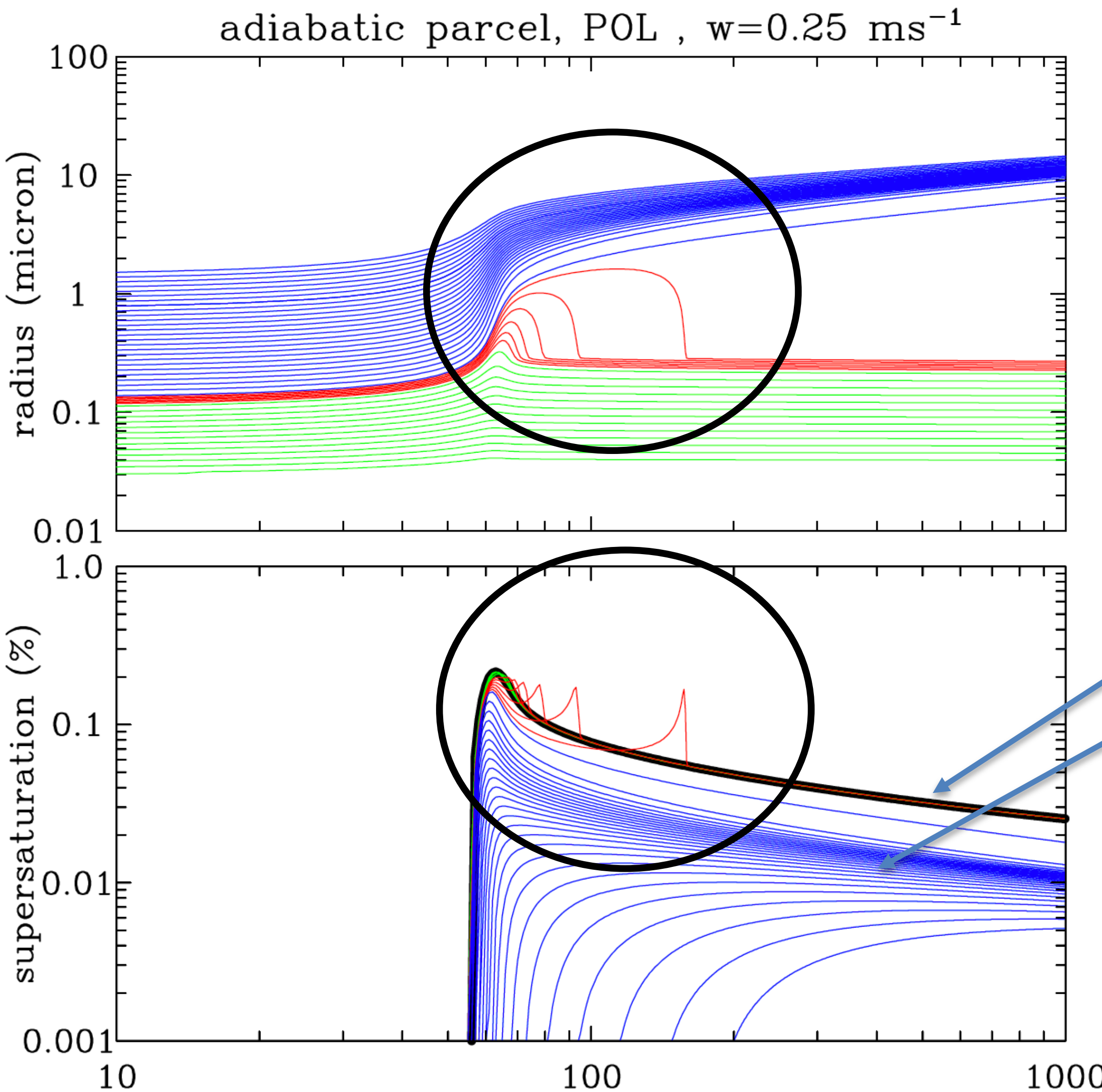
Lorraine A. Remer, Jaya Ramaprasad, and Yoram J. Kaufman
NASA Goddard Space Flight Center, Greenbelt, Maryland, USA

Three types of response are identified: (1) cloud droplet concentrations increase with increasing aerosol loading, followed by saturation in the response at high concentrations; (2) as in type 1, followed by increasing droplet concentrations with further increases in aerosol loading. This increase in droplet concentration is due to the suppression of supersaturation by abundant large particles, which prevents the activation of smaller particles. This enables renewed activation of larger particles when smoke loadings exceed some threshold; (3) as in type 1, followed by a decrease in droplet number concentrations with increasing aerosol loading as intense competition for vapor evaporates the smaller droplets. The latter implies an unexpected increase in drop size with increasing smoke loading. The conditions under which each of these responses are expected to occur are discussed. It is shown that although to first-order smoke optical depth is a good proxy for aerosol indirect forcing, under some conditions the size distribution and hygroscopicity can be important factors. We find no evidence that indirect forcing depends on precipitable water vapor.

Geophysical Research Letters 2023

Adiabatic Evolution of Cloud Droplet Spectral Width: A New Look at an Old Problem

Wojciech W. Grabowski[1] and Hanna Pawlowska[2]



weak updraft, polluted clouds…

$$\frac{dr^2}{dt} \sim (S - S_{eq})$$

black line

$$S_{eq} = \frac{a}{r} - \frac{b}{r^3}$$

Geophysical Research Letters 2023

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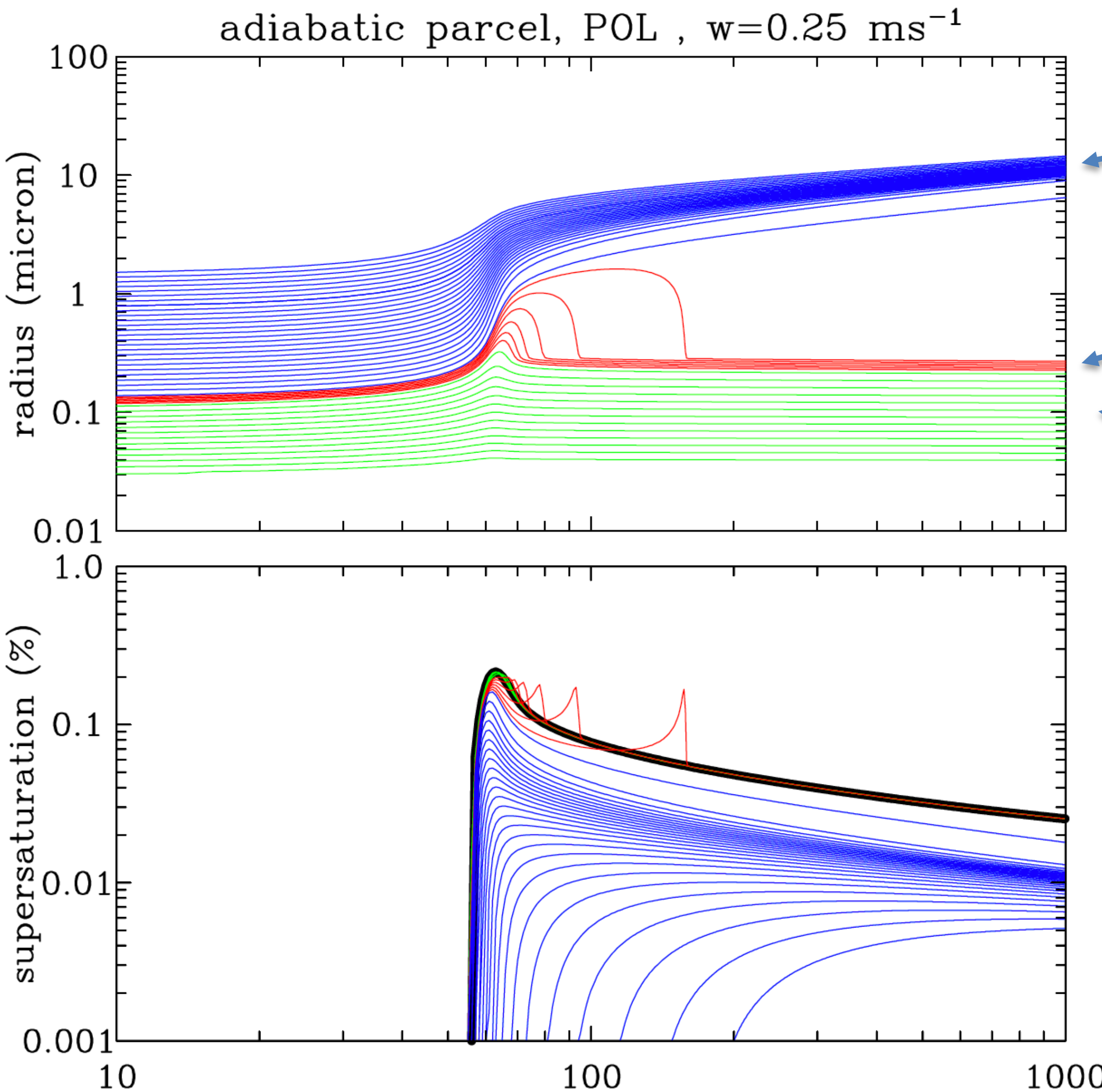


cloud droplets (activated CCN)

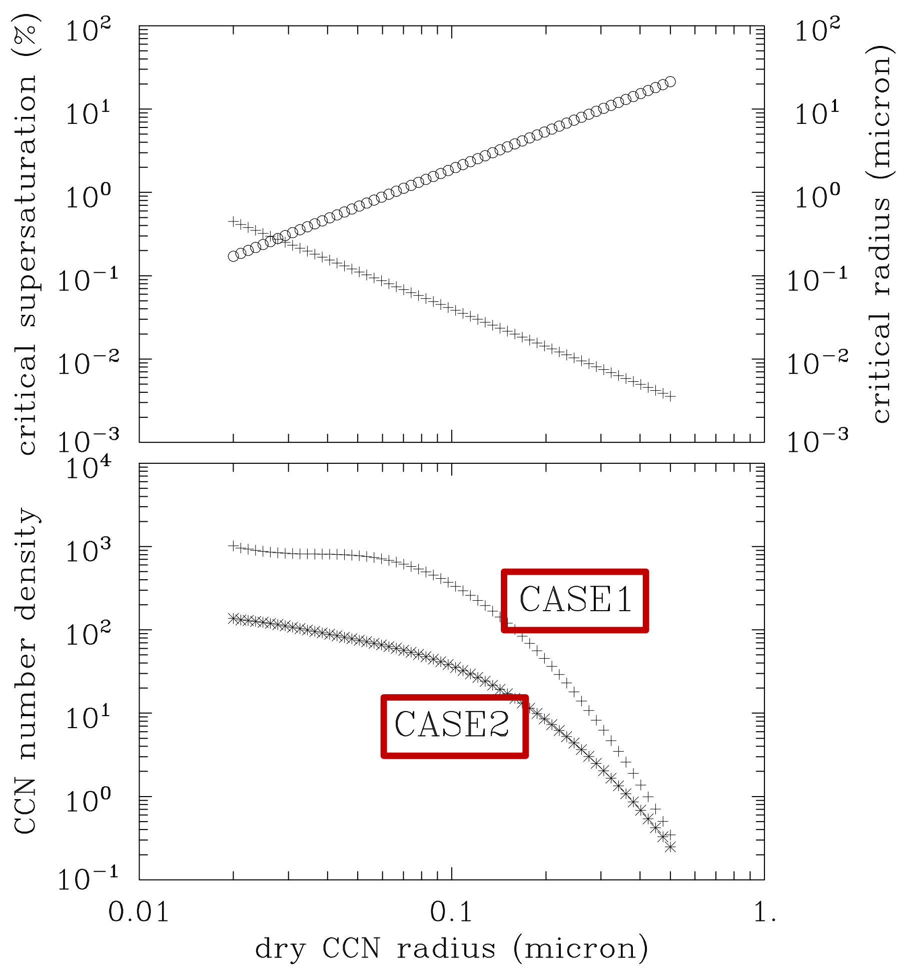
activated and then deactivated CCN

unactivated (interstitial) CCN

Simulations applying CCN observed over India (Gani et al. *ACP* 2020)

Parameters for CCN two-mode lognormal distributions

	CASE 1 Highly polluted	CASE 2 Moderately Polluted
Geometric Mean Radius	RM1 = 0.0125 um RM2 = 0.048 um	RM1 =0.016 um RM2 = 0.051 um
Standard Deviation	Sigma1 = 1.7 Sigma2 = 1.82	Sigma1 = 1.8 Sigma2 = 2.0
Total aerosol concentration	NANEWI =26.5 *10^9/m3 NANEW2 =20.5 *10^9/m3	NANEWI =32.0 *10^8/m3 NANEW2 =18.35 *10^8/m3



Total concentration:	47,000	5,035
Concentration (cm^{-3}) in 20 to 500 nm range:	24,500	2,870

60 CCN bins, equally spaced in log scale between 20 and 500 nm

Adiabatic parcel simulations as in Grabowski and Pawlowska (GRL 2023):

Run simulations with CASE1 and CASE2 aerosols with different updrafts, 0.25, 1, and 4 m s^{-1}, and compare results.

$$c_p \frac{dT}{dt} = -gw + L_v C,$$

T – temperature
q_v – water vapor mixing ratio
w – updraft speed

C – condensation rate $\quad C = \frac{d}{dt} \sum_i \frac{4}{3} \pi r_i^3 N_i \frac{\rho_w}{\rho_0}$

$$\frac{dq_v}{dt} = -C,$$

$g = 9.81$ ms^{-2} – gravitational acceleration
$L_v = 2.5 \times 10^6$ J/kg - latent heat of condensation

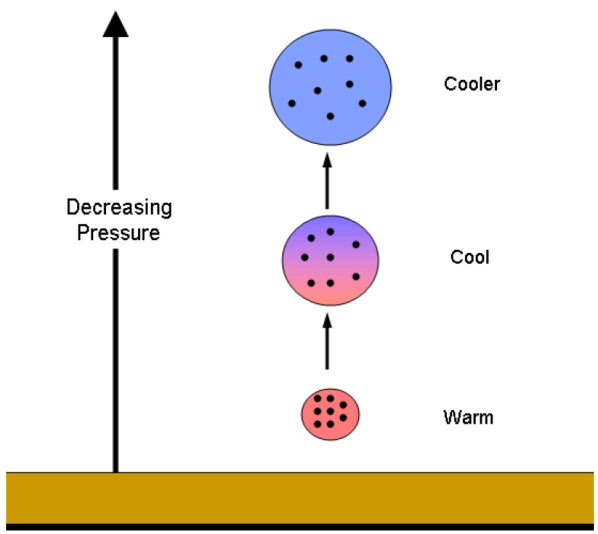
$$\frac{dp}{dt} = -\rho_0 w g,$$

p – environmental air pressure
ρ_0 – environmental air density (1 kg m^{-3})

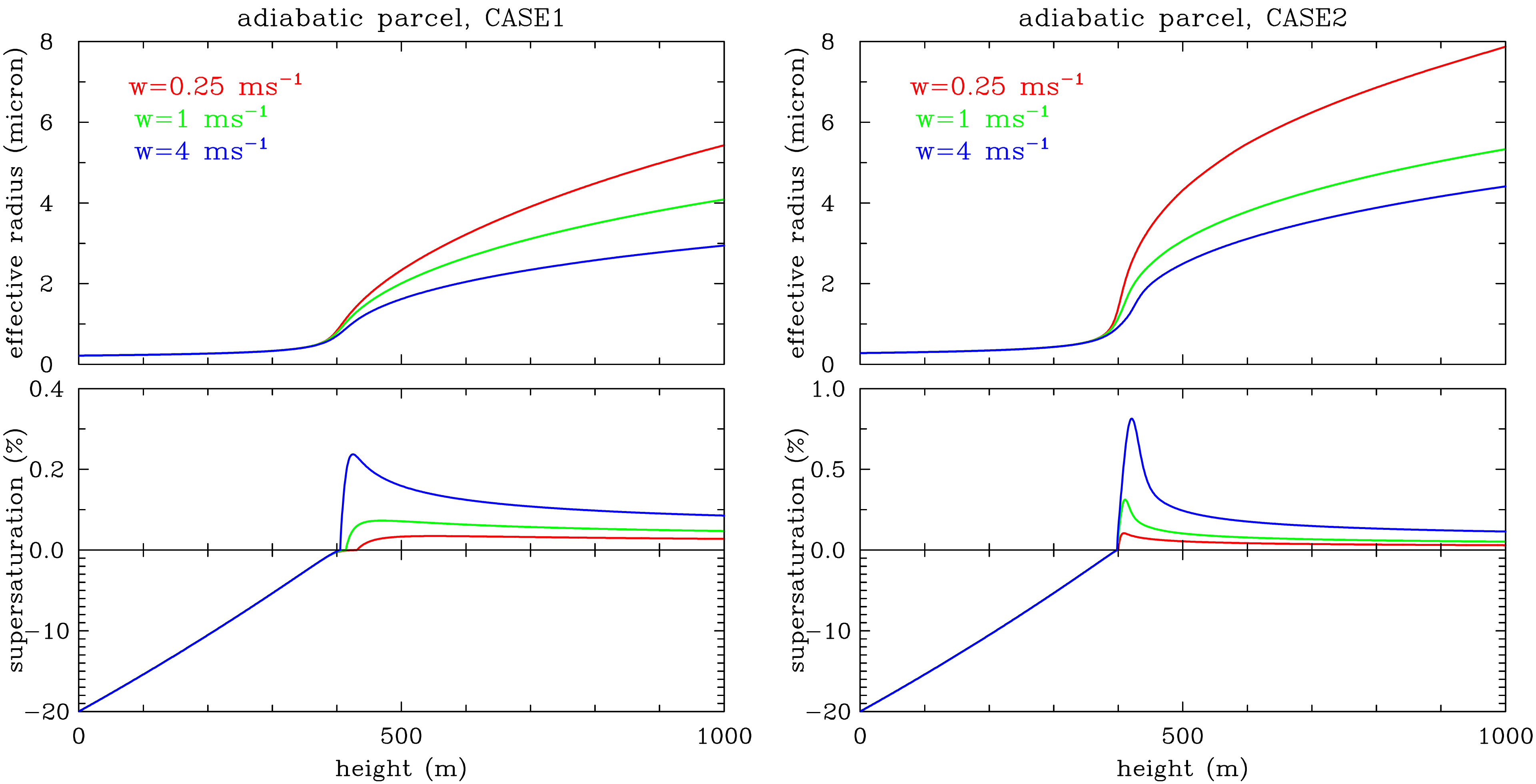
$$\frac{dr^2}{dt} \sim (S - S_{eq})$$

r – droplet radius
S – supersaturation ($S = q_v/q_{vs} - 1$)

$$S_{eq} = \frac{a}{r} - \frac{b}{r^3}$$

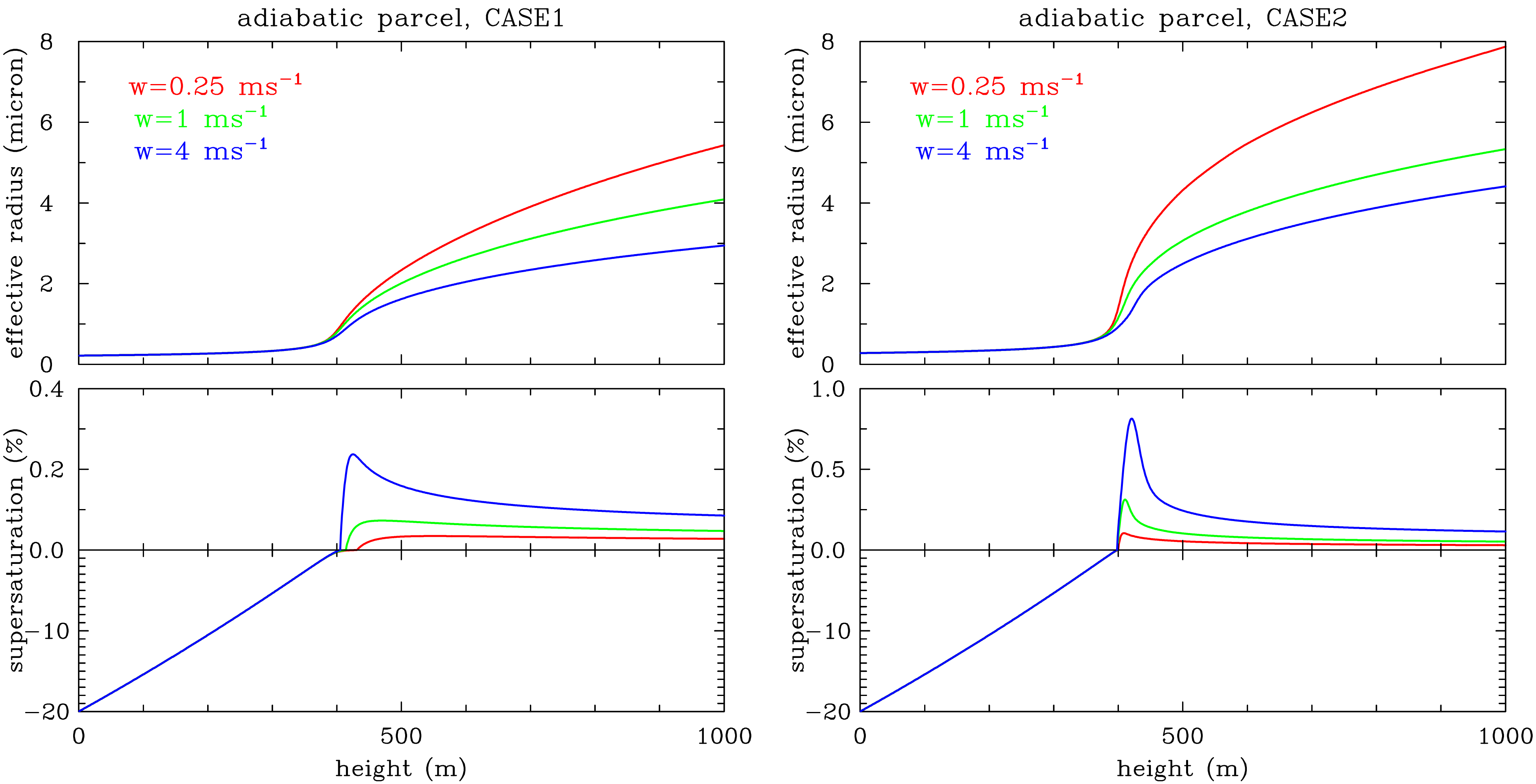


Adiabatic parcel simulations as in Grabowski and Pawlowska (GRL 2023)



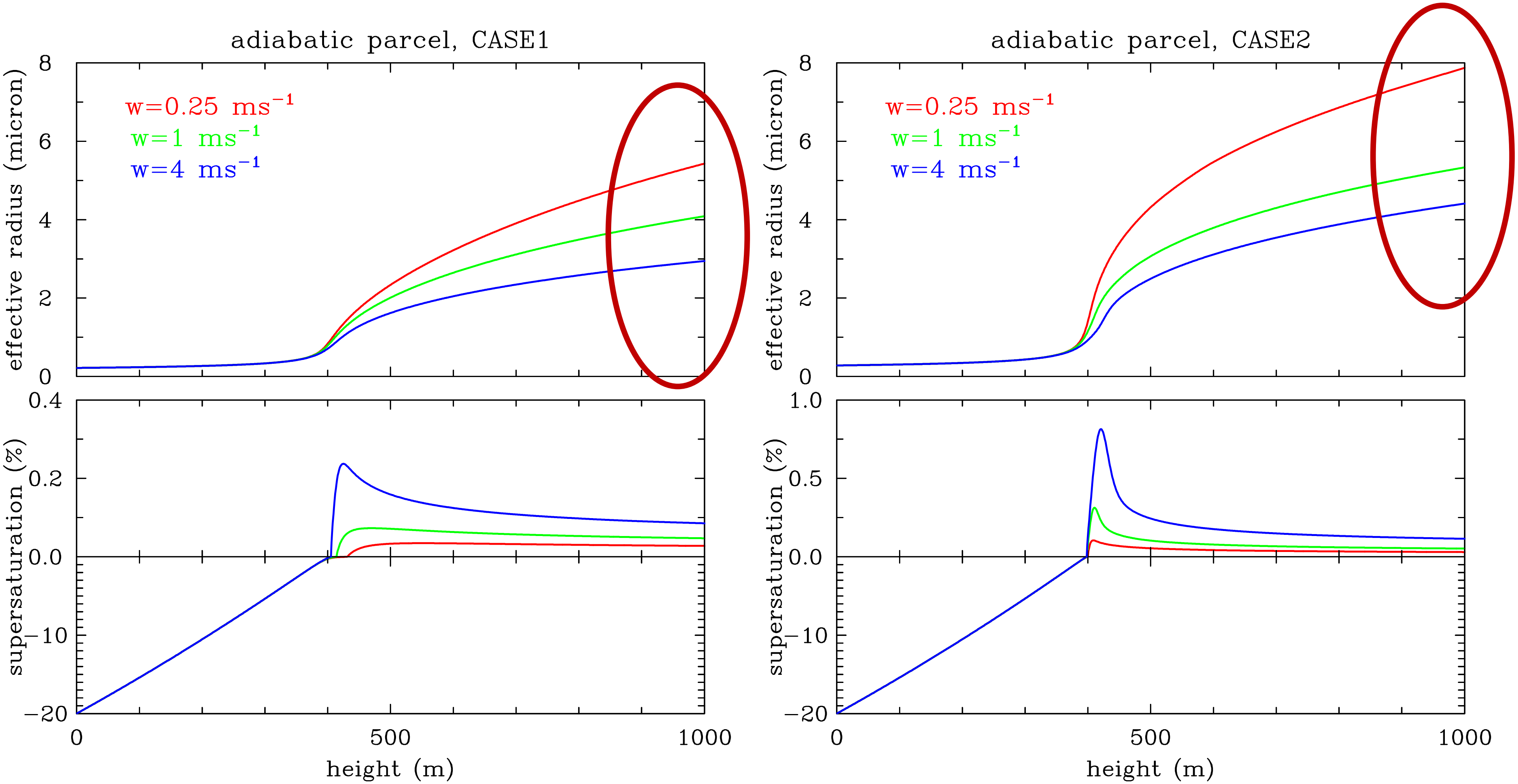
effective radius: ratio of the third and second moments of the droplet size distribution is the key parameter for the radiative transfer; this is what is “observed” by satellites…

Adiabatic parcel simulations as in Grabowski and Pawlowska (GRL 2023)



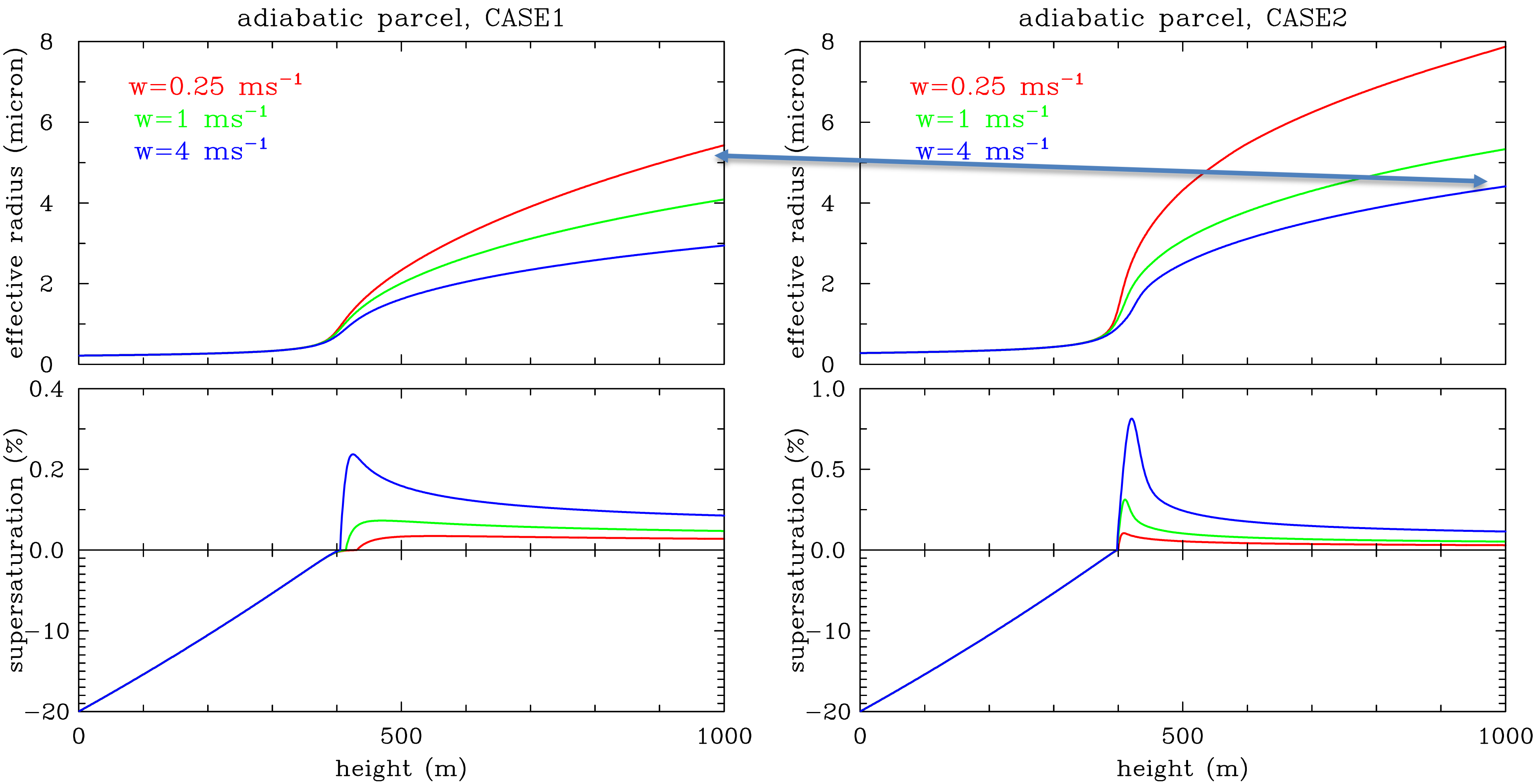
Larger vertical velocity implies higher cloud base supersaturation and thus higher concentration of cloud droplets; this gives smaller mean radius and thus smaller effective radius…

Adiabatic parcel simulations as in Grabowski and Pawlowska (GRL 2023)



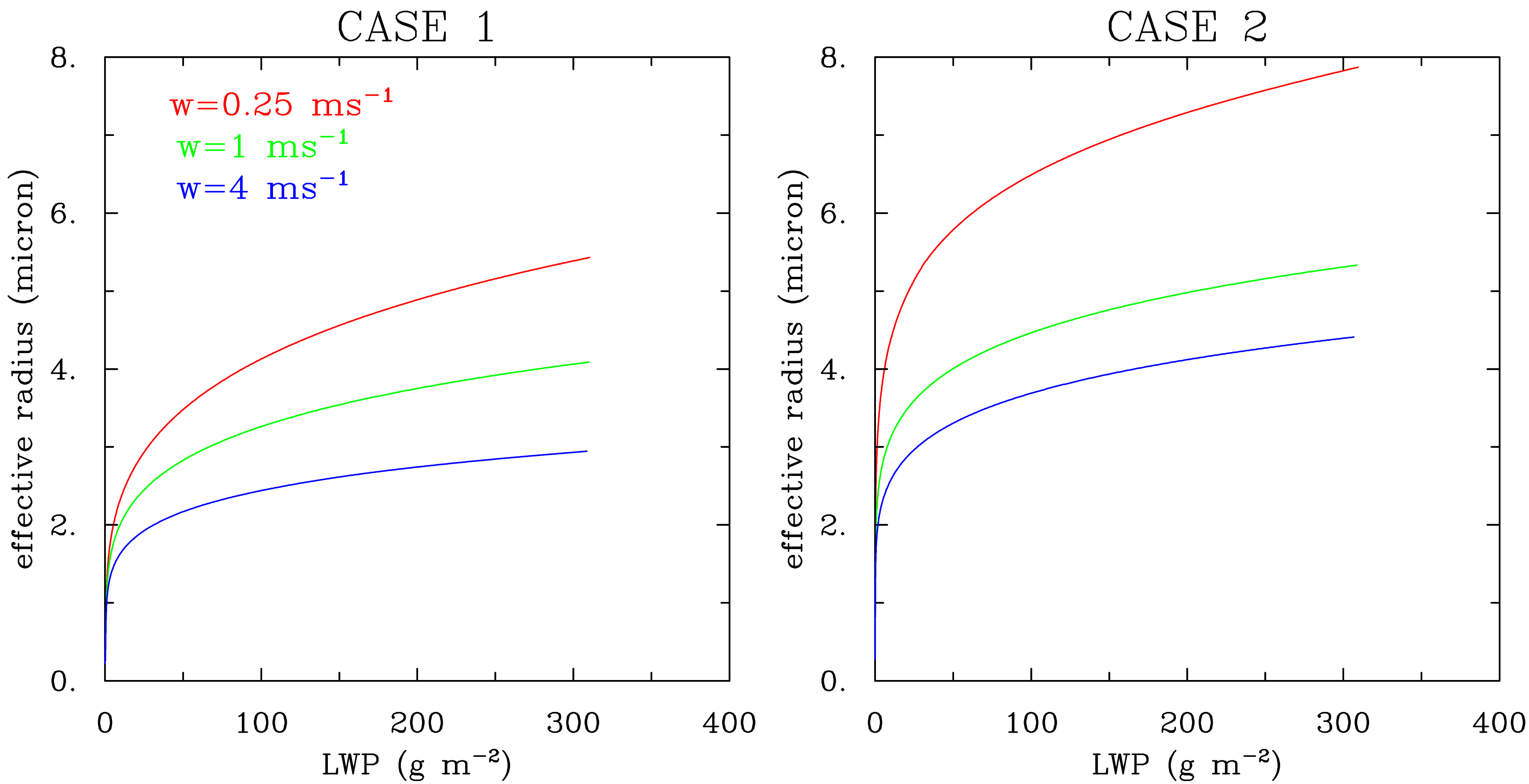
CASE1 has smaller effective radii than CASE2 for the same vertical velocity:
this agrees with the Twomey effect!

Adiabatic parcel simulations as in Grabowski and Pawlowska (GRL 2023)

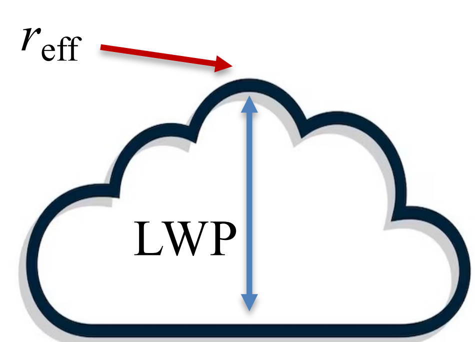


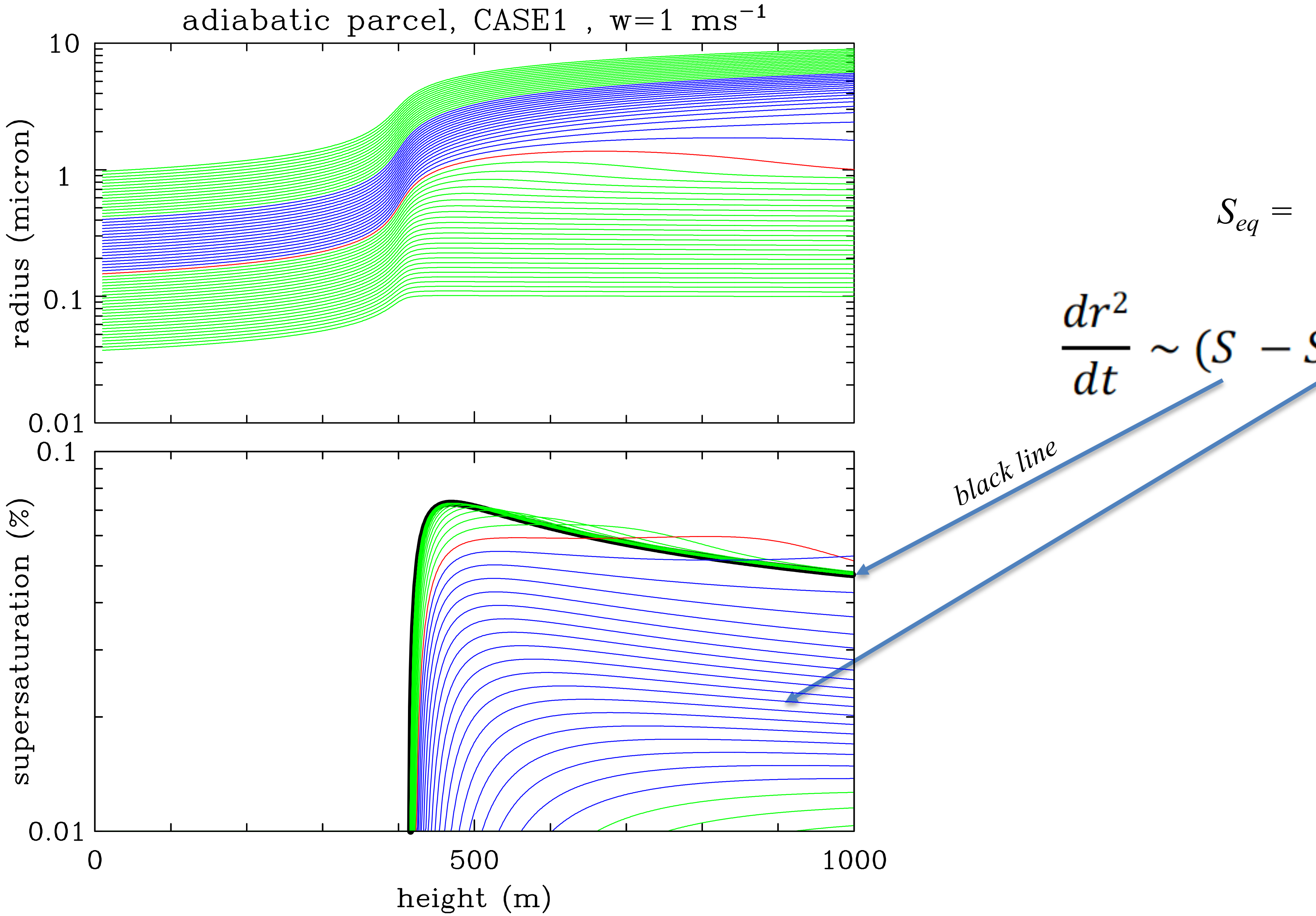
CASE1 weak updraft has larger effective radius than CASE2 strong updraft:

Does this explain reversed Twomey effect? Unlikely…



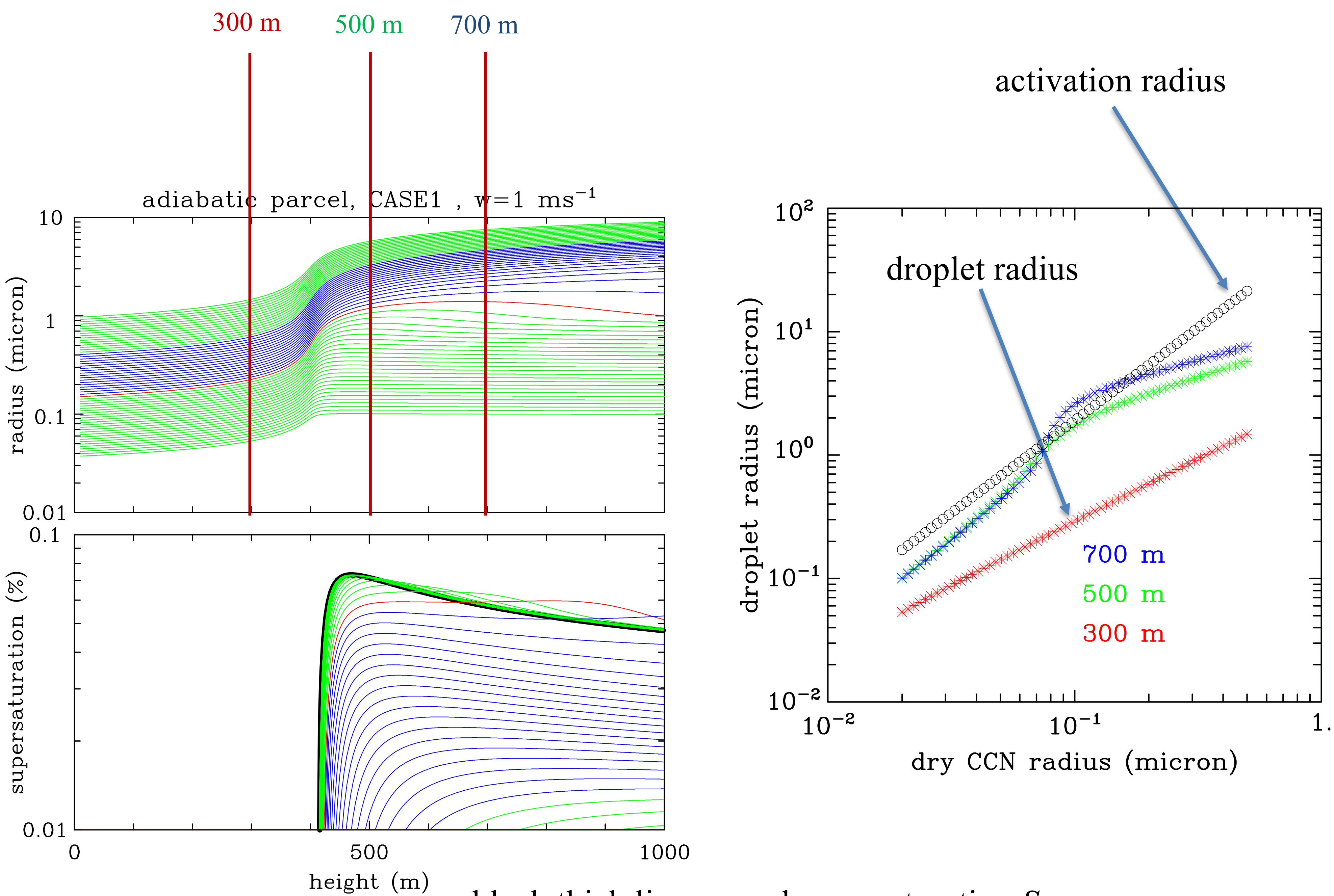
Effective radius at height z is combined with LWP (vertical integral of cloud water content) below the height z.





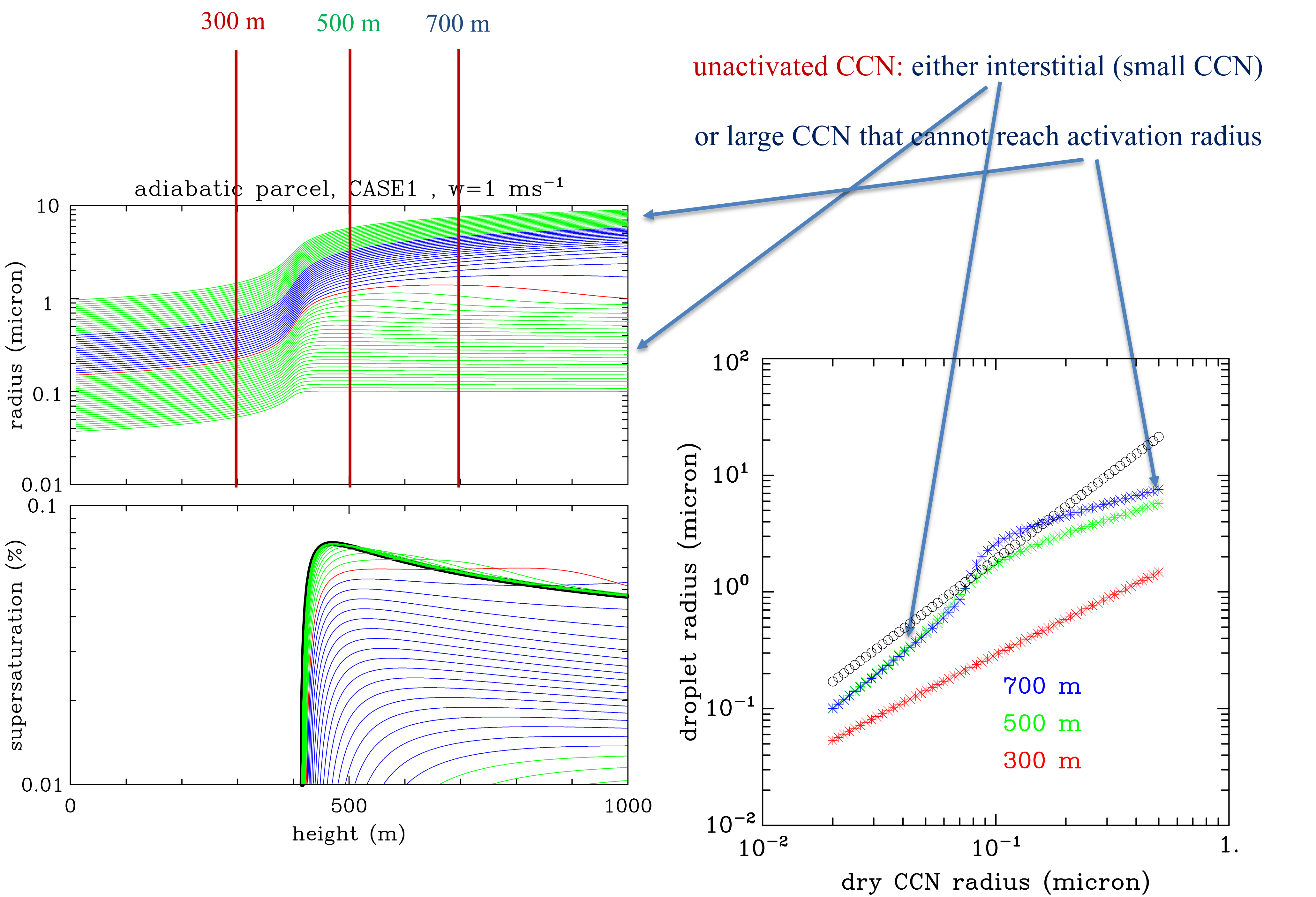
$$S_{eq} = \frac{a}{r} - \frac{b}{r^3}$$

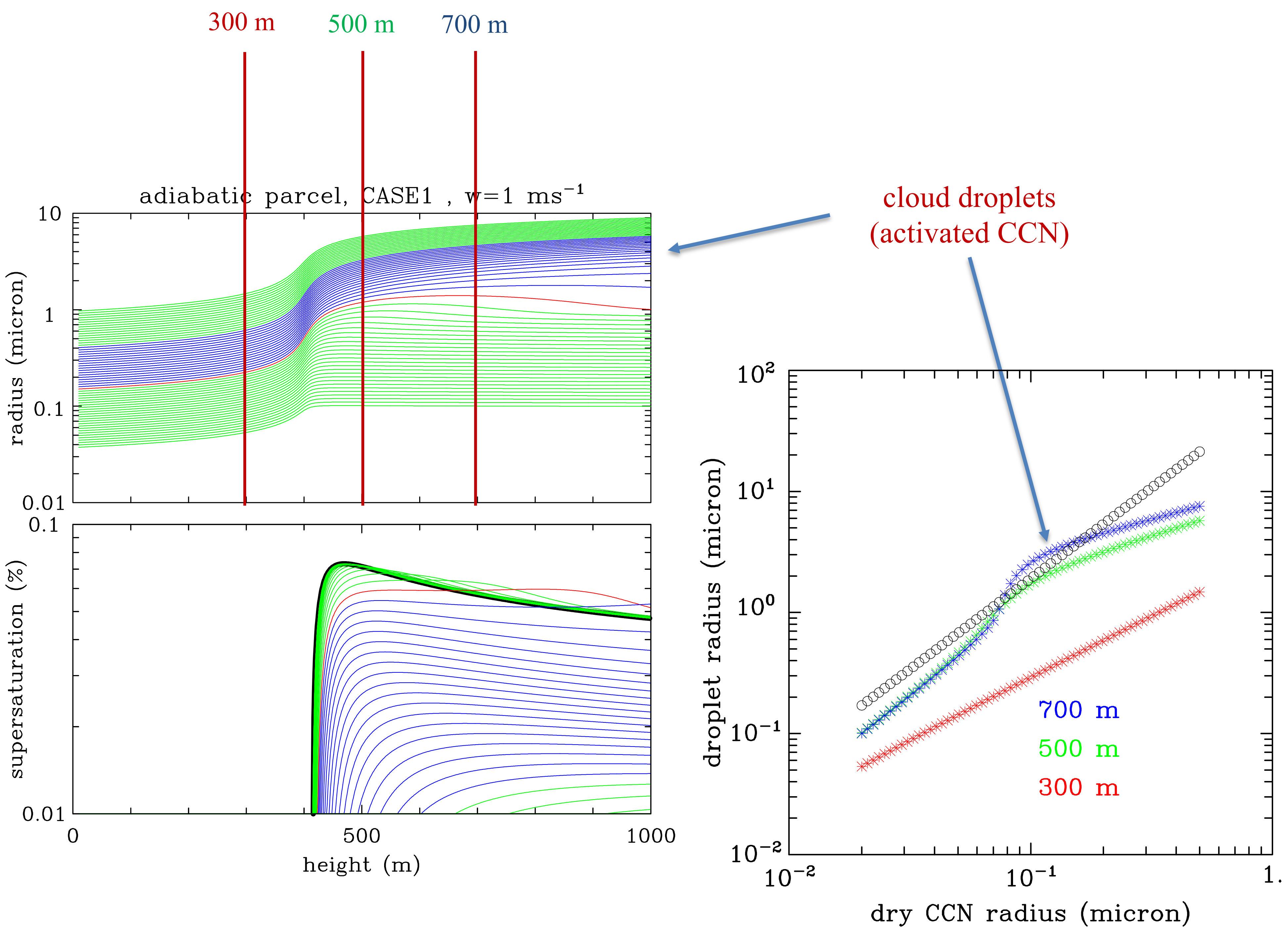
$$\frac{dr^2}{dt} \sim (S - S_{eq})$$



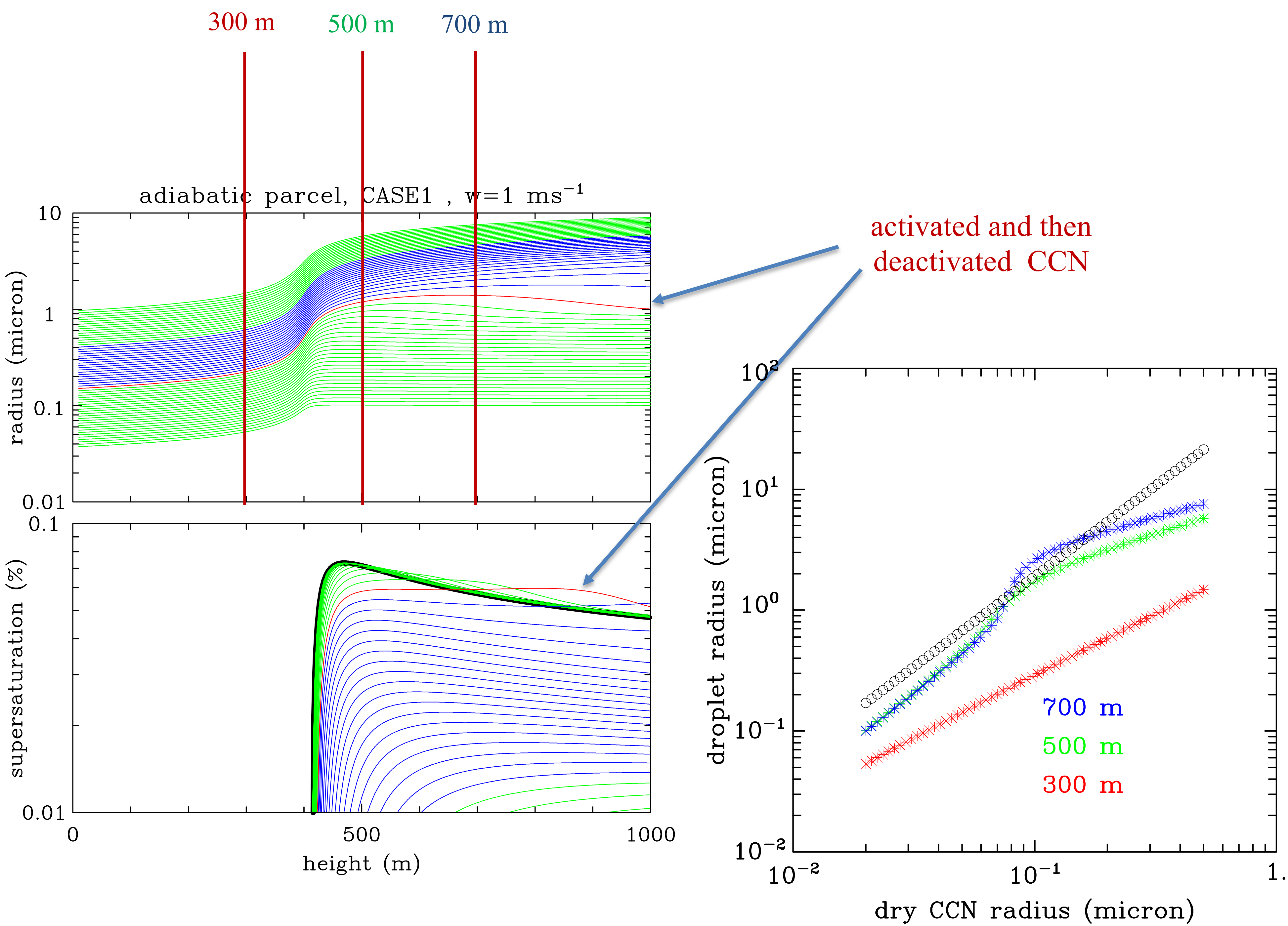
black thick line: parcel supersaturation S

green/blue/red lines – equilibrium supersaturation S_{eq}

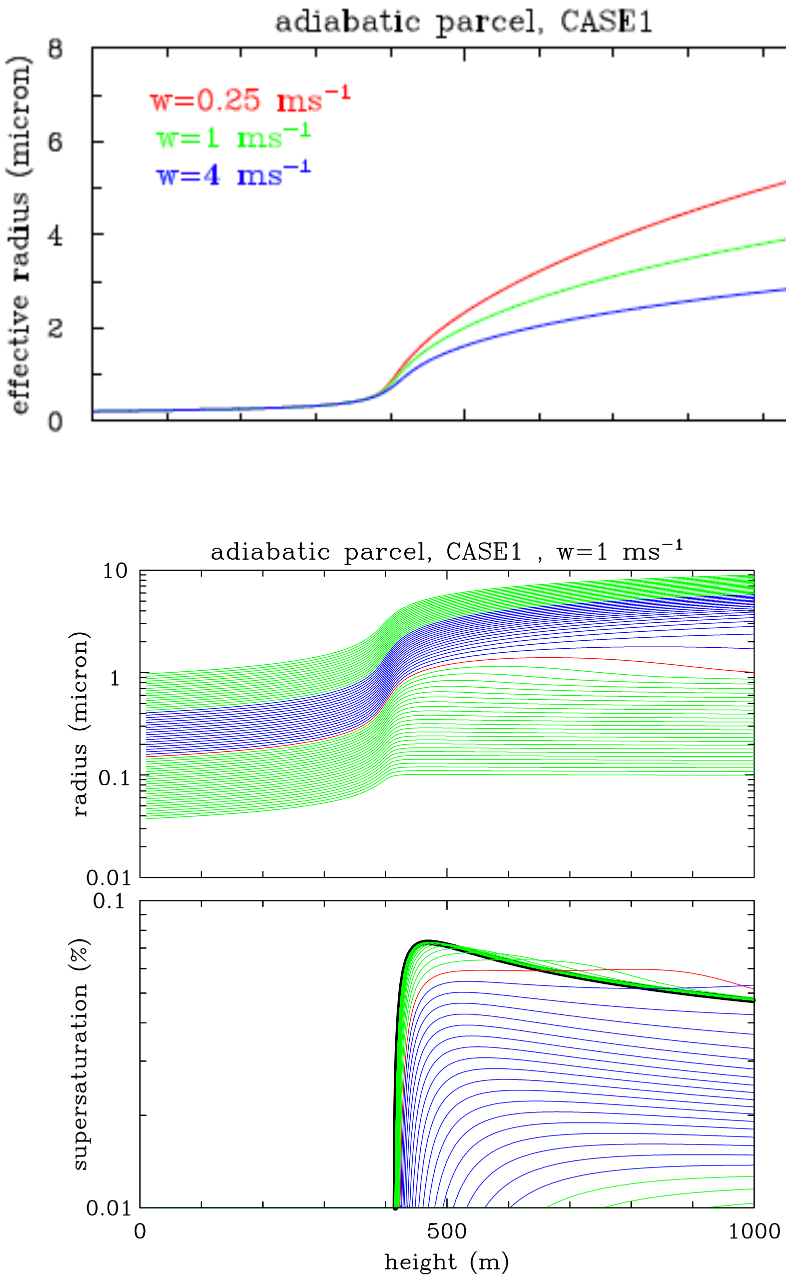

300 m
500 m
700 m
unactivated CCN: either interstitial (small CCN)
or large CCN that cannot reach activation radius
adiabatic parcel, CASE1 , w=1 ms^{-1}
radius (micron)
supersaturation (%)
height (m)
droplet radius (micron)
dry CCN radius (micron)
700 m
500 m
300 m



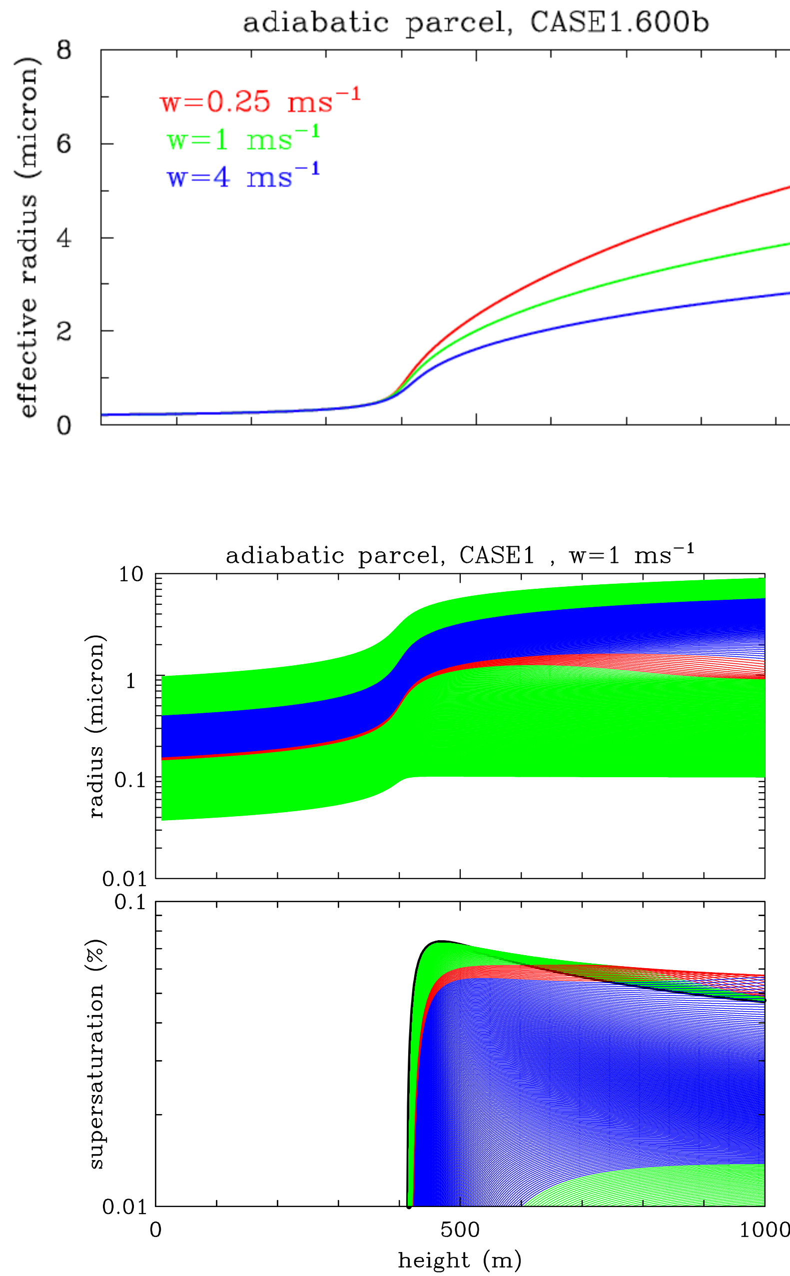
300 m
500 m
700 m
adiabatic parcel, CASE1 , w=1 ms⁻¹
cloud droplets
(activated CCN)
10
1
0.1
0.01
radius (micron)
0.1
0.01
supersaturation (%)
0
500
1000
height (m)
droplet radius (micron)
10²
10¹
10⁰
10⁻¹
10⁻²
700 m
500 m
300 m
10⁻²
10⁻¹
1.
dry CCN radius (micron)


300 m
500 m
700 m
adiabatic parcel, CASE1 , w=1 ms⁻¹
radius (micron)
supersaturation (%)
height (m)
activated and then
deactivated CCN
droplet radius (micron)
dry CCN radius (micron)
700 m
500 m
300 m

60 CCN bins



600 CCN bins



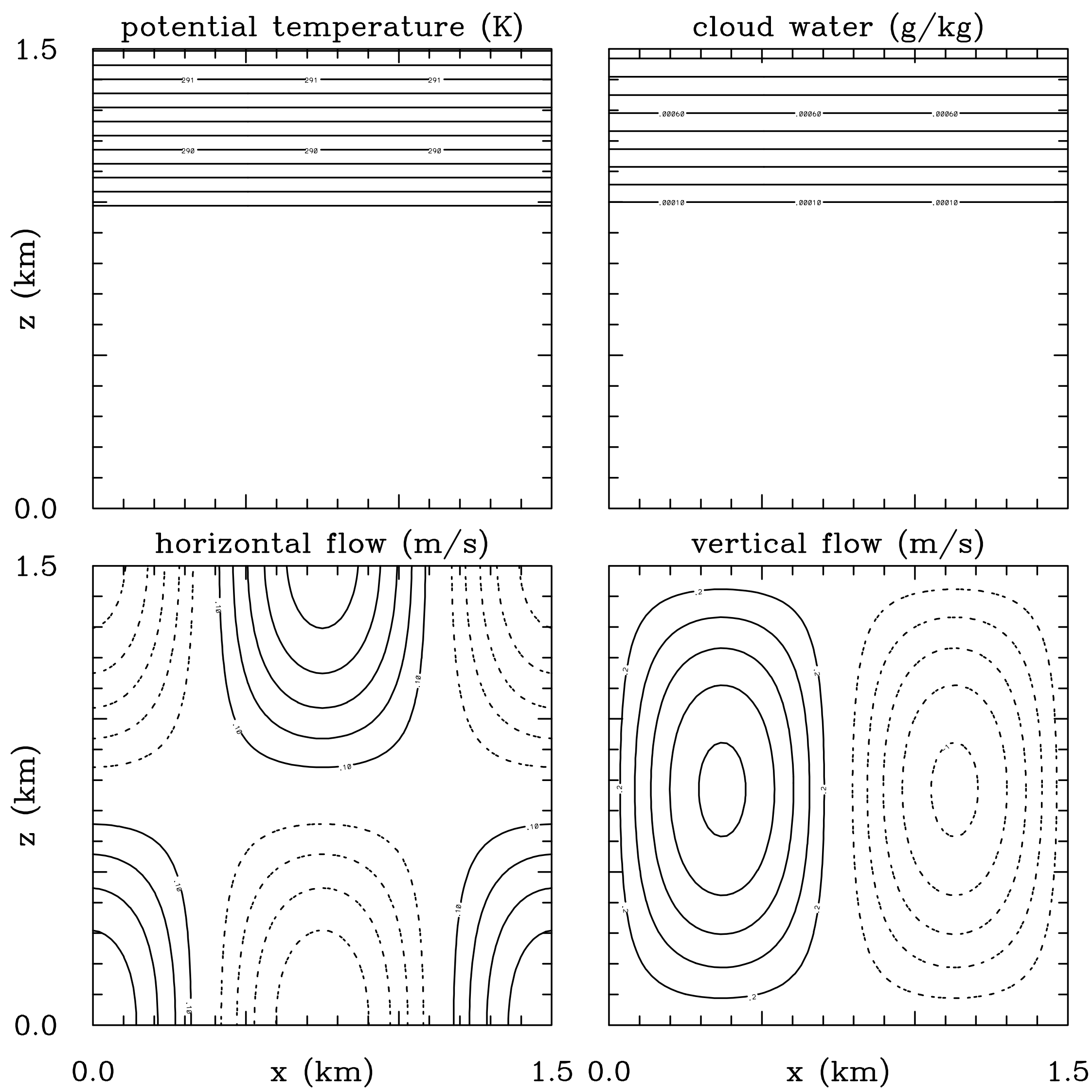
Evaporation of some already activated droplets leads to no reversal of the Twomey effect in adiabatic parcel simulations.

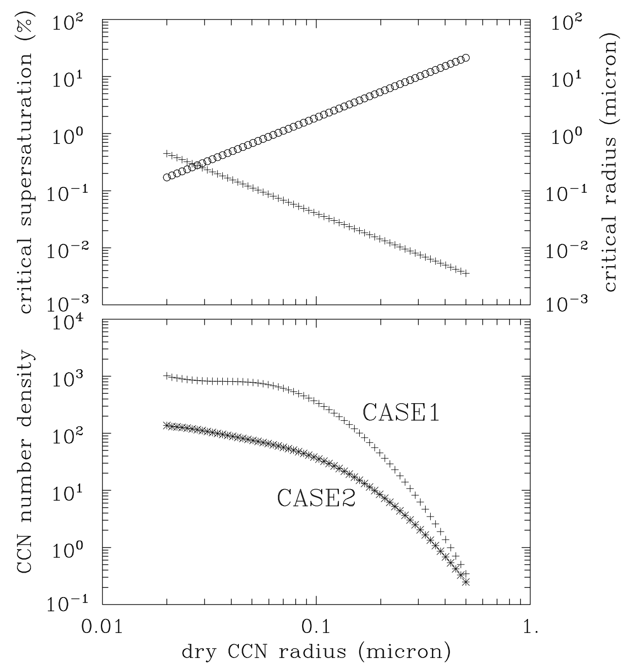
What about a more realistic simulation framework?

Kinematic (prescribed flow) simulation allows a range of updrafts at the cloud base. Let us try that…

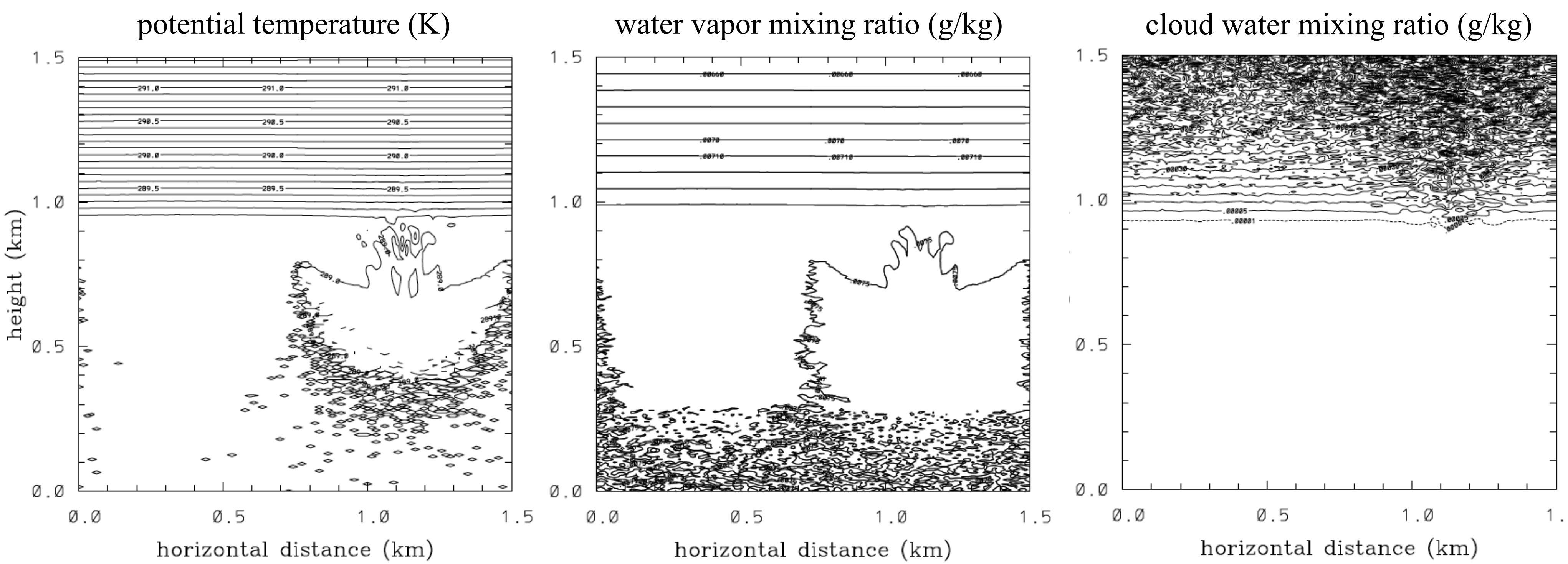
2D prescribed flow model with superdroplets mimicking a stratocumulus cloud:

1.5 x 1.5 km^2 domain, about 1 m/s maximum updraft, about 1 g/kg cloud water near the top;
5/15 m vertical/horizontal grid length, 60 superdroplets per grid box;
condensation/evaporation only;
run for 2 hours that is sufficient to achieve approximate steady state.

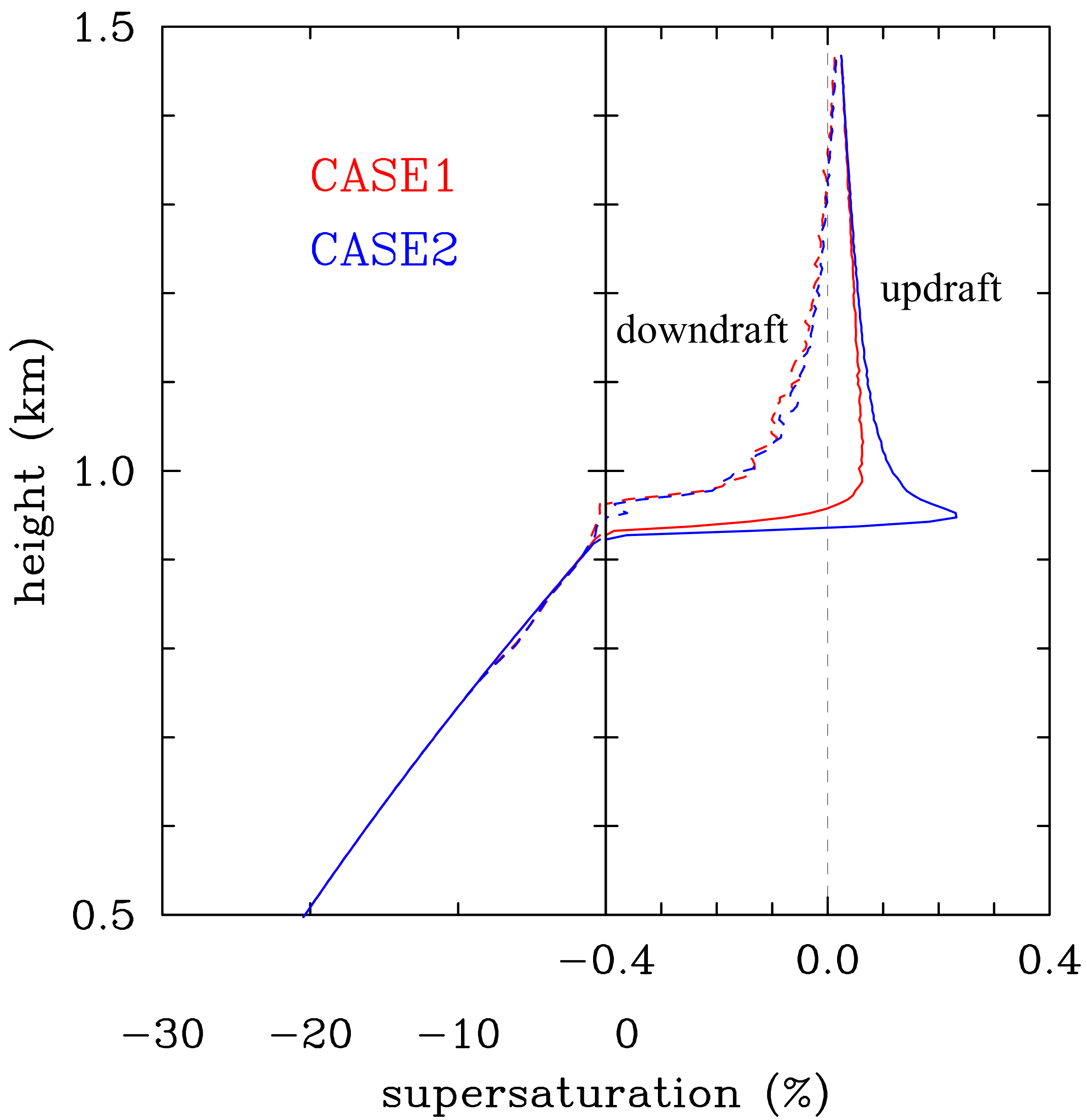




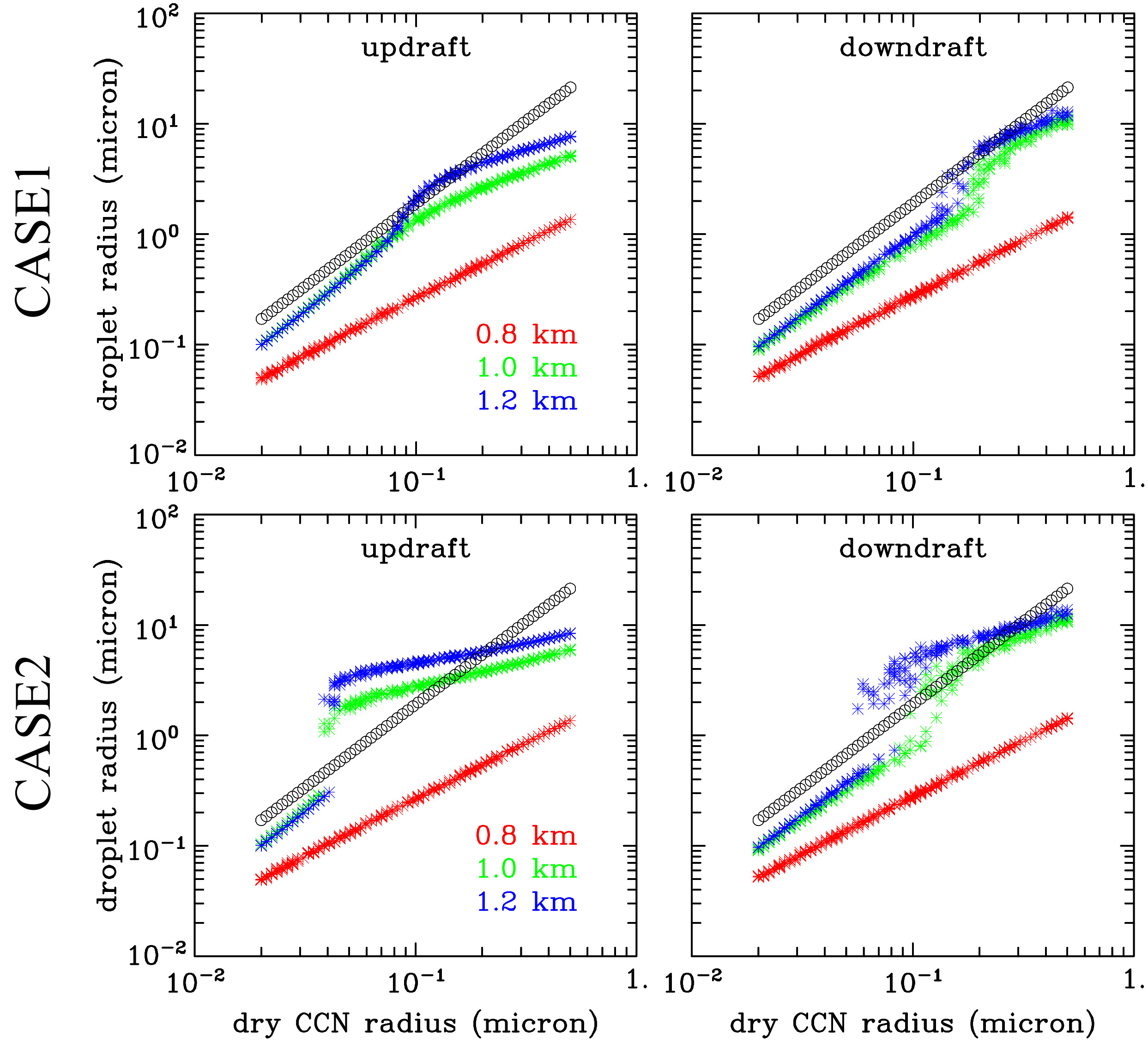
Kinematic model results after 2 hours



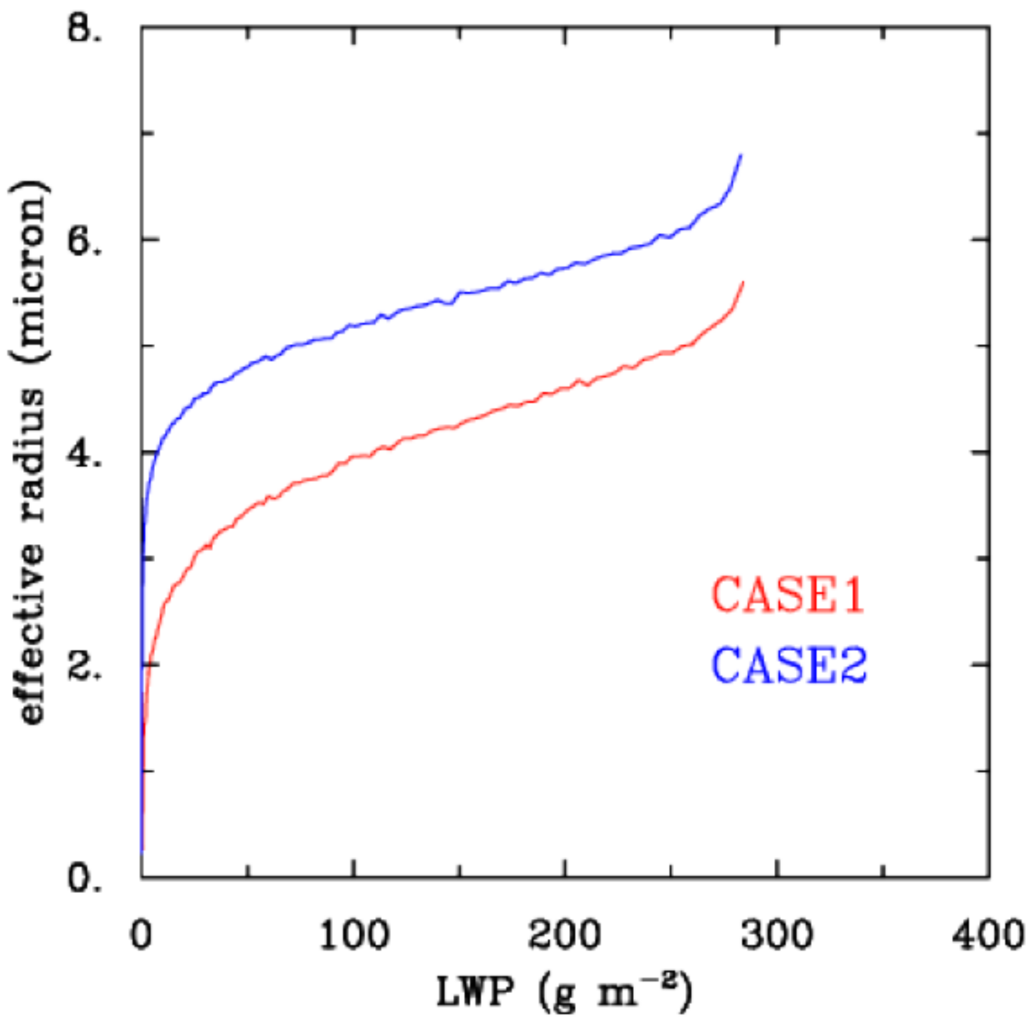
Noise in the figures comes from stochasticity of the super-droplet (SD) transport in the physical space: with 60 SDs, standard deviation of the number of SDs per grid volume is about sqrt(60) or about 8, that is, 13%...

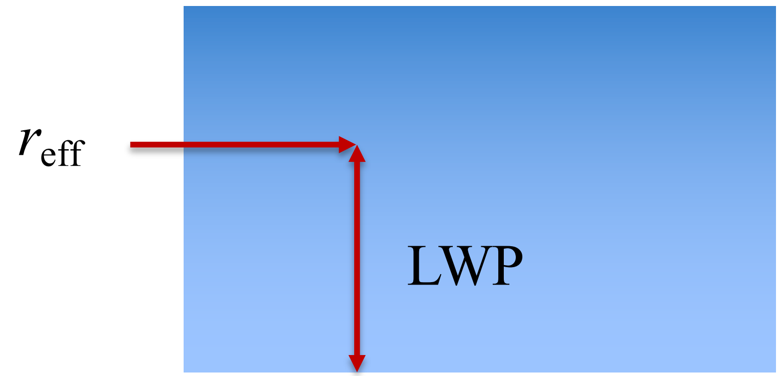


Mean saturation profiles near the center of the updraft and downdraft



Droplet radius versus dry CCN radius near the center of updraft and downdraft for CASE1 and CASE2.





Effective radius at height *z* is combined with LWP (vertical integral of cloud water content) below the height *z*. Horizontally-averaged over all model columns.

No reversal of the Twomey effect in kinematic flow simulations either…

What about 3D cloud field simulations?
These include physical processes excluded in previous simulations, entrainment and mixing in particular…

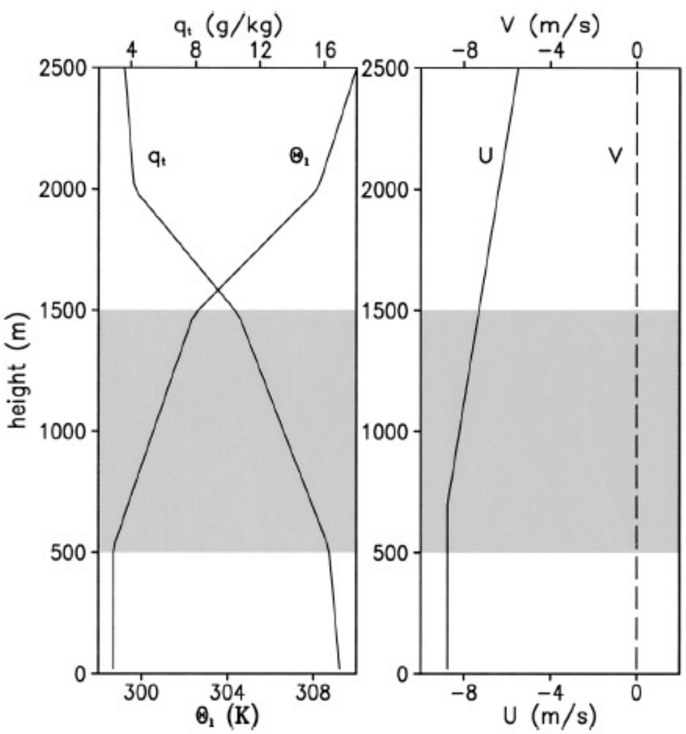
CM1 cloud model with Shima's SDM
BOMEX setup, 100 m horizontal 40 m vertical grid lengths

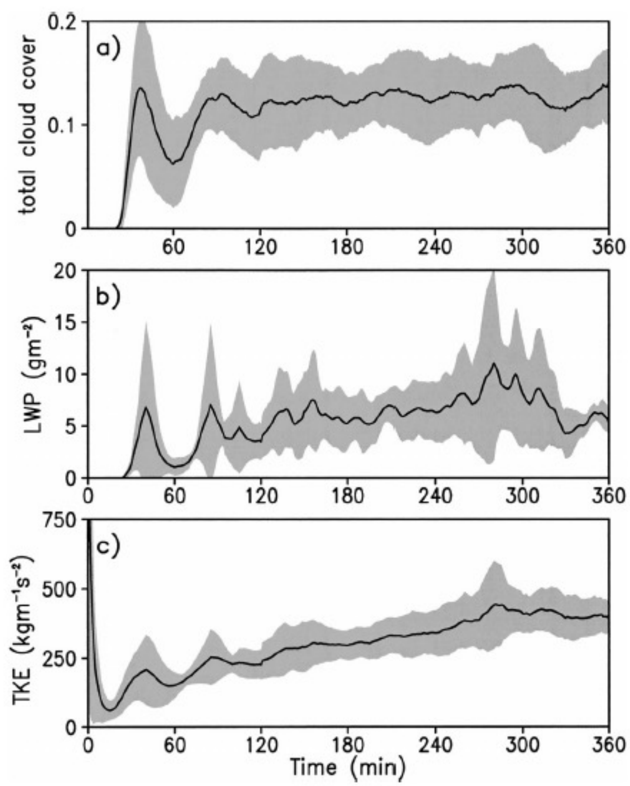
CASE1 and CASE2 CCN

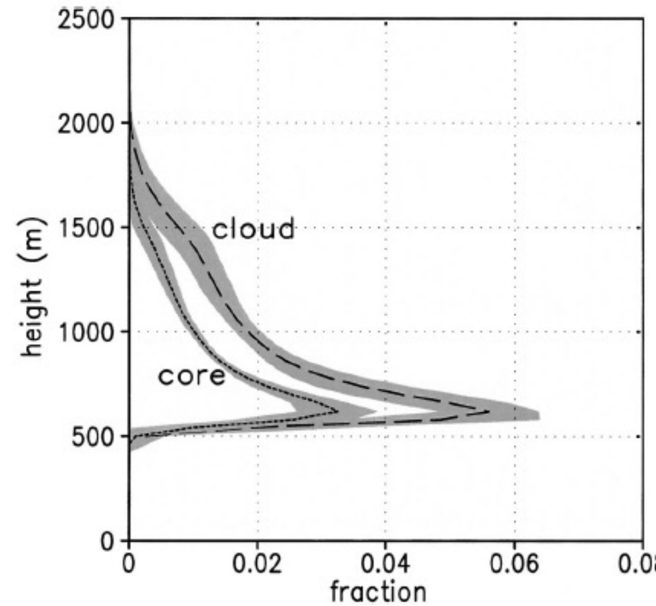
A Large Eddy Simulation Intercomparison Study of Shallow Cumulus Convection

A. PIER SIEBESMA,[a] CHRISTOPHER S. BRETHERTON,[b] ANDREW BROWN,[c] ANDREAS CHLOND,[d] JOAN CUXART,[e] PETER G. DUYNKERKE,[f*] HONGLI JIANG,[g] MARAT KHAIROUTDINOV,[h] DAVID LEWELLEN,[i] CHIN-HOH MOENG,[j] ENRIQUE SANCHEZ,[k] BJORN STEVENS,[l] AND DAVID E. STEVENS[m]

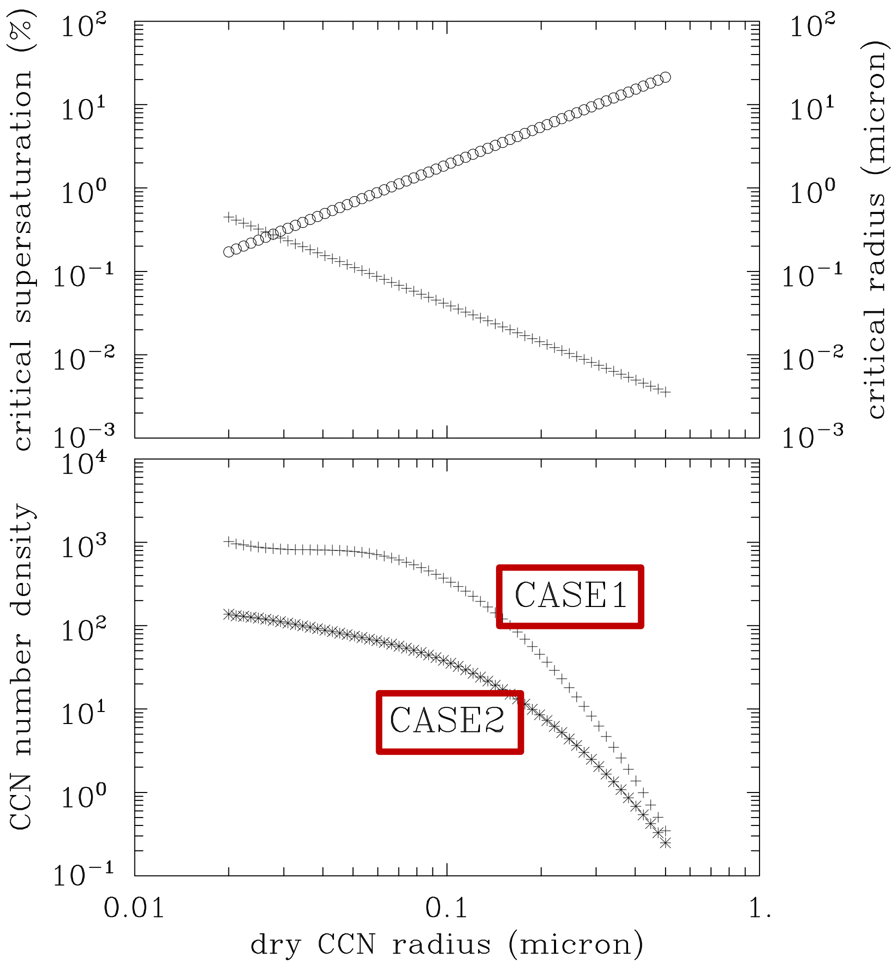
close to 800 citations…







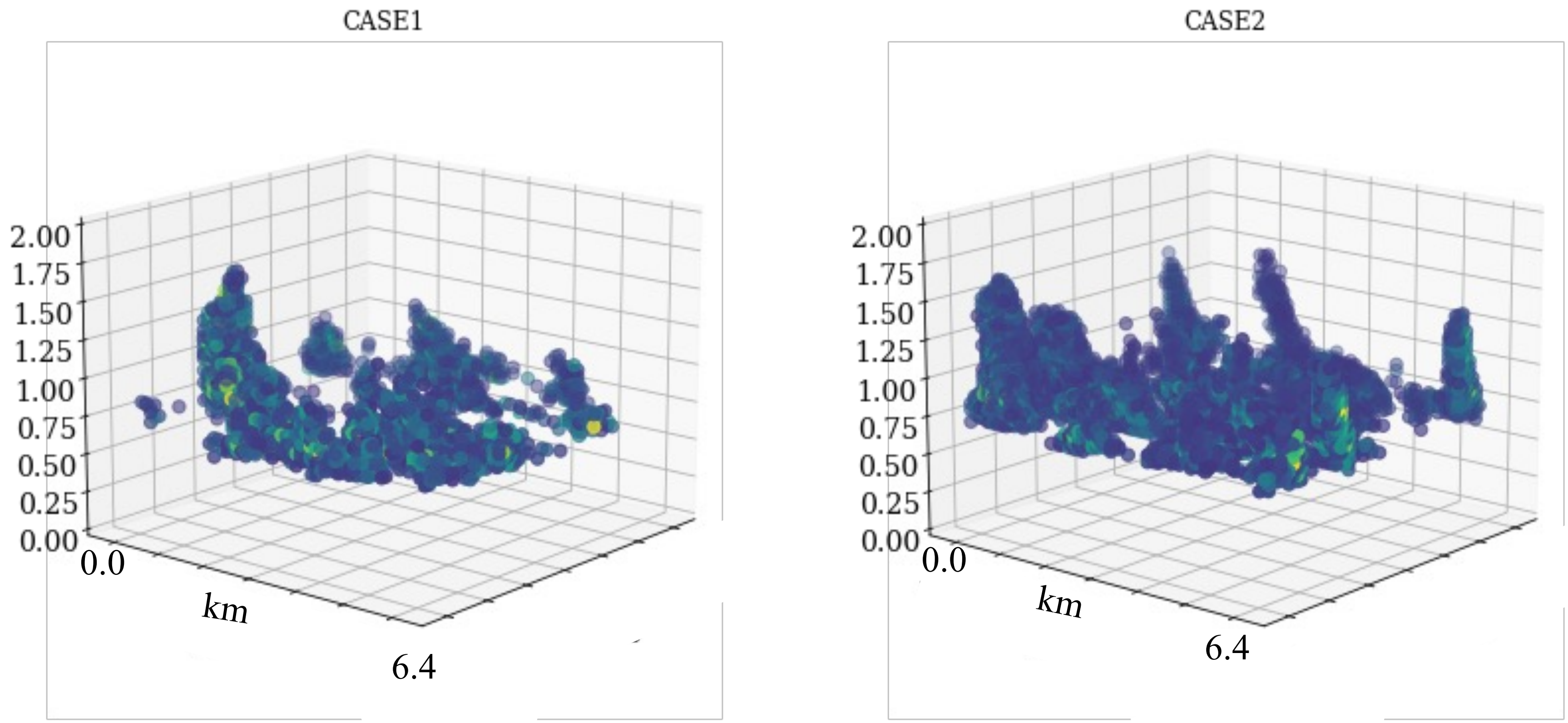
Simulations applying CCN observed over India (Gani et al. ACP 2020)



	CASE 1 Highly polluted	**CASE 2 Moderately Polluted**
Geometric Mean Radius	RM1 = 0.0125 um RM2 = 0.048 um	RM1 =0.016 um RM2 = 0.051 um
Standard Deviation	Sigma1 = 1.7 Sigma2 = 1.82	Sigma1 = 1.8 Sigma2 = 2.0
Total aerosol concentration	NANEW1 =26.5 *10^9/ m3 NANEW2 =20.5 *10^9/ m3	NANEW1 =32.0 *10^8/ m3 NANEW2 =18.35 *10^8/ m3

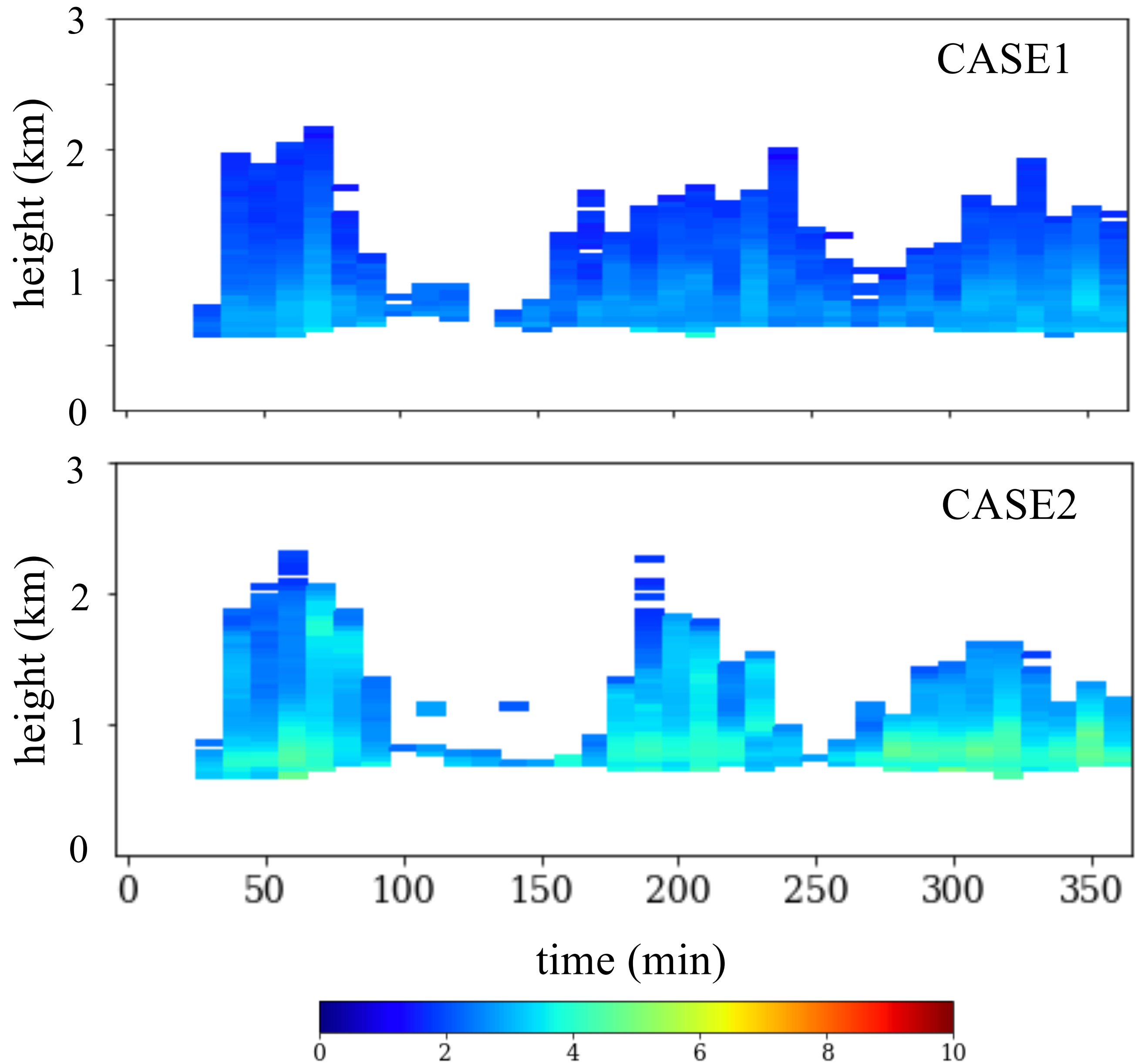
Total concentration:	47,000	5,035
Concentration (cm^{-3}) in 20 to 500 nm range:	24,500	2,870

Subin: explain the micro setup: number of SDS, range of dry CCN radius, etc…

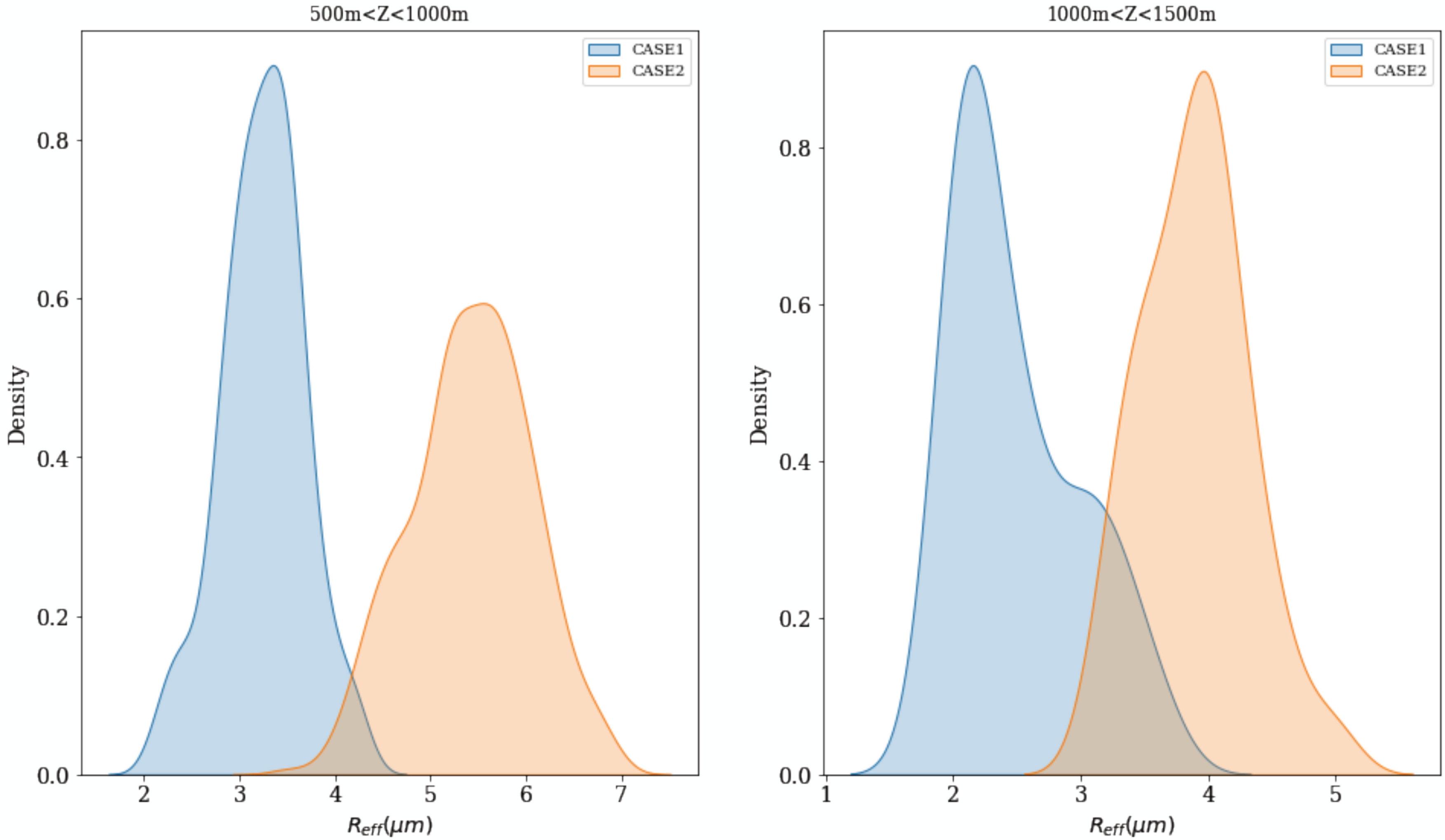


Snapshots of the cloud field in CASE and CASE2 simulations…

evolution of horizontally-averaged effective radius at each model height



Distribution of cloud effective radius at different altitudes for CASE1 & CASE 2.
Only moderately diluted clouds (0.25 <AF<0.65) are considered

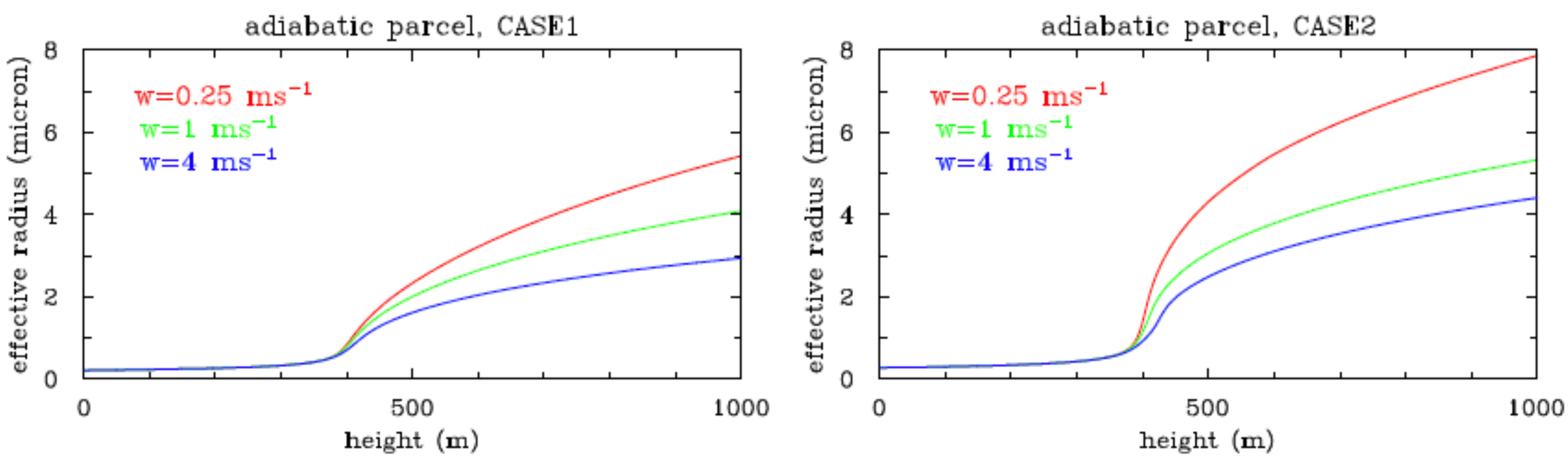


no reversal of the Twomey effect…

How to reconcile satellite observations with model simulations?

How to reconcile satellite observations with model simulations?

1. An obvious suggestion: *not all aerosols are CCN.*



If all aerosols are CCN, one expects results like in CASE 1. But if only fraction are CCN, then the outcome might be as for CASE 2, that is, larger effective radii. with aerosol distribution as in CASE1.

REMOTE SENSING LETTERS
2022, VOL. 13, NO. 10, 1002–1010
https://doi.org/10.1080/2150704X.2022.2115861

Taylor & Francis
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Type dependent role of aerosols in reversing the first indirect effect over the north Indian Ocean

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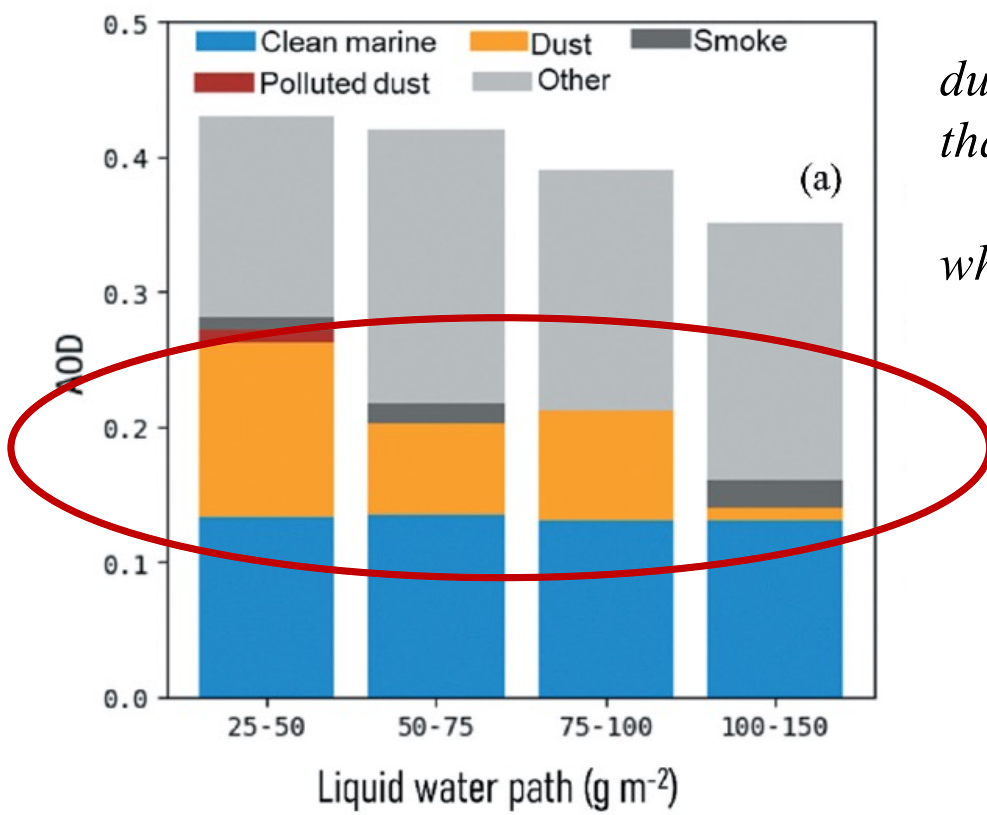


dust is likely insoluble, that is, it is not CCN.

what is "other"?

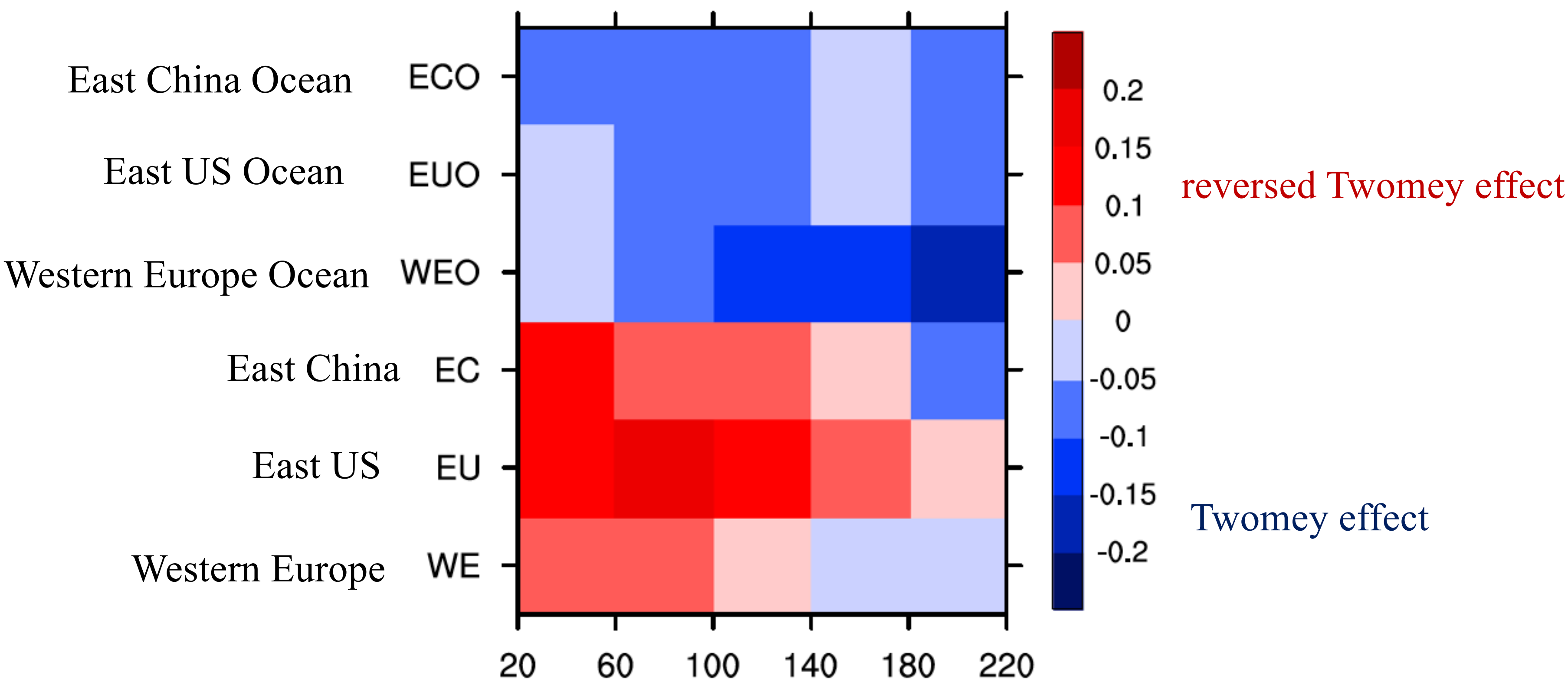
Geophysical Research Letters

RESEARCH LETTER
10.1029/2018GL077562

Key Points:
- Cloud effective radius is positively correlated to aerosol index over three industrial regions, but negatively over

Opposite Aerosol Index-Cloud Droplet Effective Radius Correlations Over Major Industrial Regions and Their Adjacent Oceans

X. Ma[1], H. Jia[1], F. Yu[2], and J. Quaas[3]



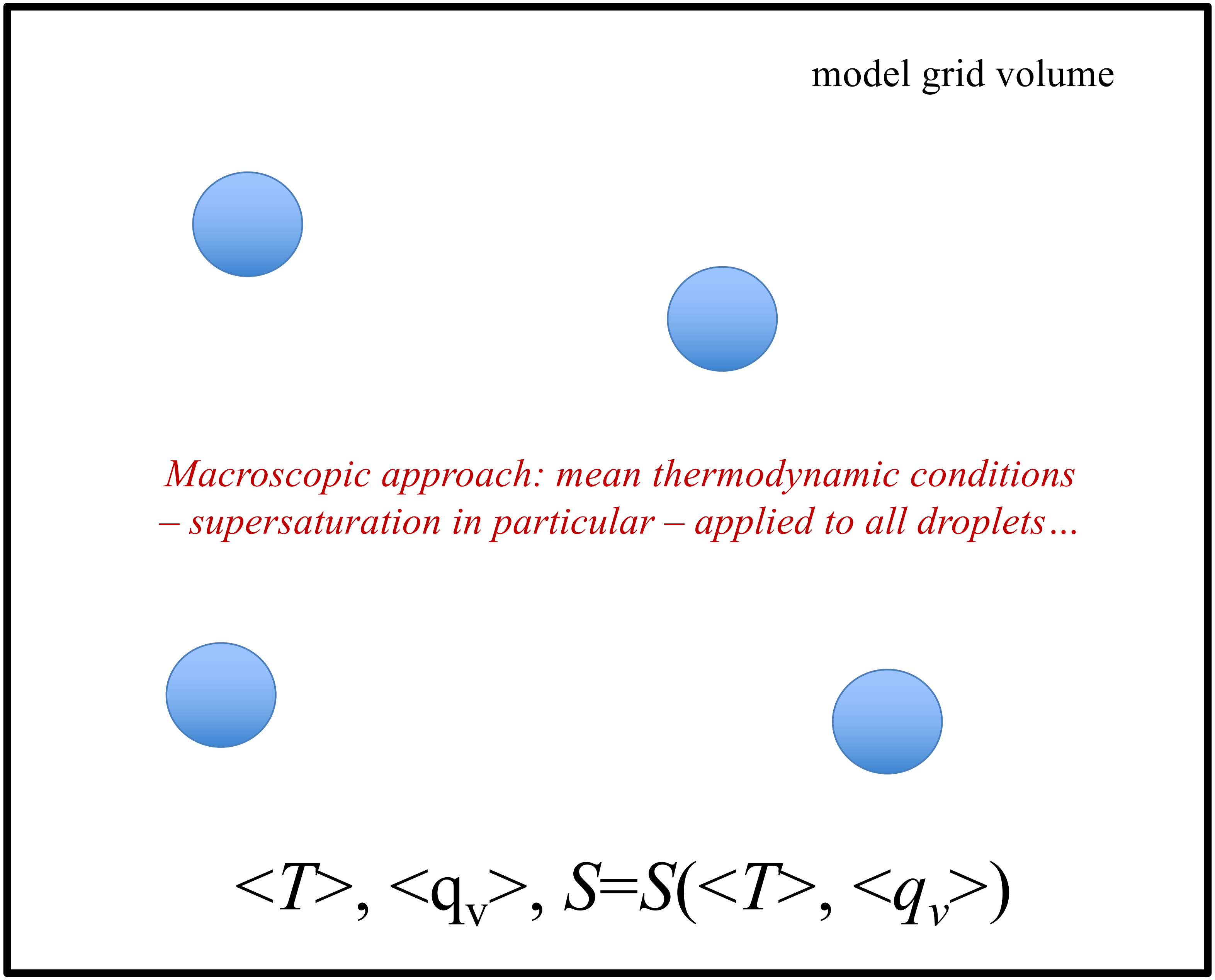
Possible explanation?

Over land: significant fraction of aerosols are insoluble, hence not CCN.

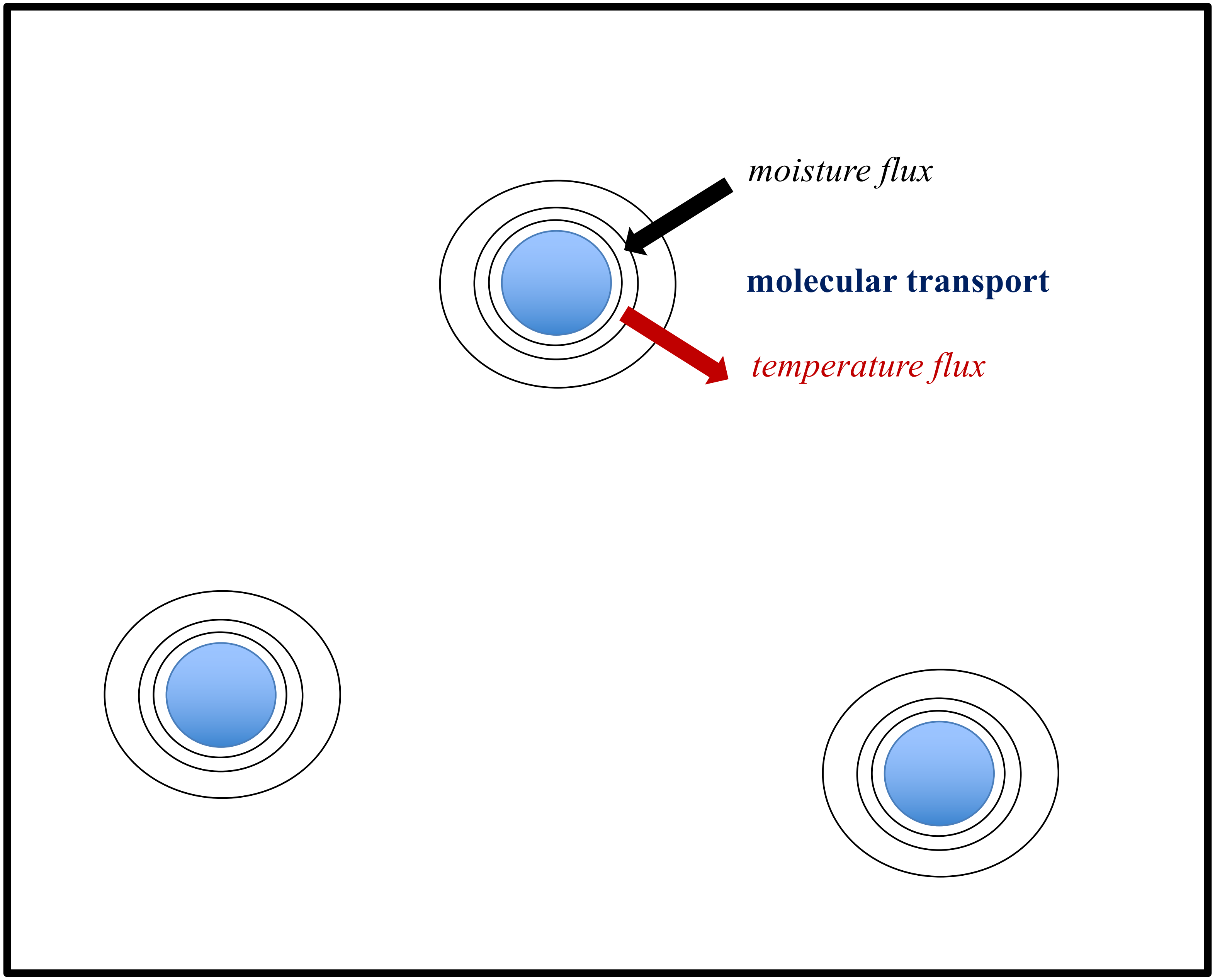
Over ocean: these are coated with sea salt and thus can act as CCN.

How to reconcile satellite observations with model simulations?

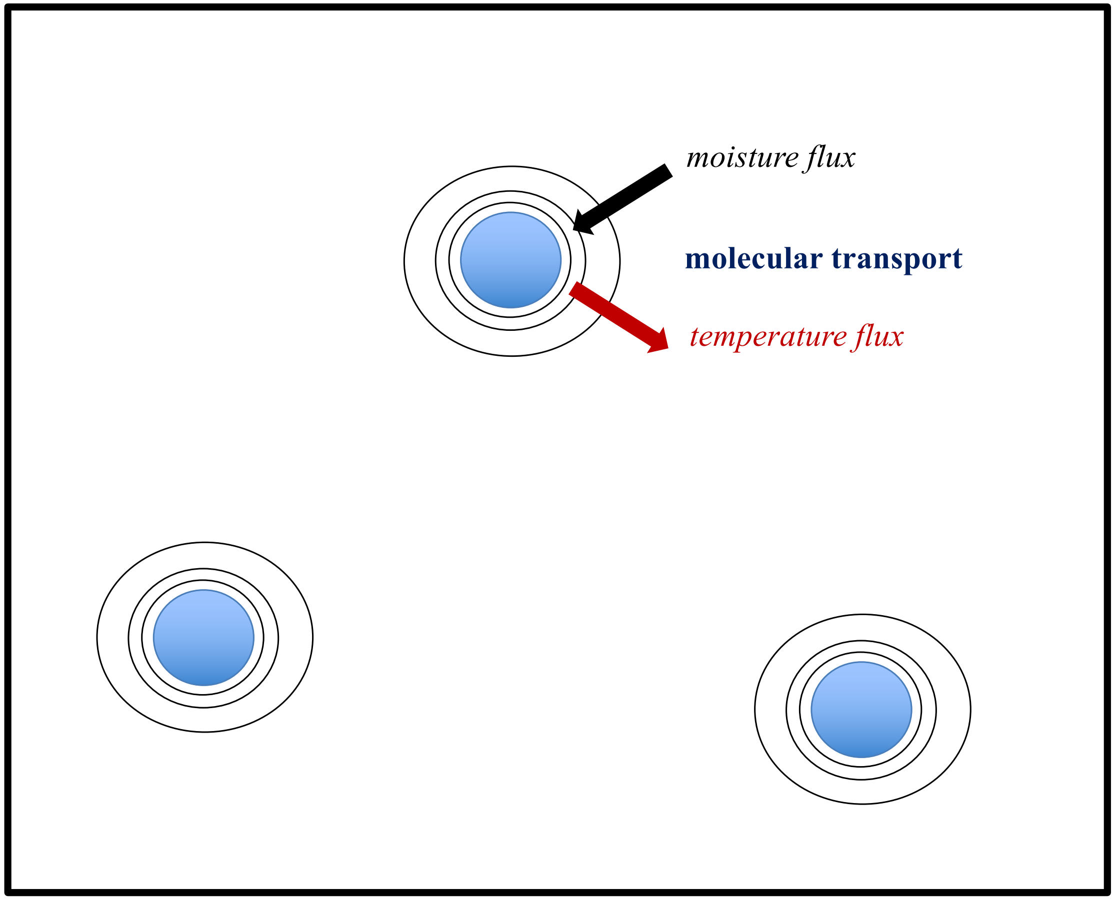
2. For droplet concentration of over 10,000 cm^{-3}, equations we solve may not be valid…



model grid volume
Macroscopic approach: mean thermodynamic conditions – supersaturation in particular – applied to all droplets…
$\langle T \rangle$, $\langle q_v \rangle$, $S = S(\langle T \rangle, \langle q_v \rangle)$



moisture flux
molecular transport
temperature flux



Molecular transport takes place over a distance of ~10 droplet radii, say, about 100 microns.

With droplet concentrations in the range of 50 to 2,000 cm^{-3}, the mean distance between droplets is between 1.7 and 0.5 mm.

But what about higher droplet concentrations, for instance, in extremely polluted environments?

JAS, 2001

Microscopic Approach to Cloud Droplet Growth by Condensation. Part I: Model Description and Results without Turbulence

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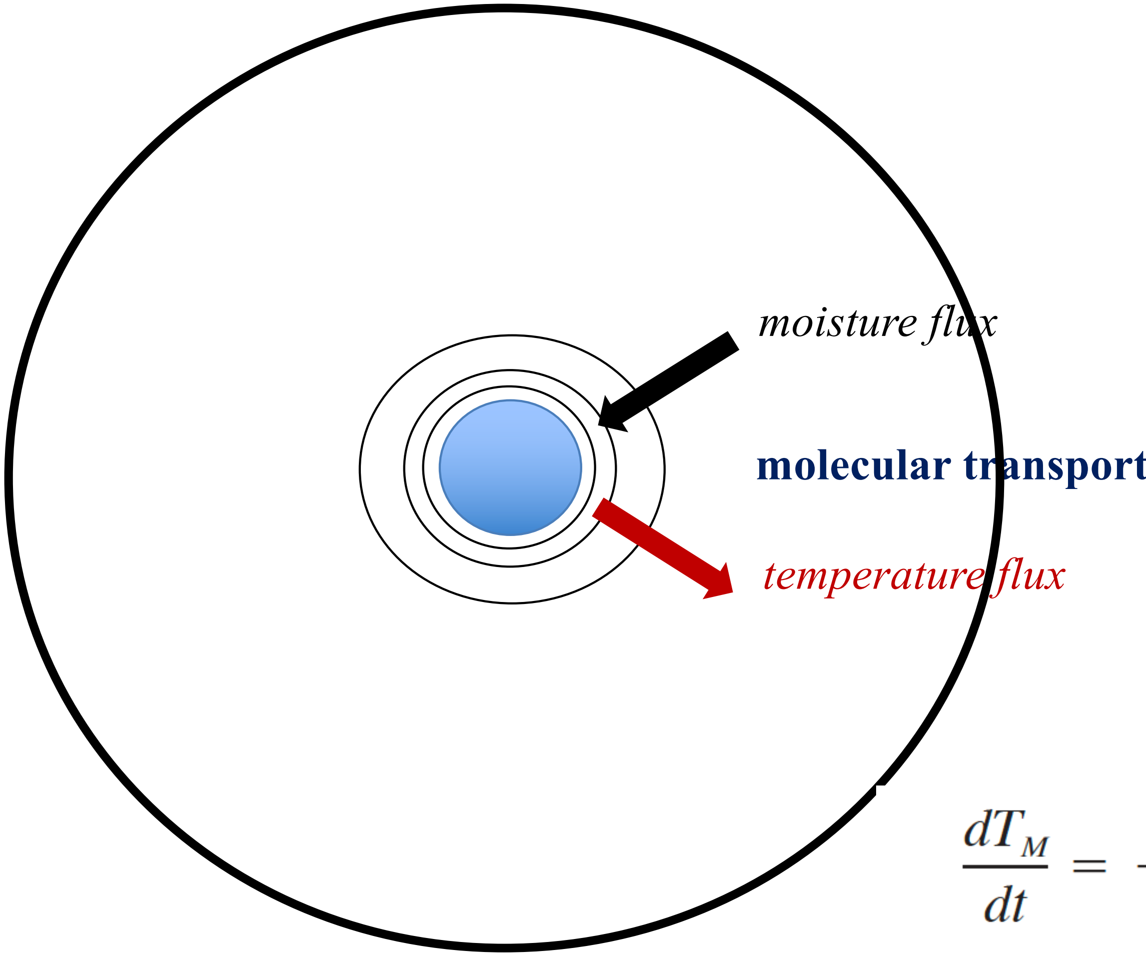
(Manuscript received 29 July 1999, in final form 31 October 2000)

Calculation of droplets growth within an adiabatic volume carrying cloud droplets and rising with a constant vertical velocity…



R_d - droplet radius

R – radius of the sphere representing mean volume occupied by a single droplet

Macroscopic equations over the volume with radius R:

$$\frac{dT_M}{dt} = -W_M\Gamma_d + \frac{L}{c_p}C_{dM},$$

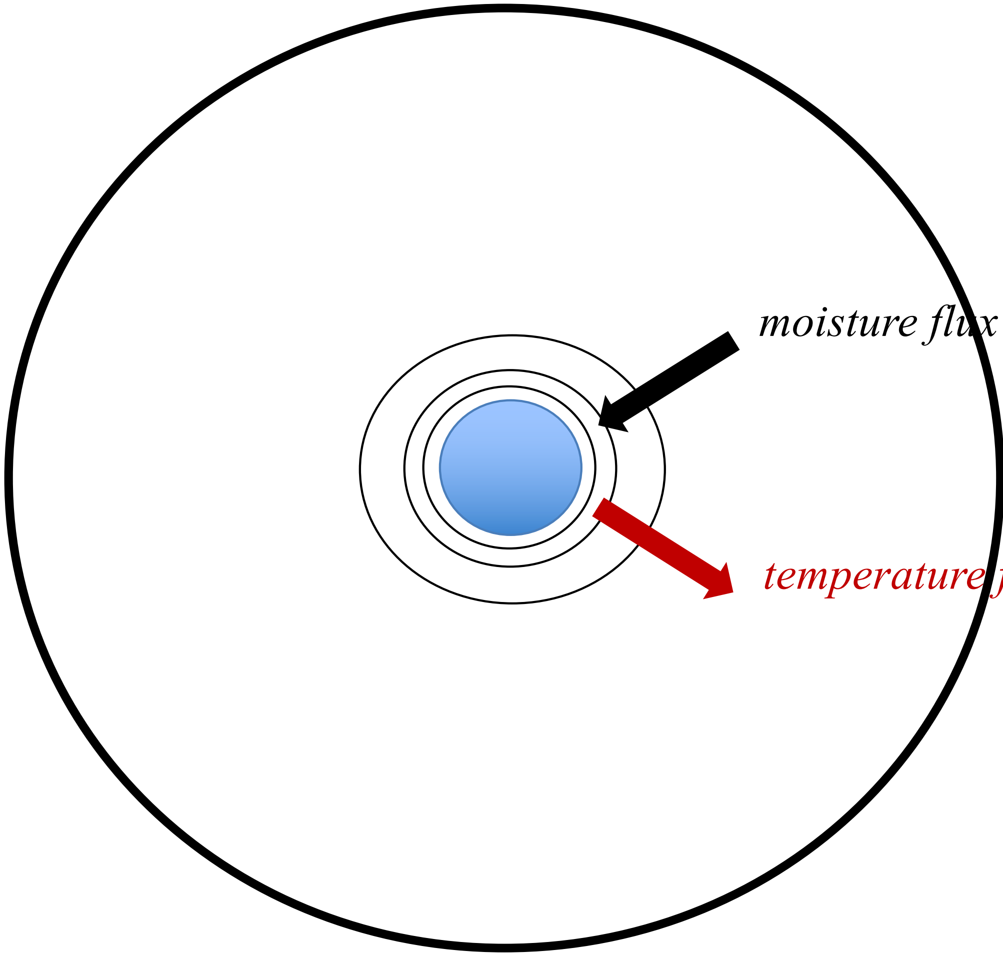
$$\frac{dq_{vM}}{dt} = -C_{dM},$$

$$\frac{dR_{iM}^2}{dt} = 2KS_M,$$

$$C_{dM} = \frac{1}{M_a}4\pi\rho_w\sum_{i=1}^{N}R_{Mi}KS_M = 4\pi\frac{\rho_w}{\rho_a}NKS_M\overline{R}_M,$$

$$\Gamma_d = g/c_p$$

$W_m = 1$ m/s



R_d - droplet radius

R – radius of the sphere representing mean volume occupied by a single droplet

Microscopic equations over the volume with radius R:

$$\frac{\partial T}{\partial t} = k_a \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial T}{\partial r}\right) - g/c_p w$$

$$\frac{\partial q}{\partial t} = D_v \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial q}{\partial r}\right)$$

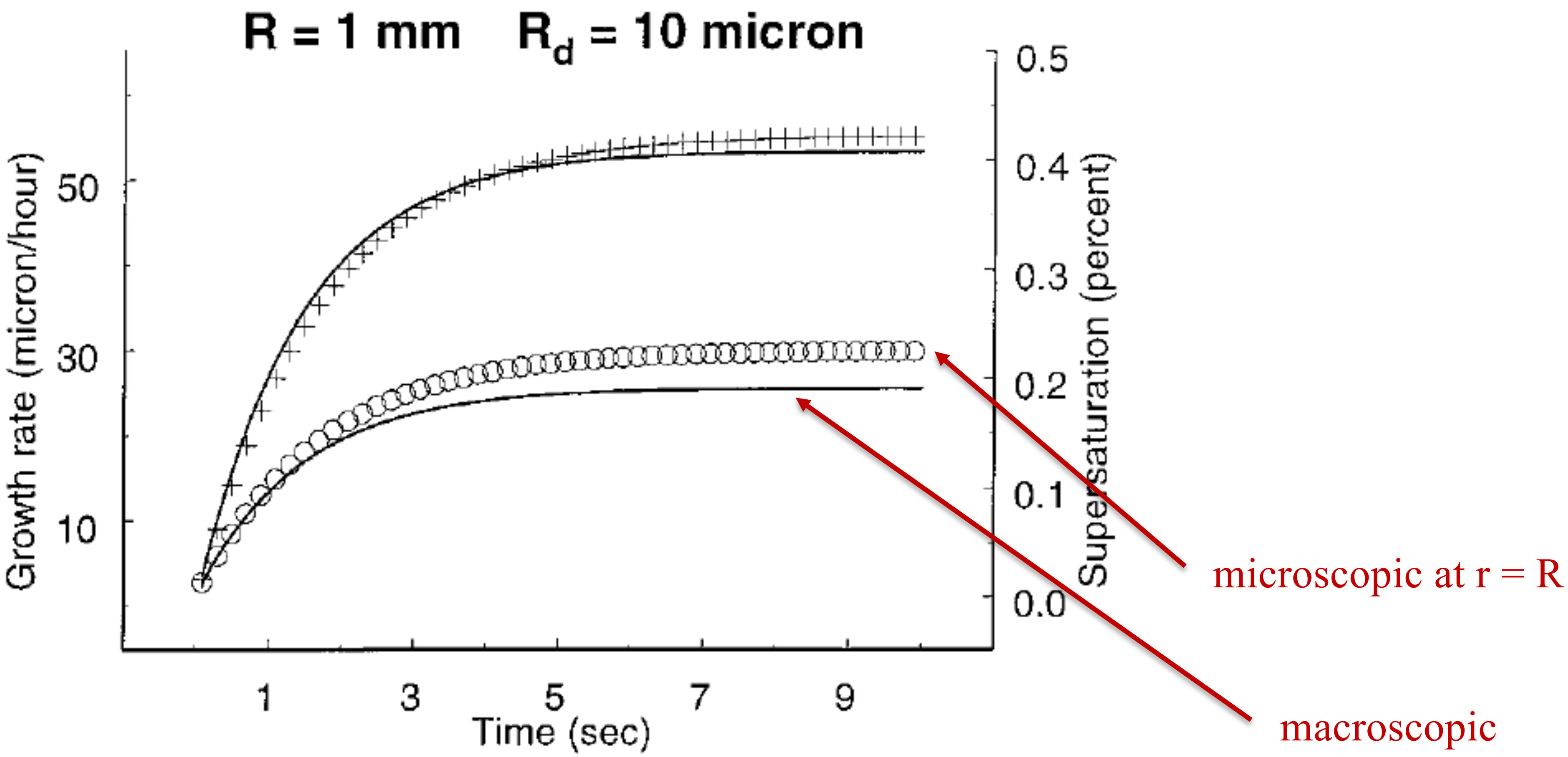
$$\frac{\partial T}{\partial r} = 0, \qquad \frac{\partial q}{\partial r} = 0 \qquad \text{for } r = R$$

$$T = T_d \qquad q = q_s(T_d, p) \qquad \text{for } r = R_d,$$

$$m_d c_w \frac{dT_d}{dt} = 4\pi L R_d^2 D_v \left(\frac{\partial q}{\partial r}\right)_{r=R_d} + 4\pi \rho_a c_p R_d^2 k_a \left(\frac{\partial T}{\partial r}\right)_{r=R_d}$$

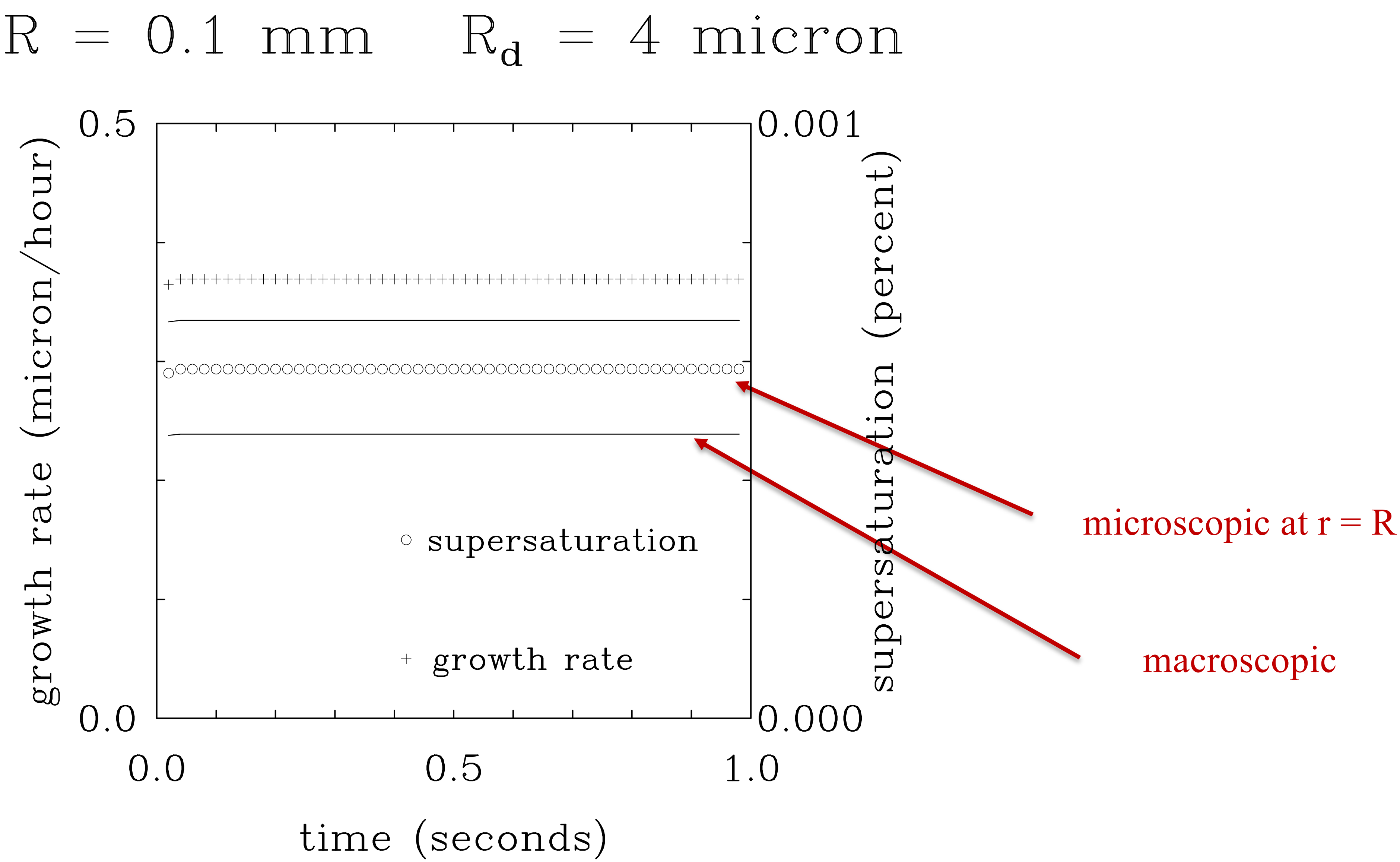
$w = 1$ m/s

concentration: ~250 cm^{-3}



Vaillancourt et al. JAS 2001

concentration: ~250,000 cm^{-3}



Larger differences, but probably not making our approach invalid…

Summary:

Microphysical explanation for the reversed Twomey effect, the competition between smaller and larger CCN/droplets during activation, does not seem to work. Although observations of the reversed Twomey effect remain uncertain, one should not reject its presence based on microphysical arguments only.

A simple explanation of the reverse Twomey effect is that not all aerosols that make larger aerosol index in polluted conditions are CCN. Detailed aerosol observations in heavily polluted environments are needed.

Other explanations have been suggested in the literature, like the "radiative pathway", that is, changing the atmospheric stability through solar radiation absorption within the polluted atmosphere and thus affecting cloud process. It is unclear how this might work as one explanation offered in the literature (e.g., Khatri et al. Nature/Scientific Reports 2022) involve complicated and uncertain logic. More modeling is needed to provide a scientifically robust explanation.

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Increased aerosols can reverse Twomey effect in water clouds through radiative pathway

Pradeep Khatri[1]✉, Tadahiro Hayasaka[1], Brent N. Holben[2], Ramesh P. Singh[3], Husi Letu[4] & Sachchida N. Tripathi[5]