Cloud Processing of Aerosol

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Measurements from two aircraft field projects;

Marine Stratus/Stratocumulus Experiment (MASE)
Ice in Clouds Experiment-Tropical (ICE-T)

polluted stratus off the central California coast in July 2005
small low altitude cumuli of the eastern Caribbean in July 2011

DOE Gulfstream 1 airplane
NCAR C-130 airplane

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DRI high resolution CCN spectrometers have revealed bimodal spectra caused by within cloud processes (Hoppel et al. 1985) by chemical or physical transformations (coalescence or Brownian capture of small interstitial material by cloud droplets).

These enlarge (decrease S_c) CCN dissolved within activated cloud droplets while unactivated particles that did not produce cloud droplets remain the same size and S_c.

After evaporation of cloud water, which happens to most clouds, there is a size separation gap (**Hoppel minimum**) that defines a bimodal spectrum from a precloud unimodal aerosol.

The two modes are processed (accumulation) and unprocessed (Aitken) (H15).

Hudson, J.G., S. Noble, and S. Tabor, 2015: Cloud supersaturations from CCN spectra Hoppel minima. *J. Geophys. Res., Atmos.*, *120*, Issue 8, 27 April , 3436–3452, doi:10.1002/2014JD022669.

Variability of CCN bimodality was quantified on a 1-8 scale.

Unqualified bimodal spectra designated 1.

Strictly unimodal spectra designated 8.

Intermediate designations for spectra with unequal or un-separated modes;

i.e., one mode a shoulder of another.

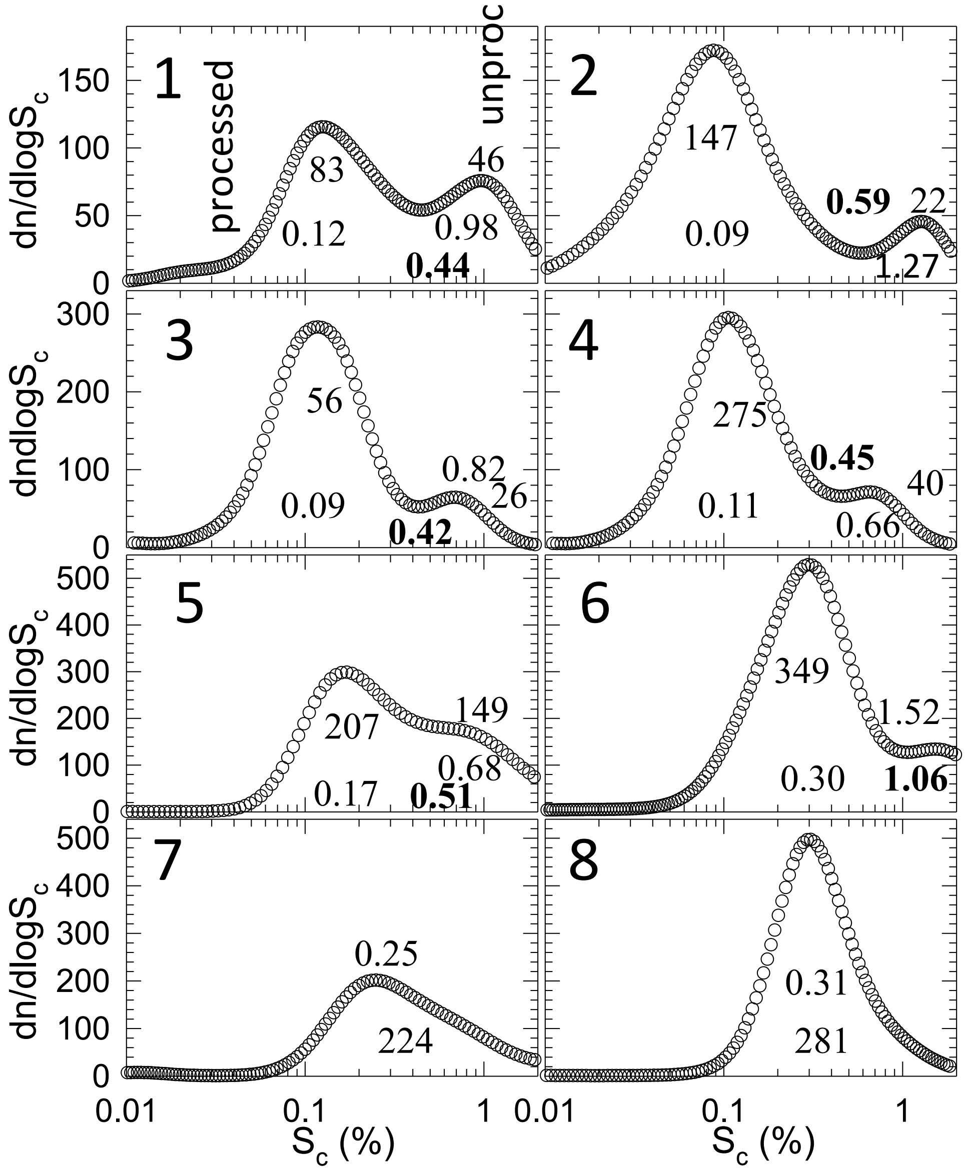


Fig. 1. Differential CCN concentrations (per cm^3) against critical supersaturation (S_c) for ICE-T below cloud examples of the 8 modal ratings. S in percent for Hoppel minima (**bold**), the modes of the processed distributions (lower S_c) and the unprocessed distributions (higher S_c) are quantified in 1-5. The unimodal modes (S_m) are quantified in 6-8. N_{CCN}P (N_p), N_{CCN}U (N_u) and N_{CCN}M (N_m) in number per cm^3 are also displayed. Revision of Fig. 3 of Hudson et al. (2015).

Subjective categorization; but we also have 2 objective categorizations:
$N_u - N_p$; high numbers unimodal, low or negative numbers bimodal
Classified $N_u - N_p$; sort $N_u - N_p$ into 8 groups of equal cases; lowest group 1 unimodal,
highest group 8 bimodal; reduces extremes

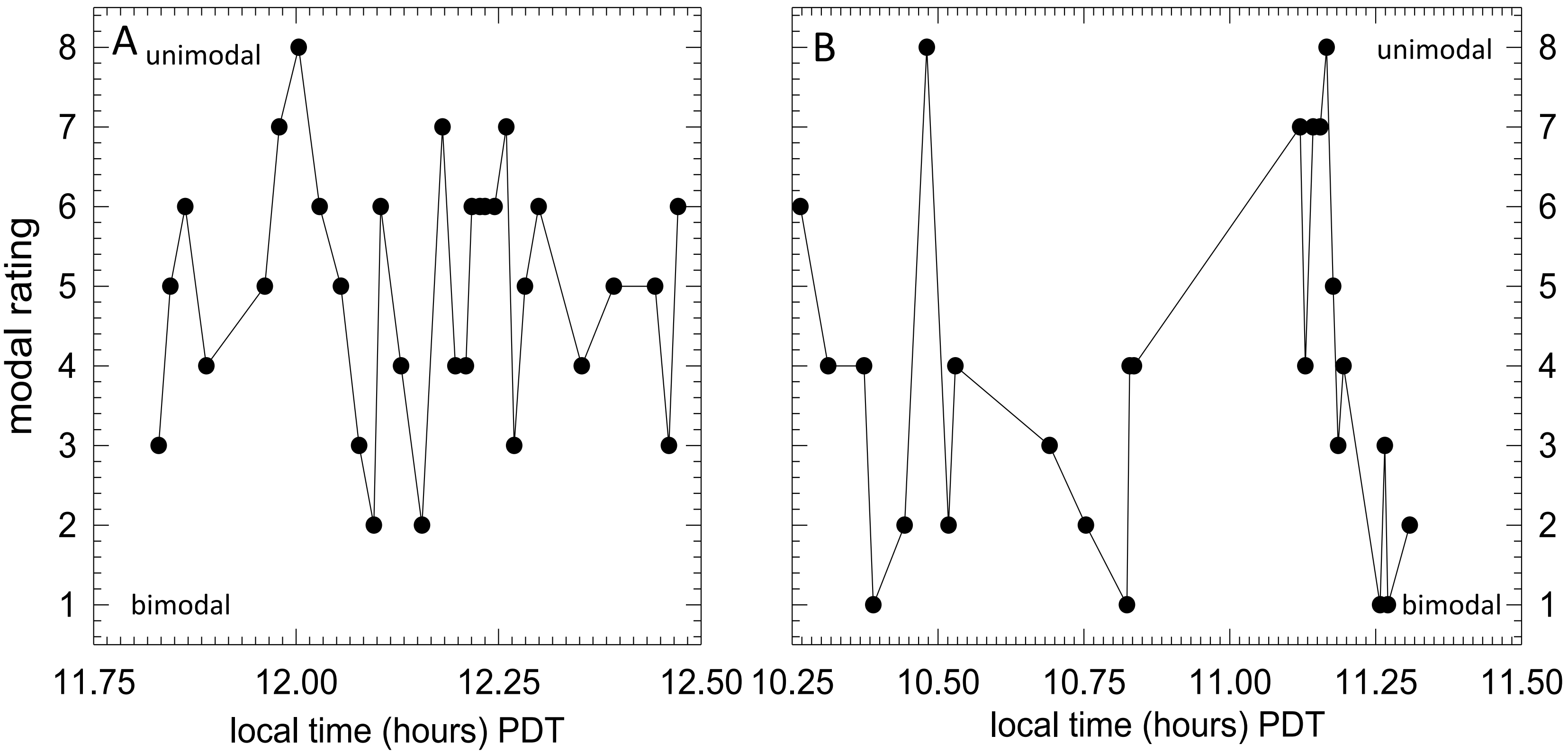


Fig. 2. Time plots of CCN spectral modality under MASE stratus; (a) 18 July, (b) 23 July. Minor tick marks are minutes. From Hudson et al. (2015)

Aerosol/CCN modality varies over short time intervals.

Compared CCN spectral modality with microphysics of the closest clouds

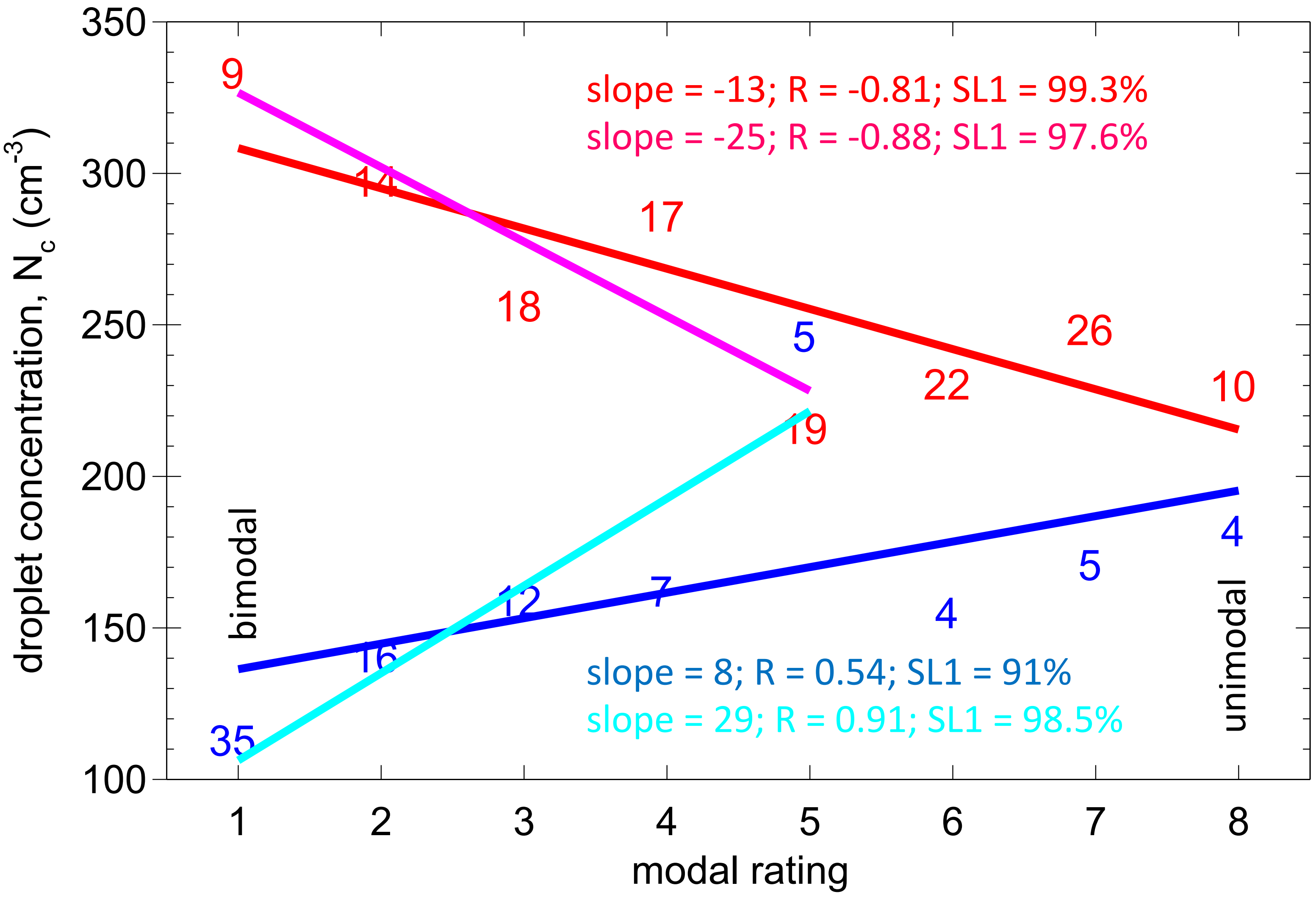


Fig. 3. Mean cloud droplet concentrations, N_c, against closest CCN modal rating for MASE (red and pink; polluted stratus) and ICE-T (blue and cyan; Caribbean cumuli). Correlation coefficients (R) and one-tailed significance levels (SL1) are shown. Pink and cyan regressions consider only modes 1-5 that have Hoppel minima. Data points are plotted as numerals showing the numbers of cases. From Hudson et al. (2015).

More chemical processing by gas-to-particle conversion within droplets (especially sulfate but also nitrate) and Brownian capture in MASE polluted stratus.
Lower cloud supersaturations (S) because of lower vertical wind, W, and higher N_{CCN} leave many interstitial particles that can be Brownian captured by cloud droplets.

More coalescence processing in thicker higher LWC ICE-T cumuli, which have larger droplets.
Higher W and S leave few interstitial CCN.

Hudson et al. (2015)

MASE

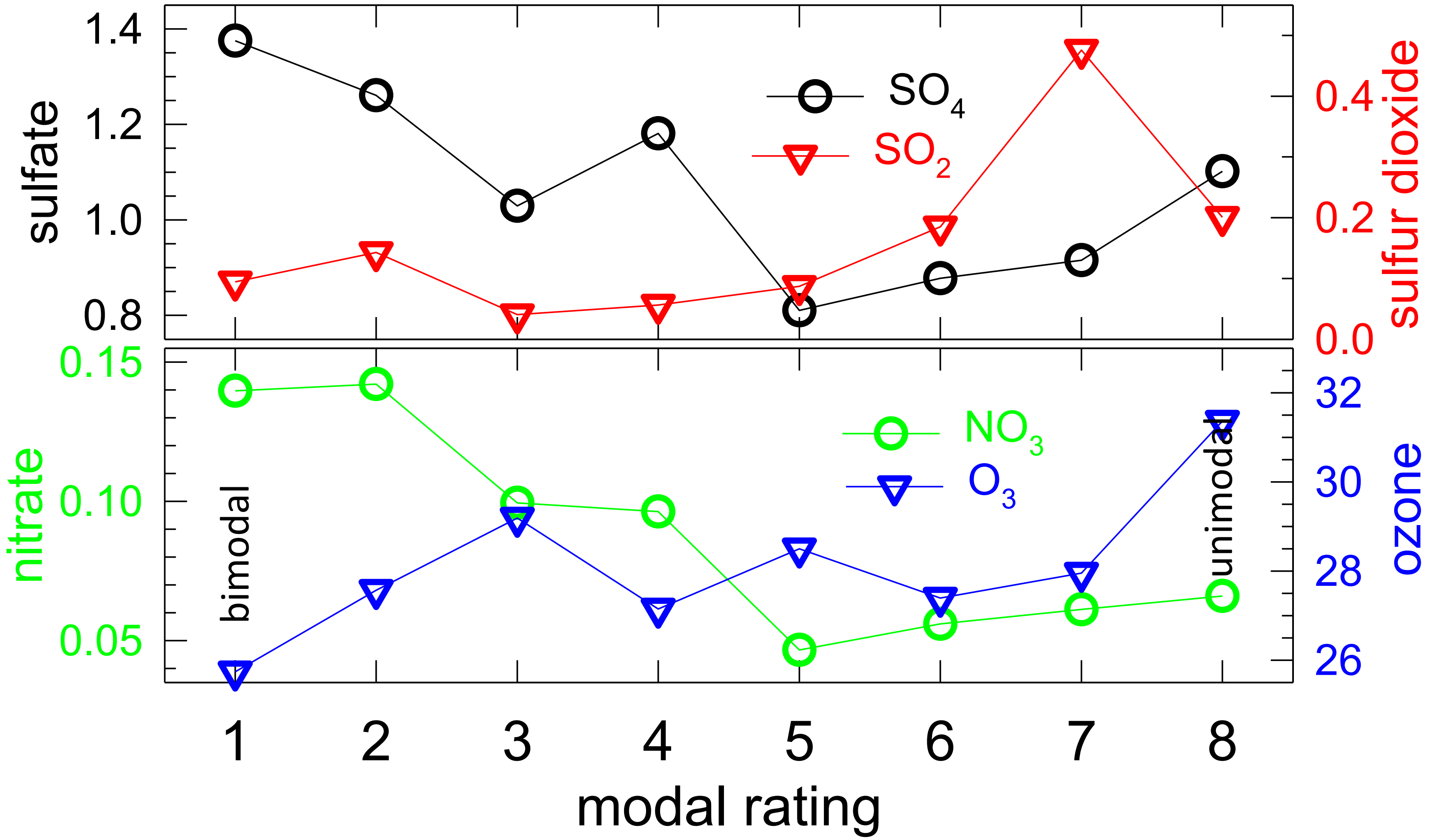


Fig. 4. Sulfur dioxide conversion to sulfate; nitrogen oxides to nitrate; ozone aides reactions

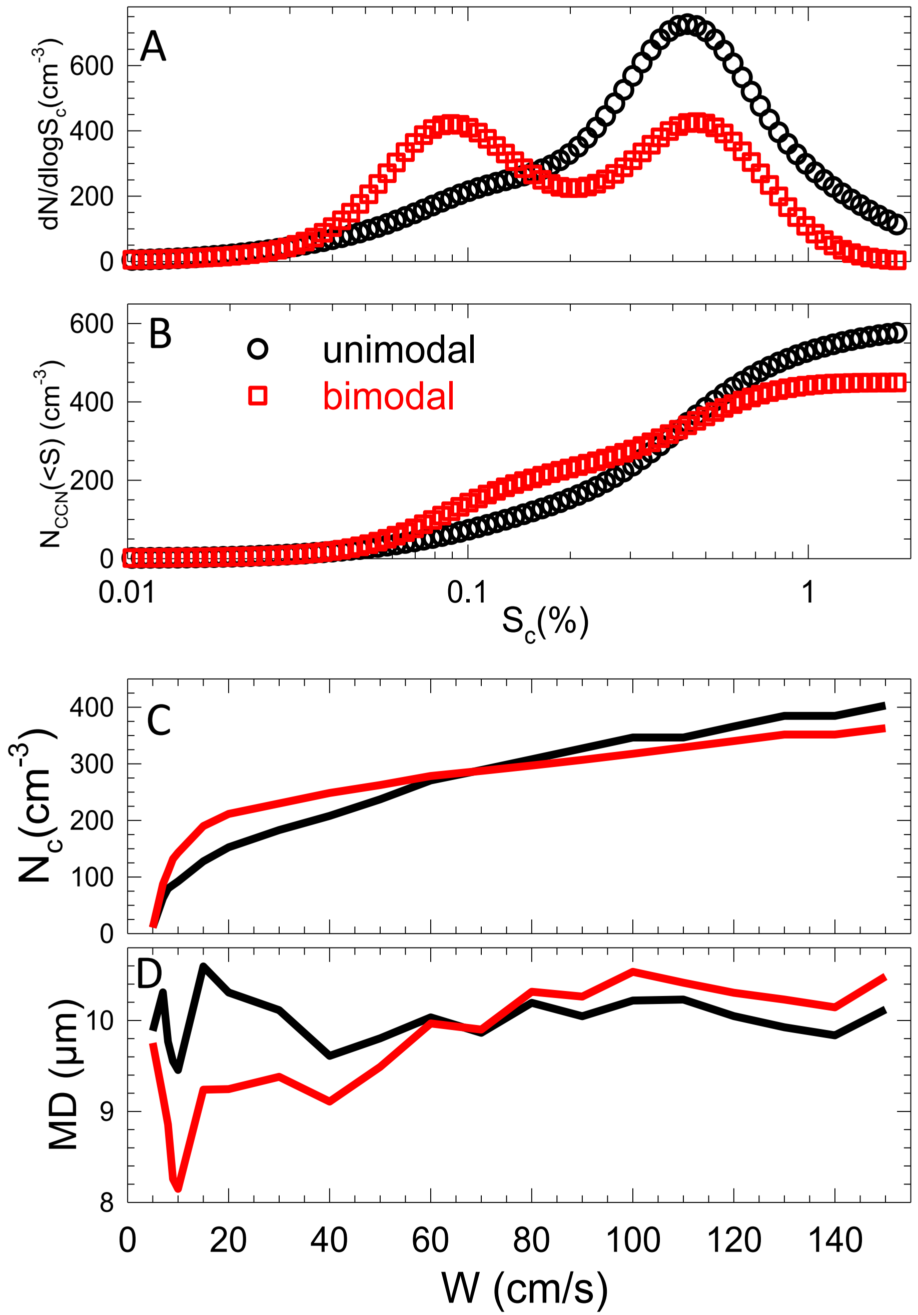
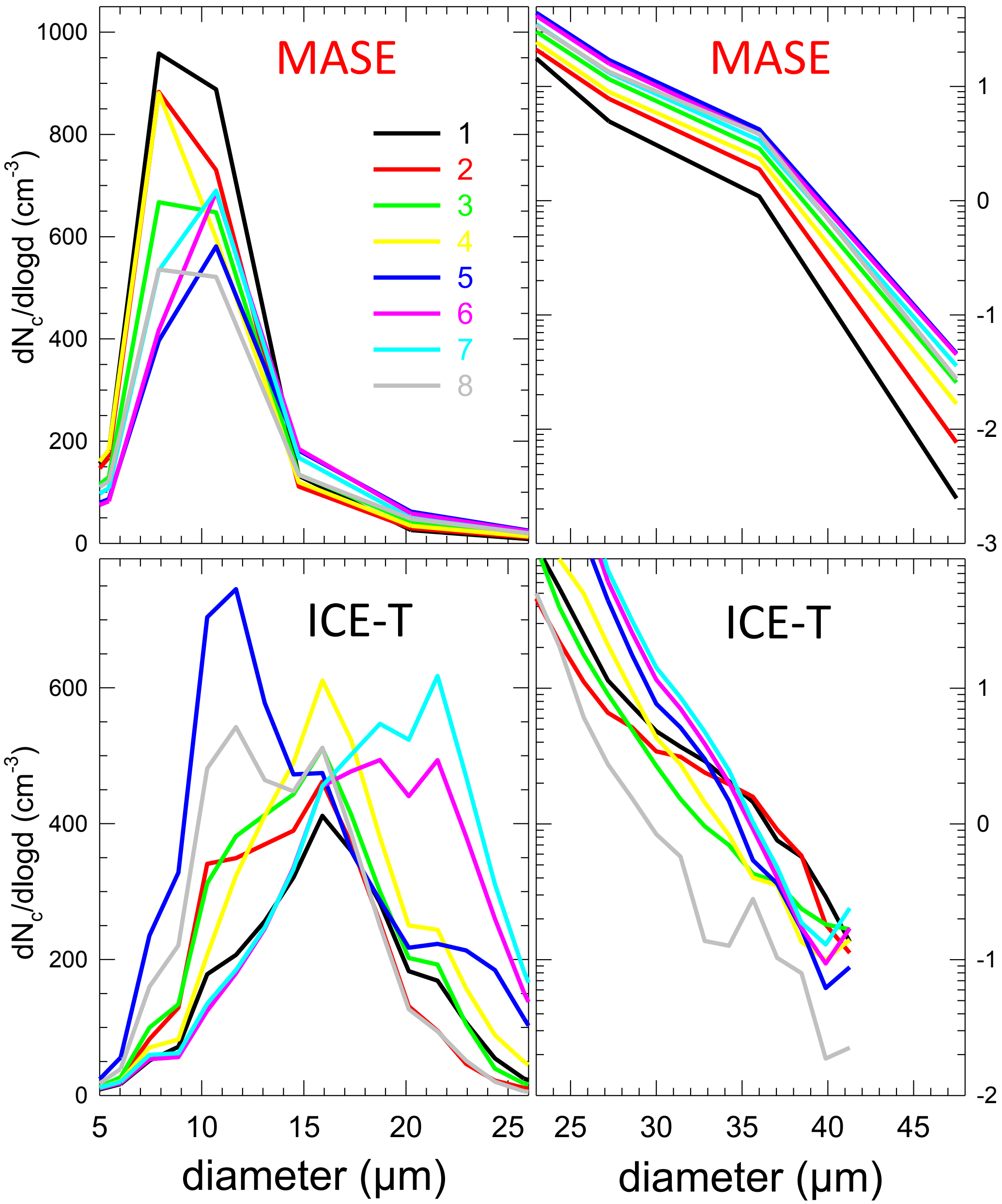


Fig. 5. (a) and (b) are measured CCN spectra during MASE. (a**)** is differential and (b) is cumulative presentation. (c and d) are cloud microphysics predictions by an adiabatic model (Robinson 1984) that applies various W to these two CCN spectra. (c) Shows cloud droplet concentrations. (N_c)
(d) is the mean diameter of the cloud droplet spectra at the same distances above cloud base for clouds grown on each CCN spectrum.

At low W typical of stratus, droplet concentration, N_c, is greater for clouds grown on bimodal CCN and droplet mean diameter (MD) is smaller.
At high W typical of cumuli N_c is greater for clouds grown on unimodal CCN and MD is smaller.

Similar results are found for ICE-T CCN spectra.



Fig, 6. Differential N_c against diameter for the various modal ratings noted by the legend colors 1-8. Right side labels show only exponents of the common-log scale.

In MASE small droplets are more numerous in clouds grown on bimodal CCN (lower ratings; black, red, green), but larger droplets are more numerous in clouds grown on unimodal CCN (higher ratings; gray, cyan, pink).

In ICE-T small droplets are more numerous in clouds grown on unimodal CCN (higher ratings; gray, cyan, pink), but larger droplets are more numerous in clouds grown on bimodal CCN (lower ratings; black, red, green).

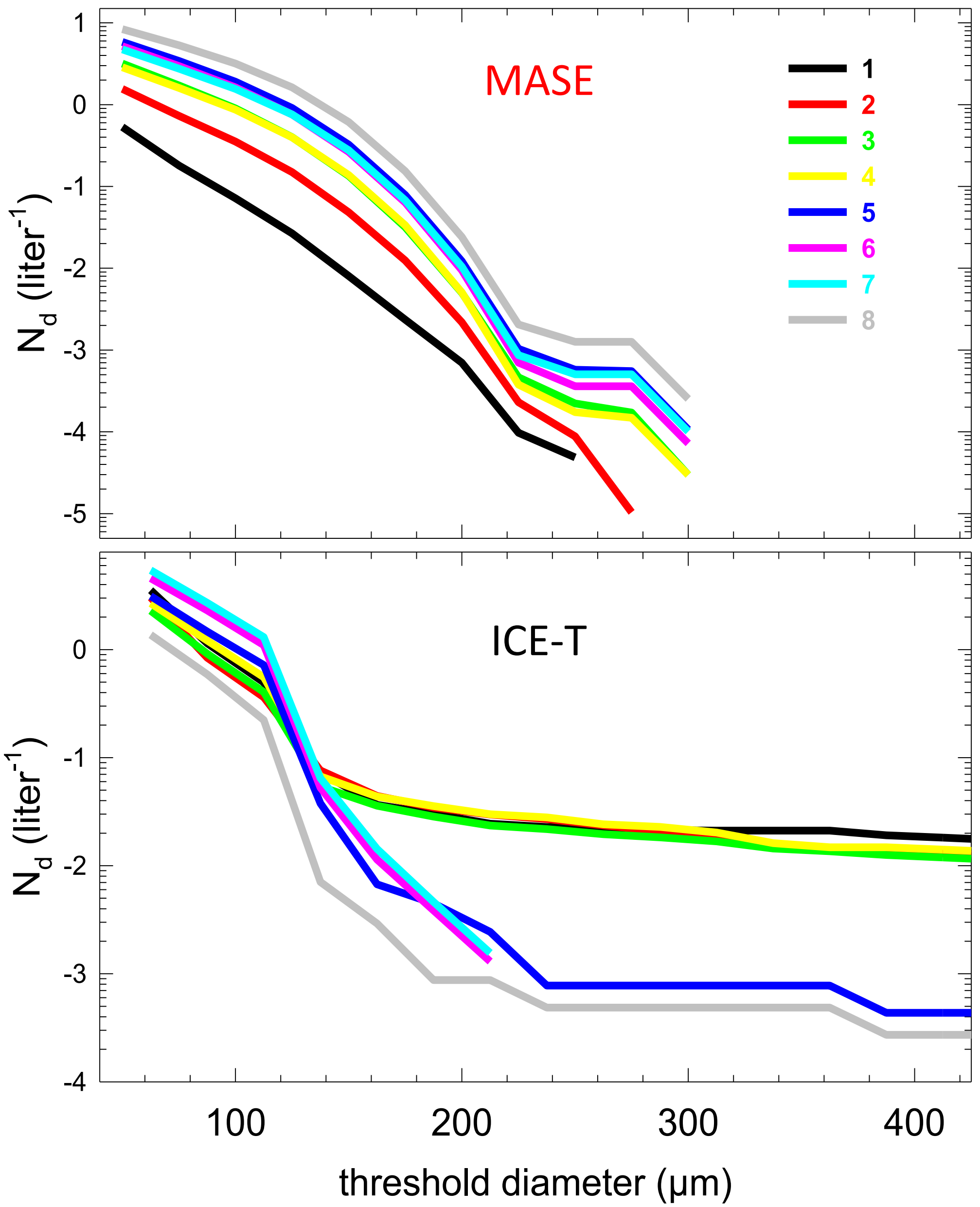


Fig. 7. Mean cumulative drizzle drop concentrations (N_d) larger than various threshold diameters for various modal ratings denoted by the legend colors 1-8.

In MASE stratus there is more drizzle in clouds grown on unimodal CCN (high ratings; gray, cyan, pink, blue).

In ICE-T cumuli there is more drizzle in clouds grown on bimodal CCN (low ratings; black, red, green, yellow).

Similar results for drizzle LWC

Opposite results in these two projects

CCN modality has opposite cloud microphysics relationships in two different cloud systems.

Thus, cloud processing enhances both components of the indirect aerosol effect (IAE) in polluted stratus,

Cloud processing reduces both IAE in cumuli.

If cloud processing continues to show importance then trace gas concentrations (sulfur dioxide, ozone, hydrogen peroxide, etc.), emissions and predictions will be as necessary as CCN measurements and predictions for assessing IAE.

Therefore, it would be necessary to learn more about natural versus relevant anthropogenic trace gases and their distributions relative to cloud fields that are most significant for climate; i.e., mainly marine stratus that largely control IAE, and where chemical processing dominates.

This would have to include better assessments of the relative anthropogenic/natural ratios of the relevant trace gases. Not just anthropogenic/natural ratios of CCN

These cloud processing effects are above and beyond the effects of different CCN concentrations (N_{CCN}) caused for instance by anthropogenic sources that cause the original indirect aerosol effect (IAE).

However, further cloud processing investigations might reveal the opposite;
Cloud processing makes the best CCN, lowest S_c.
Cloud processing makes most of the accumulation mode.
S is lower in stratus due to lower vertical wind.
Polluted status S is suppressed by high N_{CCN}.
Thus, only a small subset of CCN (lowest S_c) are activated and thus cloud processed.

In subsequent cloud cycles slightly higher S_c CCN thus are made irrelevant.

With the cloud processing improvement, in the next cycle not even all of the previously processed CCN make droplets. This further suppresses cloud S; and this makes irrelevant even more higher S_c CCN.

Anthropogenic CCN tend to have high S_c.

Many high S_c CCN are lost to Brownian scavenging by cloud droplets. “Clouds are the vacuum cleaners of the atmosphere” (Volker Mohnen, circa 1985)

Cloud processing then tends to reduce IAE.

Cloud processing could therefore constitute IAE buffering (Feingold and Stephens, 2006).

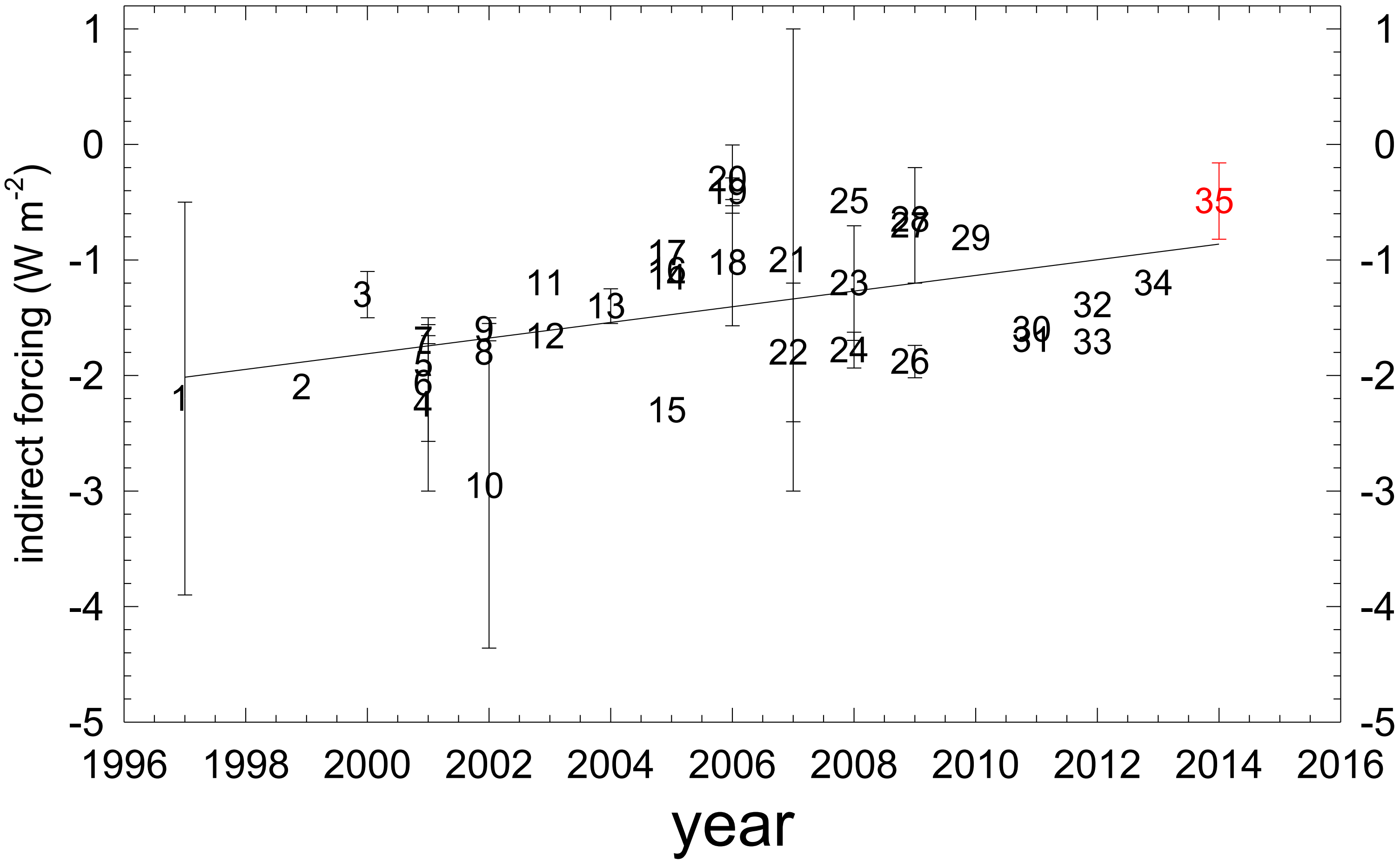


Fig. 8. IAE estimates mostly from the 5th ICCP report. 1. Lohmann and Feichter (1997); 2. Rotstayn (1999); 3. Lohmann et al. (2000); 4. Ghan et al. (2001); 5. Jones et al. (2001); 6. Rotstayn and Penner (2001); 7. Williams et al. (2001); 8. Kristjansson (2002); 9. Lohmann (2002); 10. Menon et al. (2002); 11. Peng and Lohmann (2003); 12. Penner et al. (2003); 13. Easter et al. (2004); 14. Kristjansson et al. (2005); 15. Ming et al. (2005); 16. Rotstayn and Liu (2005); 17. Takemura et al. (2005); 18. Penner et al. (2006); 19. Quaas et al. (2006); 20. Storelvmo et al. (2006); 21. Menon and Del Genio (2007); 22. Ming et al. (2007); 23. Kirkevag et al. (2008); 24. Seland et al. (2008); 25. Storelvmo et al. (2008); 26. Hoose et al. (2009); 27. Quaas et al. (2009); 28. Rotstayn and Liu (2009); 29. Storelvmo et al. (2010); 30. Ghan et al. (2011); 31. Penner et al. (2011); 32. Makkonen et al. (2012); 33. Takemura (2012); 34. Kirkevag et al. (2013); 35. Chen et al. (2014) satellite.

IAE estimates have decreased over the years

Satellite measurements and other observations do not see as much IAE as predicted by models.

Stevens, B., 2013: Aerosols: Uncertain then, irrelevant now. Nature, 503, 47.

“**Uncertainty in estimates of the effects of aerosols on climate stems from poor knowledge of the past, pristine atmosphere — so getting a better understanding of these effects might not be as useful as was thought.**”

Clouds make the best CCN