CCN Bimodality and Cloud Microphysics

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High resolution CCN spectra from MASE (polluted California stratus) and ICE-T (Caribbean cumuli) are presented along with adjacent cloud microphysics characteristics. Three cloud processes cause bimodal CCN spectra by increasing the mass of dissolved material within cloud droplets.

1) Coalescence among droplets;
2) Gas-to-particle chemical reactions, sulfate and nitrate;
3) Brownian capture of interstitial material, often CCN within unactivated droplets.

Since these cloud processes do not affect the unactivated particles a gap in the dry particle size distribution often occurs when cloud droplets evaporate. The sizes at these minima (Hoppel et al., 1985) have been used to infer cloud supersaturations (S) but those estimates depended on assumptions of particle composition in order to convert size to S. Hoppel minima in CCN spectra do not require knowledge of particle composition. The considerable variability in CCN spectral shapes is compared with cloud microphysics characteristics to ascertain if and how cloud processing affects clouds.

CCN spectral modality was quantified by subjective ratings by the authors on a 1-8 scale. The most bimodal spectra with well separated modes and somewhat equal mode peaks were rated 1. Strictly monomodal spectra were rated 8. Intermediate ratings were given to more asymmetric bimodal spectra, or bimodal spectra not so well separated (i.e., one mode a shoulder of another mode) or spectra with more than two modes. All spectra with modal ratings up to 4 provided Hoppel minima such that S_{eff} could be estimated. Rating 8 did not provide S_{eff}. Spectra with modal ratings 5, 6 and 7 provided S_{eff} at decreasing rates.

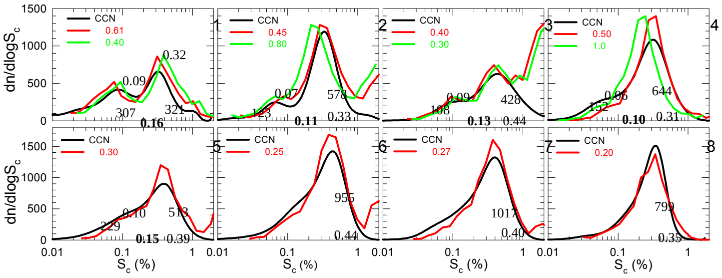

Fig. 1. MASE examples of simultaneous CCN and differential mobility analyzer (DMA; red and green) spectra for each of the 8 modal rating categories. 1 is most bimodal, 8 is strictly monomodal. CCN concentrations (cm^{-3}), N_{CCN}, within each mode (cloud processed to the left side, cloud unprocessed to the right side) and modal critical supersaturations (S_c) in percent are shown as well as the Hoppel minima S_c (**bold**). DMA sizes are converted to S_c by applying hygroscopicity (kappa) values. Kappa that provide best agreement with the CCN spectra are the ambient kappas shown in red and green of the legends. Variations of kappa with size or S_c are shown by the two different values in some panels.

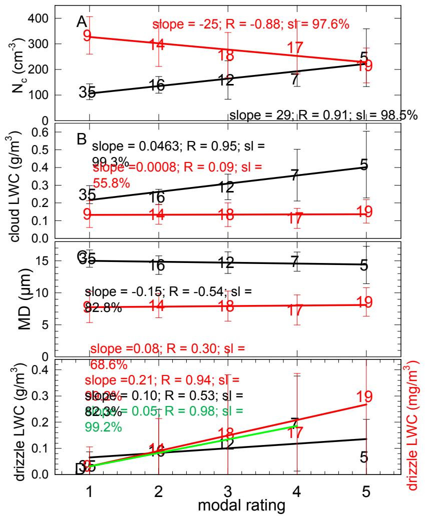

Fig. 2. Mean (displayed as number of cases) and standard deviation, sd (error bars), of cloud microphysics for categories of CCN spectra (demonstrated in Fig. 1) of nearby out-of-cloud measurements. Only modes 1-5, which are bimodal enough to provide Hoppel minima are considered. Linear regressions with 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.

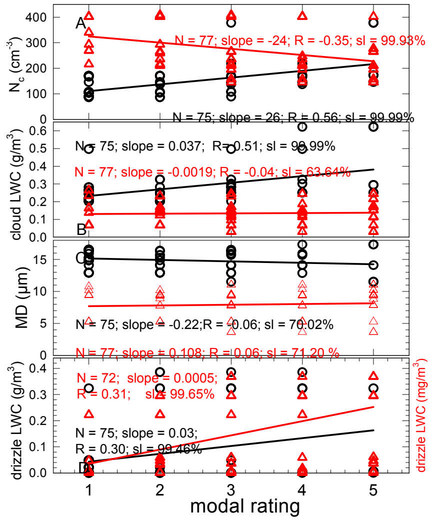

Fig. 3. As Fig. 2 but showing the individual cases.

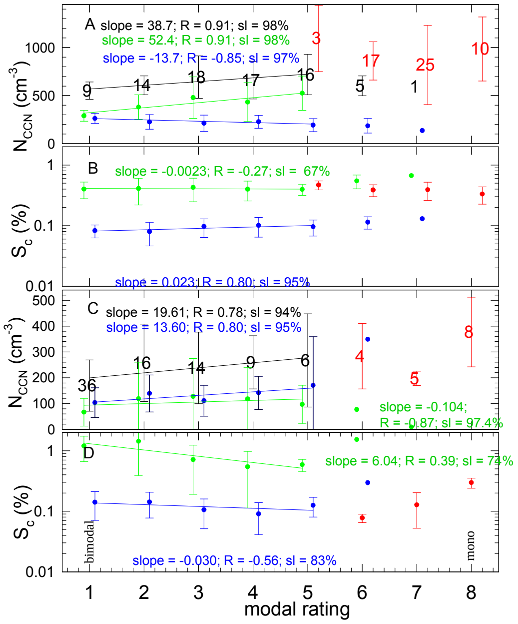

Fig. 4. Panels (a) and (c) display mean and sd of CCN concentrations, N_{CCN}, within modal categories against modal categories. Panels (b) and (d) display mean and sd of mode S_c of the three modes. Green is the unprocessed mode of the bimodal spectra. Blue is the processed mode of the bimodal spectra. 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).

Figs. 2 and 3

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.

Fig. 4 a and c show lower N_{CCN} for bimodal spectra (black) than for monomodal spectra (red) as well as lower N_{CCN} for lower modal ratings—sloping black lines. This indicates that cloud processing reduces N_{CCN}; in ICE-T by coalescence and in MASE by Brownian capture. The decreasing unprocessed N_{CCN} (green) with greater bimodality in Fig. 4a reflect Brownian capture of high S_c CCN while the opposite trend for processed CCN (blue) indicates both Brownian capture and chemistry making more better (lower S_c) CCN.

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.

Cloud-processing of CCN spectra is as important as CCN sources; it alters CCN concentrations, N_{CCN}, S_c spectra, cloud supersaturations (S_{eff}), particle hygroscopicity (kappa), droplet concentration (N_c), droplet sizes (MD), droplet spectral width (σ) and consequently cloud albedo (first indirect aerosol effect, IAE) and precipitation (second IAE). These changes could be additional or reverse IAE; i.e., buffering. Brownian scavenging of high N_{CCN} from anthropogenic sources could be a very important buffering effect that could considerably nullify IAE. However, this could in turn be reduced by the resulting improved (lower S_c) CCN.

Cloud processing of CCN could also influence the direct aerosol effect (e.g., Varnai and Marshak, 2014). The extent of cloud processing needs to be assessed. Further analysis of these high-resolution CCN spectra in other field projects is proceeding.

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Varnai, T. and A. Marshak (2014), Near cloud aerosol properties from the 1 km resolution MODIS ocean product, *J. Geophys. Res. Atmos., 119*, 1546-1554, doi:10.1002/2013JD020633.