Bimodal CCN spectra demonstrate aerosol cloud processing and feedback to cloud microphysics

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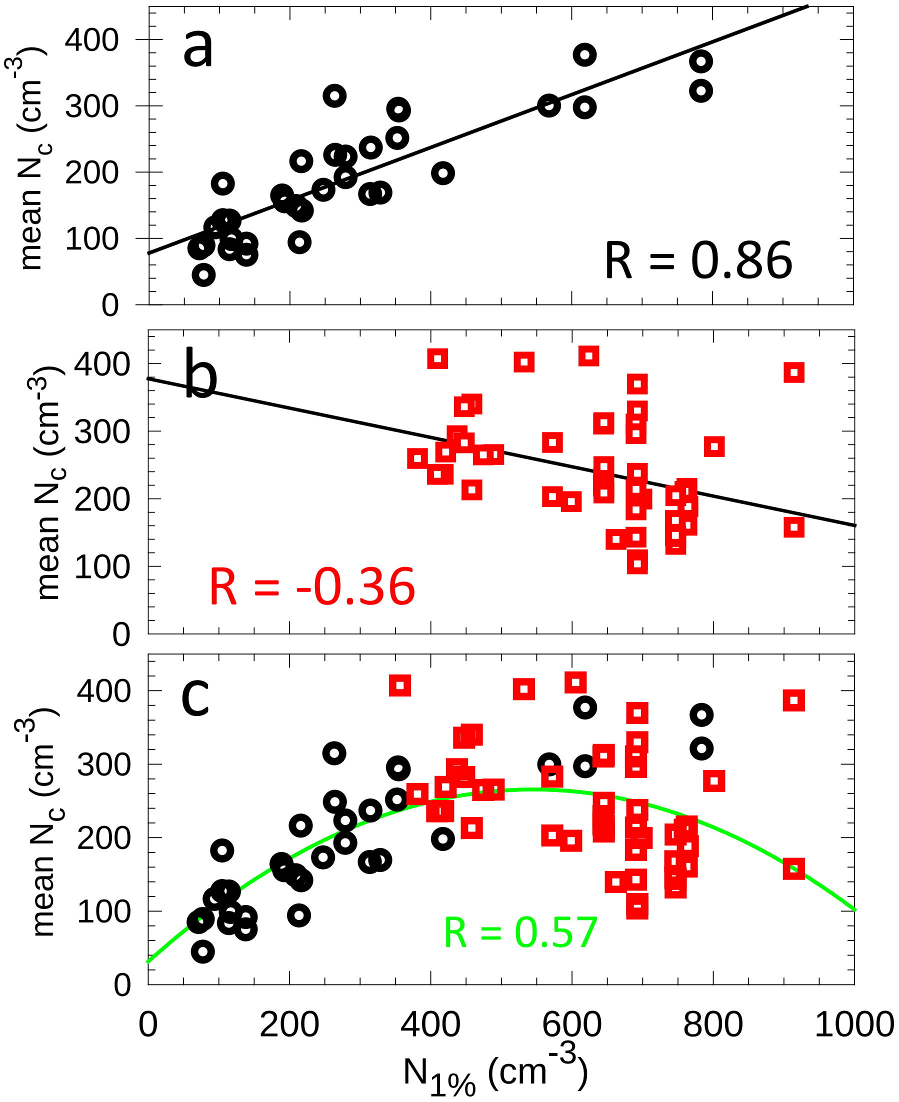


Fig. A. Mean droplet concentrations (N_c) versus below cloud CCN concentrations at 1% S ($N_{1\%}$) for 34 POST horizontal cloud passes (a) (as part of Fig. 3 of H10) and for 50 horizontal passes in MASE (b). (c) Data from panels a and b together. Linear regressions are shown in a and b, 2nd order regression is shown in c.

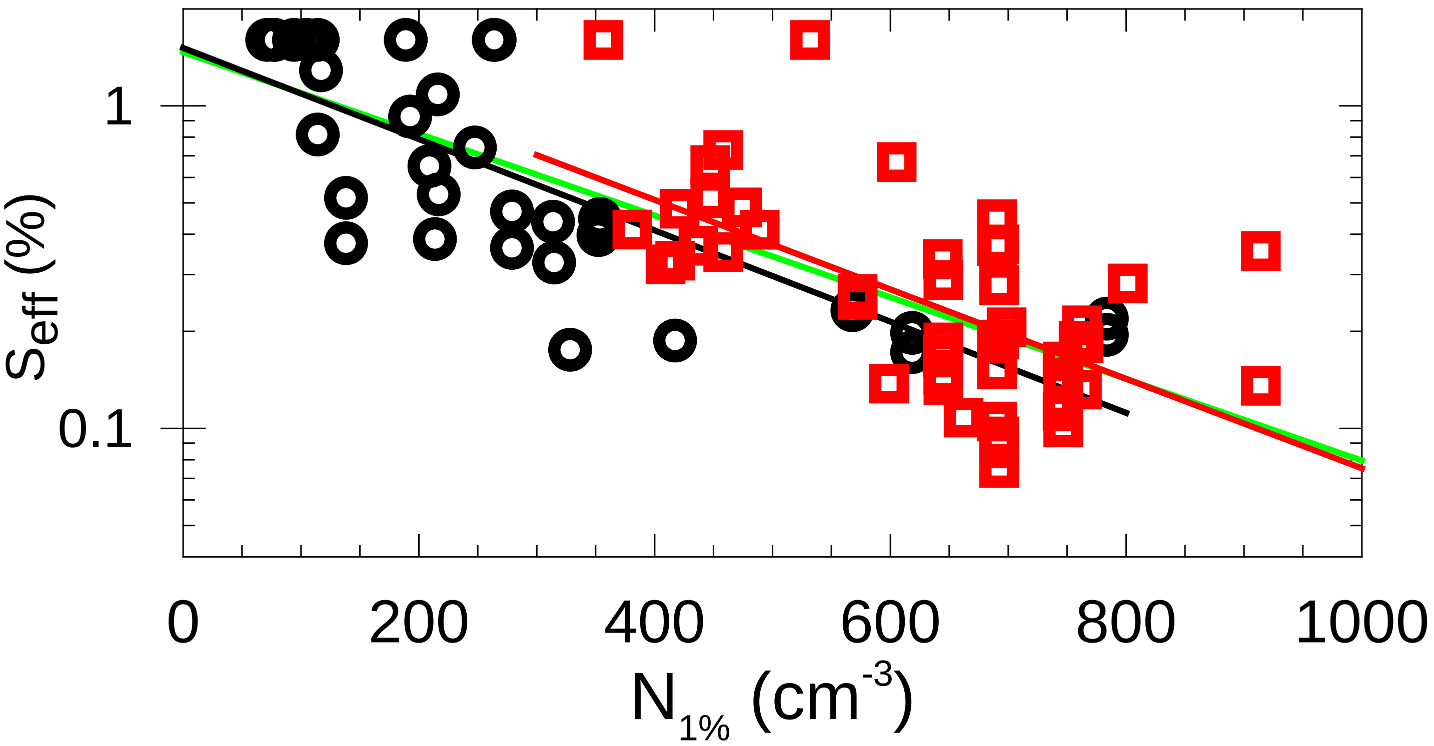


Fig. B. **E**ffective cloud supersaturation (S_{eff}) against CCN concentration at 1% S ($N_{1\%}$) black POST; red MASE. S_{eff} is the S for which nearby below cloud N_{CCN}(S) equals mean droplet concentration (N_c). Linear regression lines are shown.

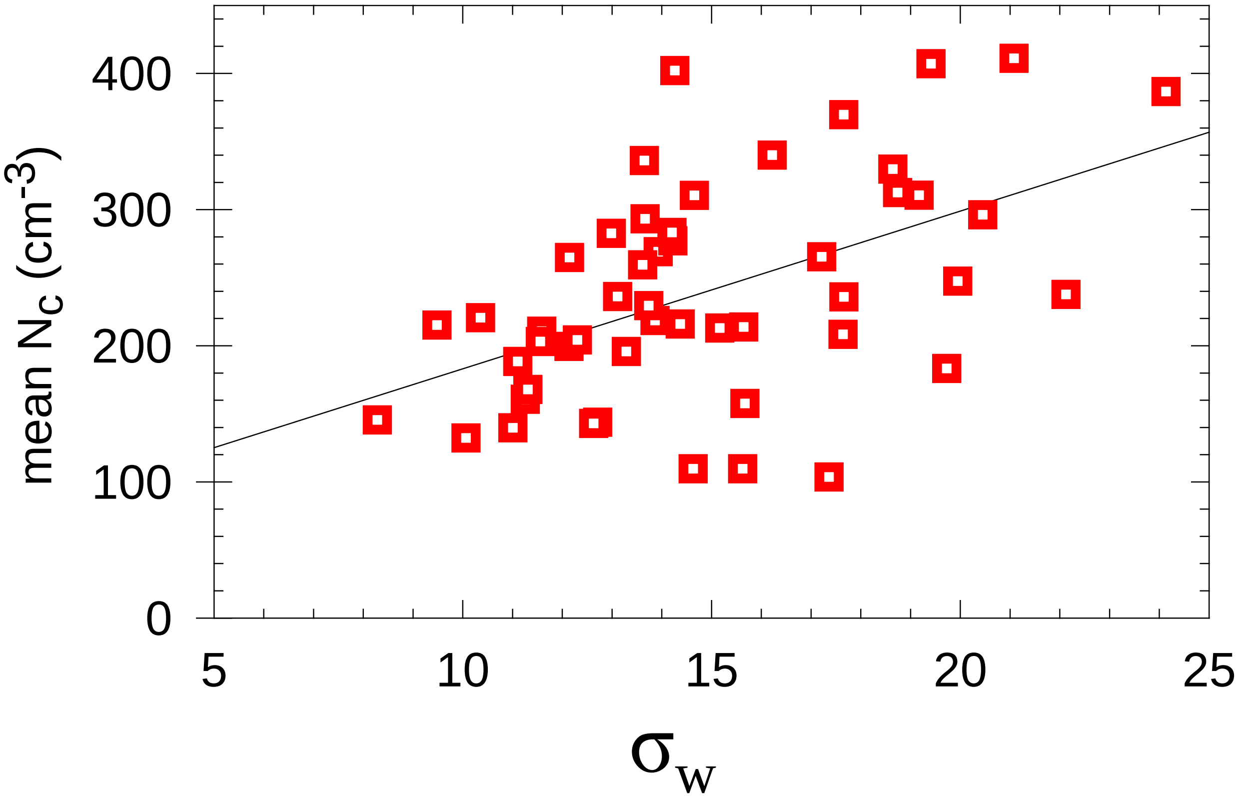


Fig. A. (a) Mean N_c versus σ_w for 50 MASE horizontal cloud passes. Linear regression is shown.

Measurements are presented from two aircraft cloud research projects:

MArine Stratus/stratocumulus Experiment
(MASE)
off the central California coast

Ice in Clouds Experiment-Tropical
(ICE-T)
cumulus clouds of the eastern Caribbean

Both in July, MASE 2005, ICE-T 2011.

Differential mobility analyzer (DMA) dry aerosol spectra below stratus clouds often display bimodality attributed to cloud processing

physical—coalescence and Brownian diffusion scavenging or chemical—reactions within droplets

that increase particle sizes and reduce

the critical supersaturation, S (S_c) of CCN

that had produced the cloud droplets.

When droplets evaporate a size gap ensues because

unactivated CCN keep their sizes and S_c whereas

activated CCN have further decreased S_c (even larger sizes).

The size at the gap between these modes has been used to infer cloud effective S, (S_{eff}) (Hoppel et al. 1986)

When all channels of the DRI CCN spectrometers are plotted, bimodality is often seen below or next to clouds.

This provides S_{eff} sans particle composition (κ).

Spectral modality is quantified on a 1-8 scale.

The most bimodal spectra with well separated equal modes are rated 1 (Fig. 1a).

Strictly monomodal spectra are rated 8 as in Fig. 1h).

Intermediate ratings for asymmetric or less separated bimodal spectra; e.g., shoulder modes (Fig. 1b-g).

Mode ratings up to 4 provided Hoppel minima, S_{eff}.

Ratings 7 and 8 did not provide S_{eff}.

Ratings 5 and 6 sometimes provided S_{eff}.

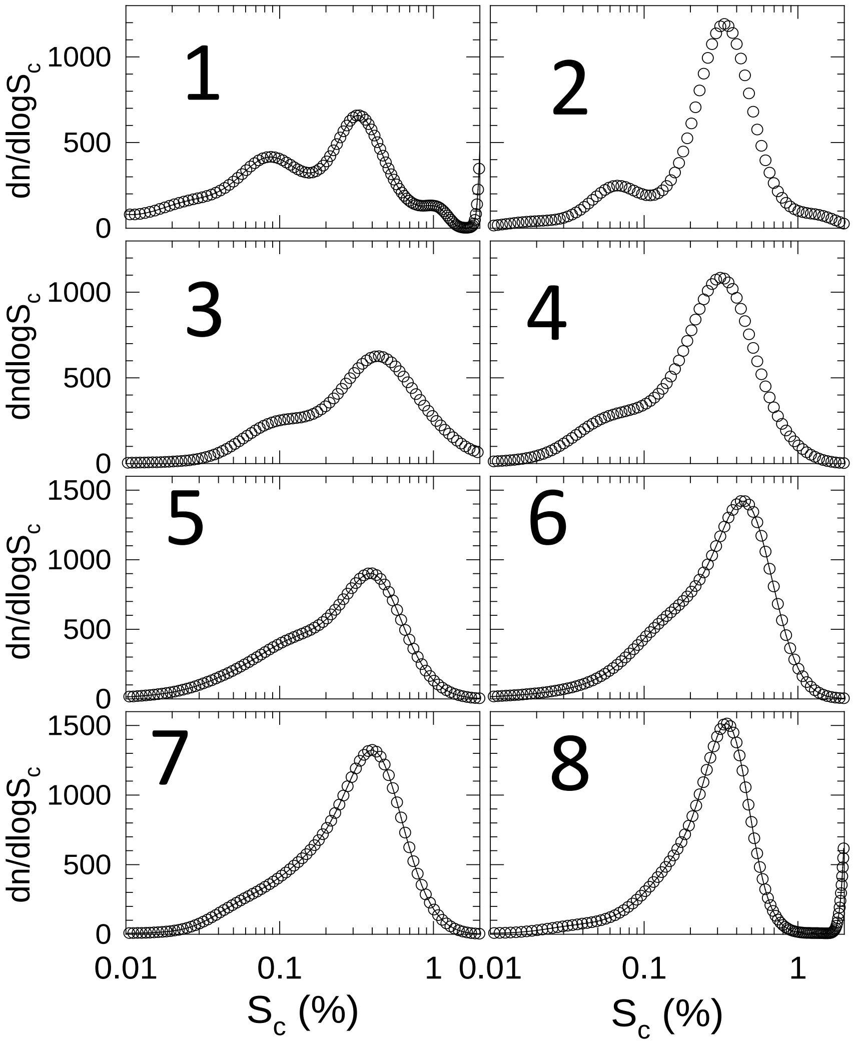


Figure 1. MASE examples of simultaneous CCN distributions for each of the 8 mode ratings.

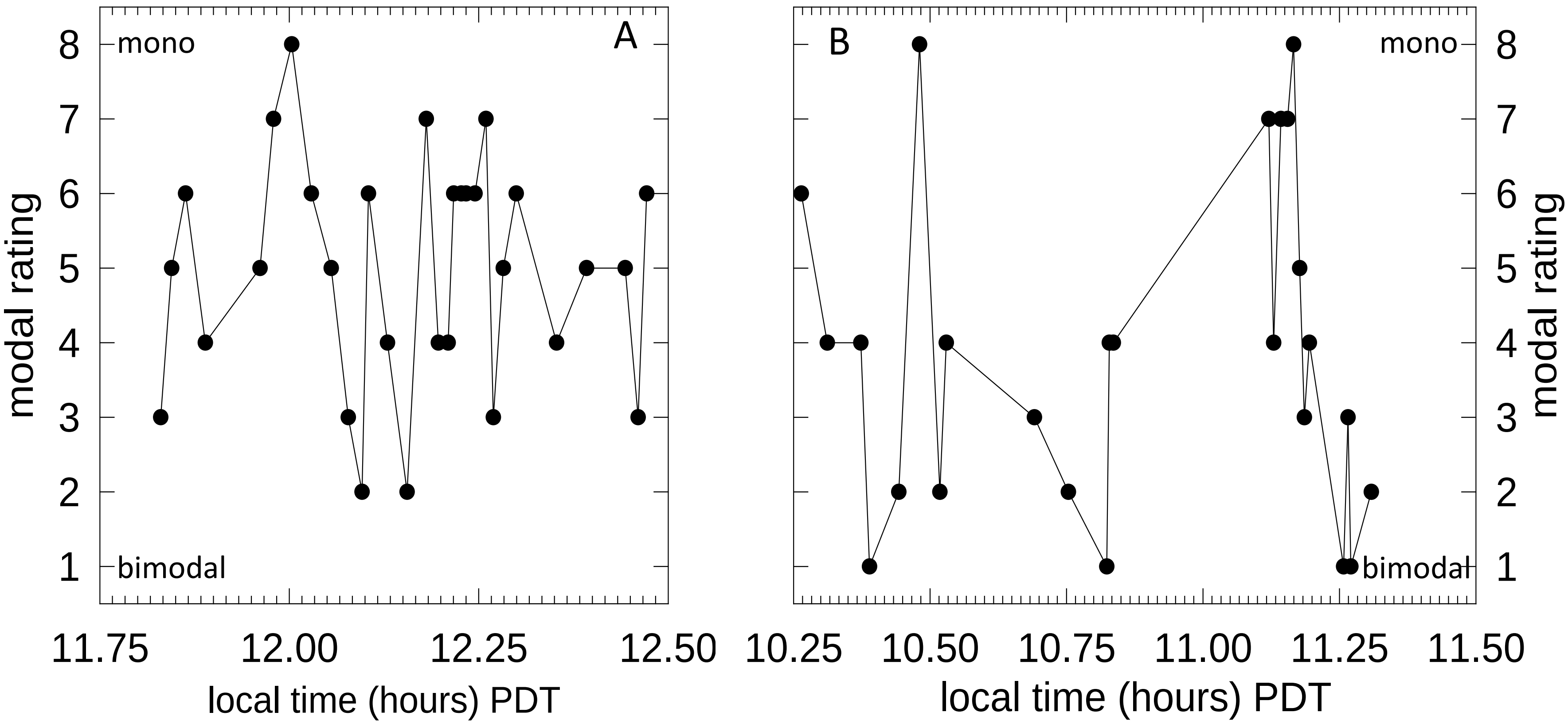


Figure 2. Time plots of CCN spectral modality under MASE stratus; (a) 18 July, (b) 23 July. Minor tick marks are minutes.

	cld	cases	mode	$S_{eff}S$(%)	$S_{eff}H$(%)
MASE	St	80	4.80	0.23	0.15
ICE-T	Cu	80	2.93	1.03	0.44

Table 1. 2nd column is the number of cases that provided Hoppel minima,

3rd column is mean modal rating,

4th column is mean traditional S_{eff} by matching below cloud CCN spectra with mean cloud droplet concentration, N_c, of the nearest cloud,

5th column is mean S_{eff} from Hoppel minima

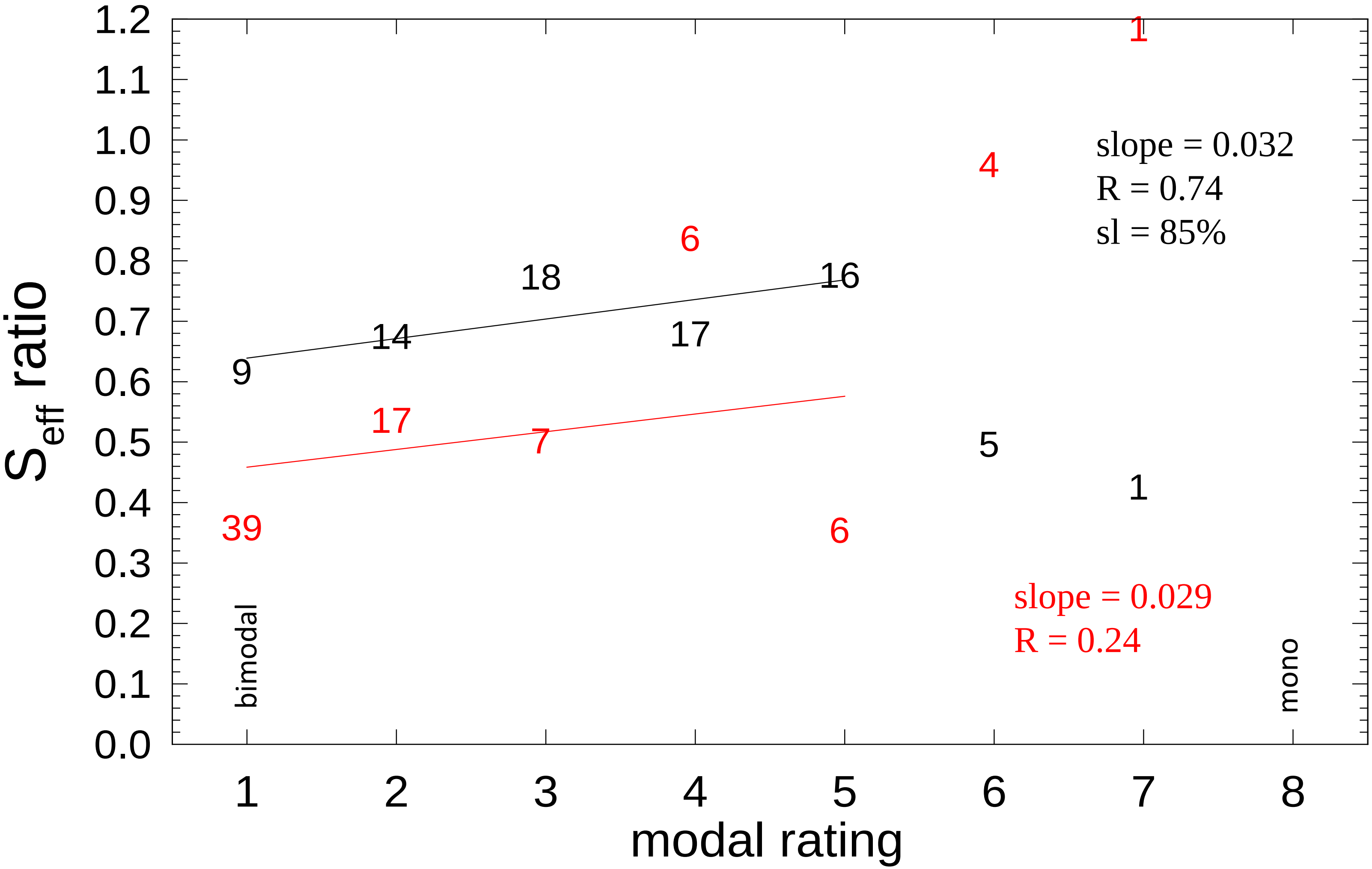


Figure 5. As Fig. 4 but ratio of S_{eff}H to S_{eff}S for MASE (black) and ICE-T (red); data from Fig. 5.

$S_{eff}H < S_{eff}S$

mainly because Hoppel minimum includes the effects of cloud processing, all 3 processes make lower S_c so Hoppel minimum is shifted toward lower S_c than the S_{eff} of the clouds.

Also smaller droplets do less processing.

But Hoppel assumed that long-lived stratus had come to equilibrium after many evaporation/condensation cycles.

Fig. 2 dispute this.

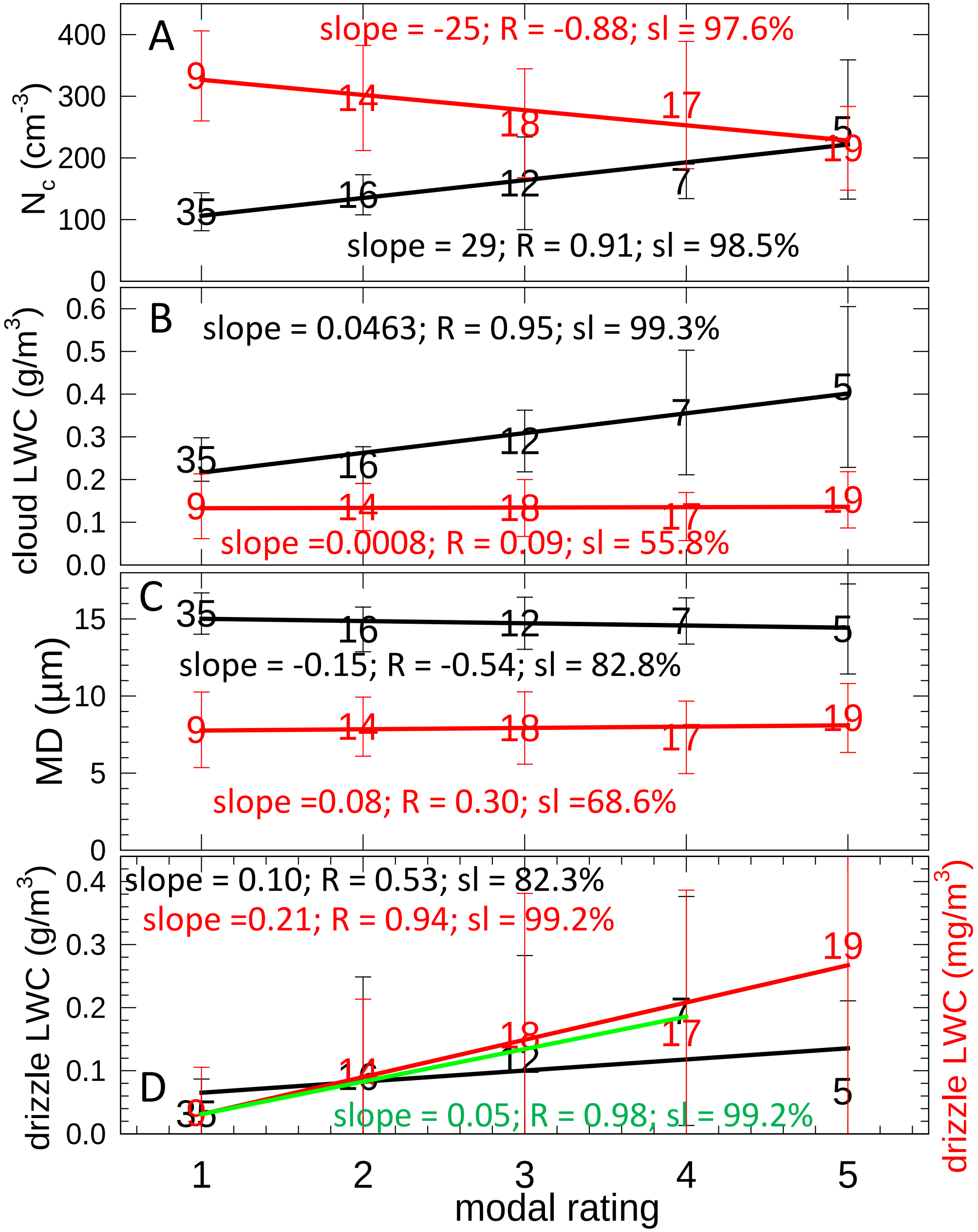


Fig. 2. Mean (displayed as number of cases) and standard deviation, sd (error bars), of cloud microphysics for categories of CCN spectra (demonstrated in Fig. 1) of nearby out-of-cloud measurements. Only modes 1-5, which are bimodal enough to provide Hoppel minima are considered. Linear regressions with slope, correlation coefficient and one-tailed significance levels are shown. Black is ICE-T, red is MASE, green is ICE-T modes 1-4. N_c is cloud droplet concentration (diameter < 50 µm), LWC is cloud droplet liquid water content, MD is cloud droplet mean diameter, drizzle is for diameter > 50 µm.

Fig. 2 A panels

Black indicates coalescence in ICE-T, which reduces droplet concentration, N_c, and CCN concentration, N_{CCN}, for more bimodal CCN (lower modal rating), see Fig. 4c black and blue. Coalescence is more likely for these cumulus clouds.

Red indicates chemical and Brownian processing in MASE, which is more likely in these polluted and stratus clouds. These processes improve CCN (lower S_c) that increase N_c and processed N_{CCN} (blue of Fig. 4a).

B panels

Black shows less cloud LWC for more bimodal CCN spectra, which is consistent with the ICE-T coalescence reducing N_c probably largely by conversion to larger drizzle sizes out of the cloud size range.

Red is weak because of the opposite effects noted above for MASE, which conflict with effects similarly noted for ICE-T.

C panels

Black and **Red** show no relationships with modal rating; MD constant with mode. This could be due to conflicting effects.

D panels

Black and **Red** display strong positive relationships, indicating that more bimodal CCN spectra are associated with clouds that have less drizzle. This seems to indicate that in both projects drizzle has fallen out of the clouds that have produced bimodal CCN spectra.

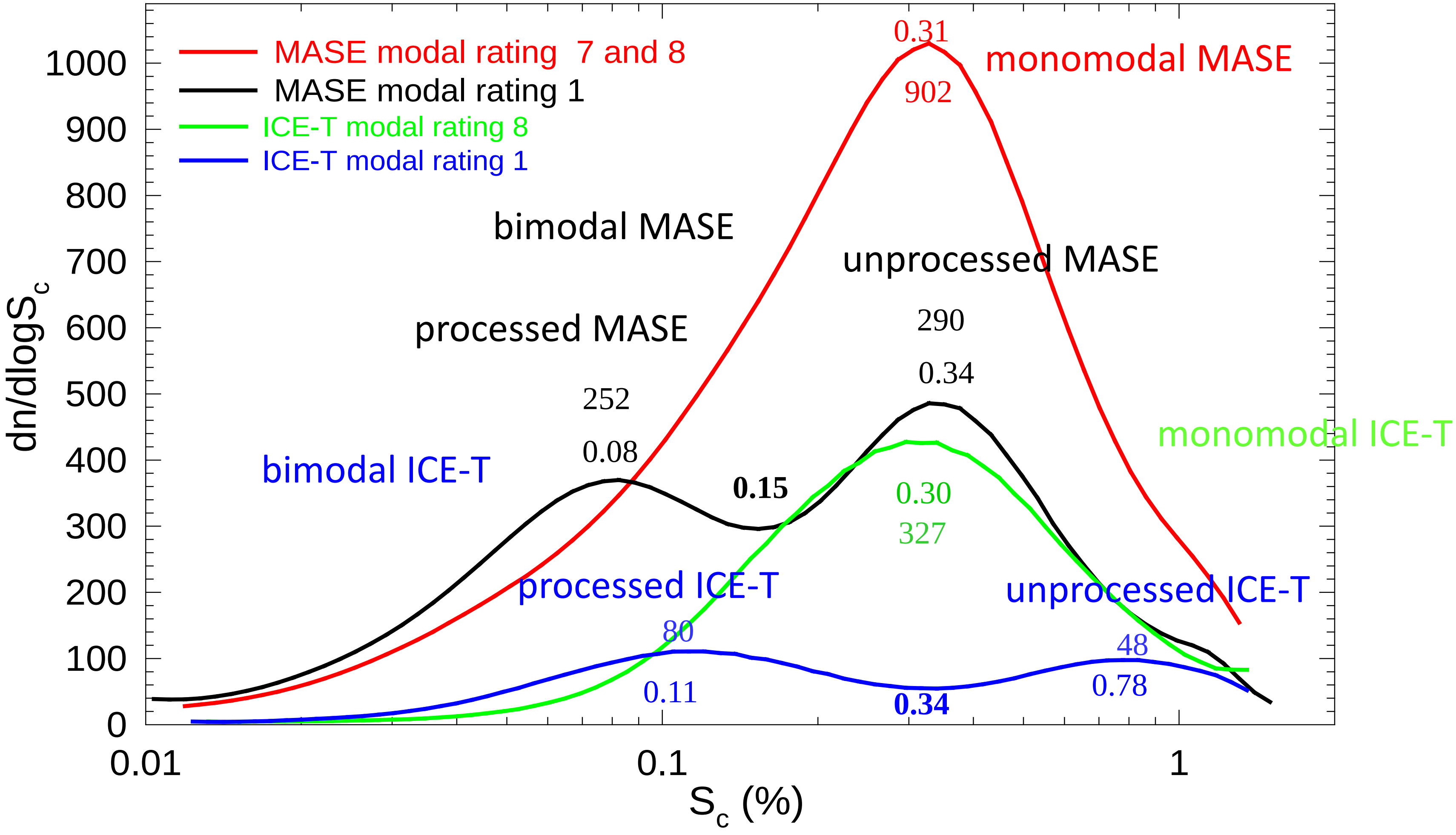
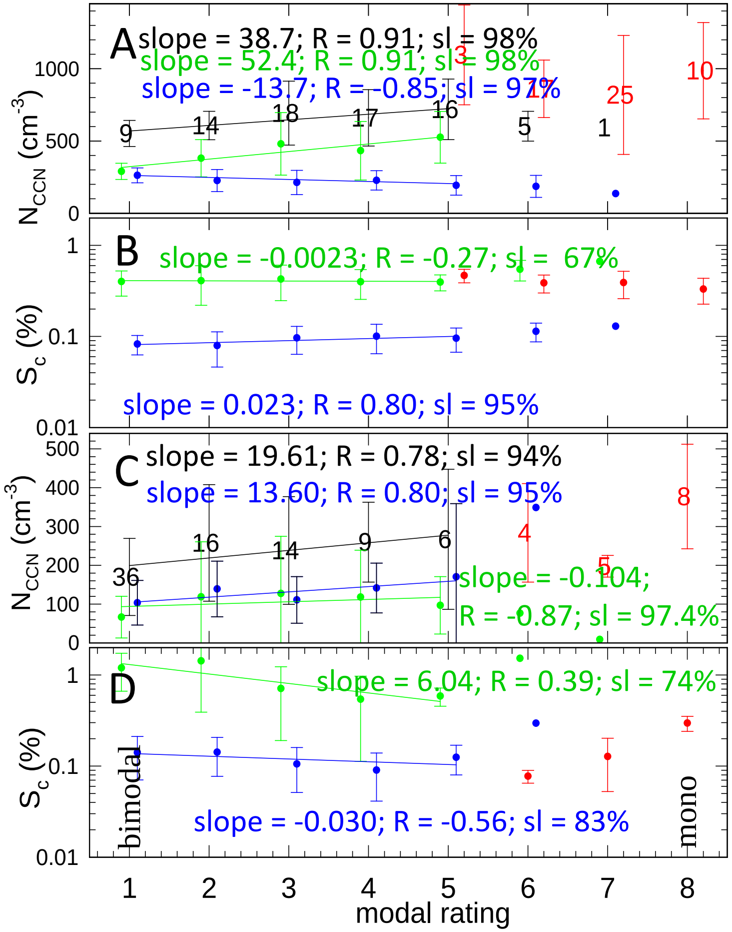


Figure 3. As Figs. 1 but composites of the MASE below monomodal (rating 7 and 8) and ICE-T monomodal (rating 8) and most bimodal (rating 1) spectra of both projects. Modes S_p, S_u and S_m, $S_{eff}H$ (%) (bold) and mean of $N_{CCN}M$, $N_{CCN}U$ and $N_{CCN}P$ are shown (cm^{-3}).

total = unprocessed + processed monomodal

MASE stratus clouds

ICE-T cumulus clouds



N_{tot} < $N_{monomodal}$ and N_{tot} decreases with bimodality for both projects implies physical processing

MASE:

N_{unp} decreases with bimodality Implies Brownian capture of highest S_c, which are most mobile

N_{proc} increases with bimodality implies chemistry and Brownian

S_{cunp} = S_{cmono}

ICE-T:

S_{cunp} > S_{cmono} > S_{cproc}

S_{cunp} increases with bimodality implies coalescence removing the lower S_{cunp}; i.e., largest droplets in that mode

Figure 4. Total concentration data points are plotted as numbers of cases.

Fig. 4. Panels (a) and (c) display mean and sd of CCN concentrations, N_{CCN}, within modal categories against modal categories. Panels (b) and (d) display mean and sd of mode S_c of the three modes. Green is the unprocessed mode of the bimodal spectra. Blue is the processed mode of the bimodal spectra. Black is the sum of processed and unprocessed modes. Red is for monomodal spectra. Panels (a) and (b) are for MASE. Panels (c) and (d) are for ICE-T. In panels (a) and (c) mean values for total bimodal spectra (black) and monomodal spectra (red) are plotted as numbers of the quantity of cases. Some data are mode rating staggered for clarity. Regressions are for modes 1-5 only. Sl are one-tailed significance levels. From Hudson et al. (2015).

Some summary thoughts:

Aerosol bimodality can be detected in DRI high-resolution CCN spectra.

Bimodality is **not** universal even under solid stratus; monomodal and bimodal spectra are often intermingled ; not in equilibrium.

Bimodal aerosol/CCN spectra are common under stratus and cumuli (next to also).

Physical, mainly coalescence processing in the cumuli of ICE-T and chemical and Brownian diffusion processing in the polluted stratus of MASE.

Chemical and Brownian diffusion cloud processing could enhance the indirect aerosol effect (IAE) by making better (lower S_c CCN), though this might destabilize the clouds and reduce IAE.

Brownian capture reduces high S_c (small) CCN from air pollution so less IAE.

Although coalescence also makes better CCN it also reduces N_{CCN}.

Now can examine high resolution CCN spectra of more than 40 previous aircraft and surface measurement projects.

The extent and effect of cloud processing on CCN and cloud microphysics requires as much attention as CCN sources.

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On 3/5/2015 8:57 AM, Jim Hudson wrote:
Tony,

I said that none of the Hoppel or Clarke papers have been disputed concerning cloud processing as the cause of bimodality.

Is this true?

Have you ever been challenged about cloud processing?

Are you aware of alternate explanations for bimodal aerosol spectra?

Are you aware of observations of bimodal aerosol spectra that are not associated with or caused by cloud processing?

Thanks, Jim

Hi Jim;
Regarding your questions---

Is this true?

As far as I know.

Have you ever been challenged about cloud processing?

No.

Are you aware of alternate explanations for bimodal aerosol spectra?

No

Are you aware of observations of bimodal aerosol spectra that are not associated with or caused by cloud processing?

Mixing of two distinctly different sources or air masses probably results in this at times but I do not think it is the typical reason.

I think I have identified this occasionally but it is not the norm.

I assumed everyone accepted this cloud processing these days.

I would challenge the reviewers to suggest a different mechanism that is more consistent with the data in the regions you are reporting on.

Tony