Bimodal CCN Spectra and Cloud Processing

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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. 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.

Since these cloud processes do not affect unactivated particles a gap in the dry particle size distribution occurs when droplets evaporate. Sizes at minima between the unprocessed and cloud processed modes are referred to as Hoppel minima (Hoppel et al., 1985). Variability in CCN spectral shapes is compared with cloud microphysics.

CCN spectral modality was quantified on a 1-8 scale. The most bimodal spectra with well separated modes and somewhat equal mode peaks were rated 1. Strictly unimodal 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).

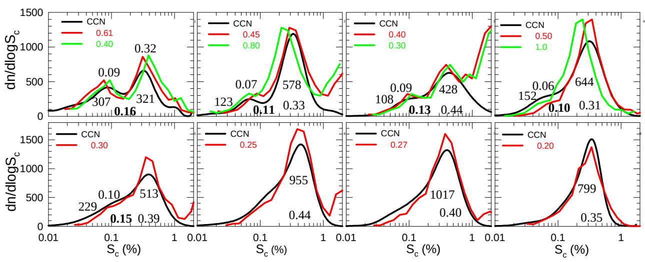

Fig. 1. MASE examples of simultaneous CCN and differential mobility analyzer (DMA; red and green) spectra for each of the 8 modal ratings. CCN concentrations (cm^{-3}), N_{CCN}, within each mode (cloud processed to the left side, cloud unprocessed to the right side) and modal critical supersaturations (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.

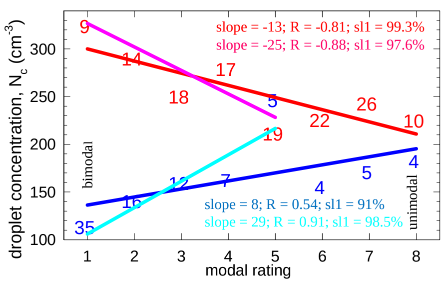

Fig. 2. Mean cloud droplet concentrations, N_c, against modal rating of the closest CCN spectra for MASE (red and pink; polluted stratus) and ICE-T (blue and cyan; Caribbean cumuli). Correlation coefficients (R) and one-tailed significance levels (sl1) are shown. Pink and cyan regressions consider only modes 1-5 that have Hoppel minima. Data points are plotted as numerals of the numbers of cases. From Hudson et al. (2015).

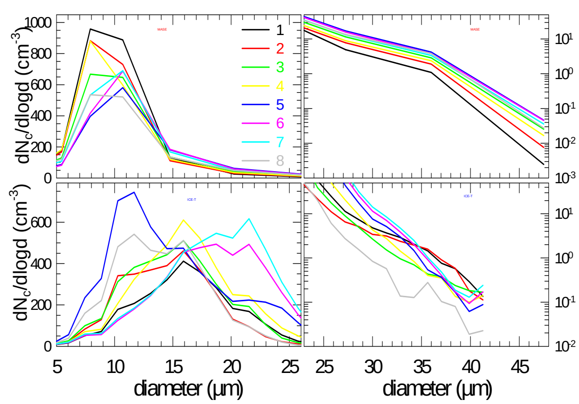

Fig. 3. Differential N_c against diameter for the various modal ratings noted by the legend colors 1-8. In MASE small droplets are more numerous in clouds grown on bimodal CCN (lower ratings), but larger droplets are more numerous in clouds grown on unimodal CCN (higher ratings). In ICE-T small droplets are more numerous in clouds grown on unimodal CCN (higher ratings), but larger droplets are more numerous in clouds grown on bimodal CCN (lower ratings). From Hudson et al. (2016).

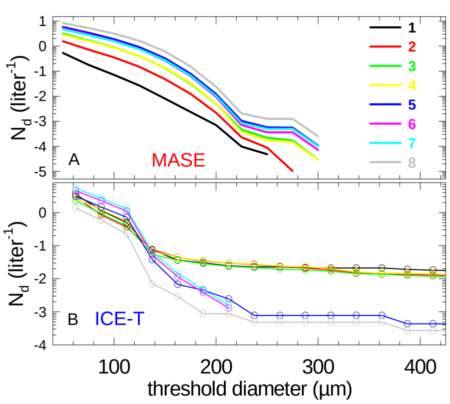

Fig. 4. Mean cumulative drizzle drop concentrations (N_d) larger than various threshold diameters within clouds closest to CCN spectra with various ratings of their modality denoted by the legend colors 1-8. In MASE stratus there is more drizzle in clouds grown on more unimodal CCN (high ratings). In ICE-T cumuli there is more drizzle in clouds grown on more bimodal CCN (low ratings). Ordinate labels are exponents of base 10 logs. Similar results for drizzle LWC. From Hudson et al. (2016).

In MASE cloud processing increases N_c and decreases droplet sizes; this decreases concentrations of larger drops and drizzle.

1st and 2nd IAE are enhanced by predominant chemical processing in stratus.

In ICE-T cloud processing decreases N_c and increases droplet sizes; this increases concentrations of larger drops and drizzle.

1st and 2nd IAE is reduced by predominant coalescence processing in cumuli

because greater vertical extent, larger droplets and greater LWC favor coalescence more than in stratus that are thinner, with smaller droplets and less LWC.

Surface measurements at SGP, May 2003

Surface aerosol bimodality/unimodality variations were correlated with remote measurements of cloud fraction (CF) and cloud base altitude (CBH). To account for aerosol travel from clouds to surface these correlation coefficients (R) were time adjusted so that earlier CF and CBH measurements are compared with later surface aerosol measurements.

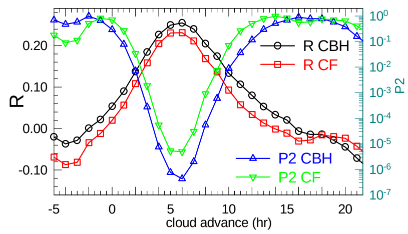

Fig. 5. Correlation coefficients (R) and two-tailed probabilities (P2) between hour averages of DMA SMPS N_u-N_p with ceilometer cloud base altitude (CBH) and cloud fraction (CF) against various hour advancements of the cloud data. CF R is multiplied by -1 in order to show that greater CF is related to lower (N_u-N_p), which is characteristic of more bimodal aerosol. From Noble et al. (2016).

As Fig. 5 shows, cloud presence was consistently implicated as the source of aerosol bimodality. A new objective categorization of aerosol modality, N_u-N_p (concentrations within the unprocessed and cloud processed modes), more precisely substantiated clouds as the source of aerosol bimodality. These compelling findings demonstrate that further analysis of abundantly available surface aerosol and remote sensing measurements could further advance understanding of cloud processing. This is important because the various cloud processes have different effects on aerosol (Figs. 2-4), which then have different effects on cloud microphysics (Figs. 2-4) that have significant implications for indirect aerosol forcing. Vast amounts of appropriate and now validated data at ARM sites can advance understanding of cloud processing. Around-the-clock and year-round coverage of surface measurements provide more data more economically than aircraft campaigns.

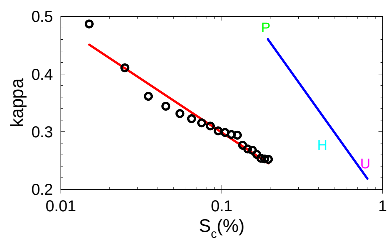

Fig. 6. Mean hygroscopicities, kappas, versus particle critical supersaturation, S_c, from 228 simultaneous CCN and DMA measurements covering 94 hours during May 2003 at SGP. P is at the peak of the processed modes, H is at the Hoppel minima, U is at the peak of the unprocessed mode. The black data are for smaller numbers of measurements when and where the two spectra were in agreement within the narrow S_c ranges.

Greater kappa for lower S_c (larger particles) and for the processed mode than the unprocessed mode indicate chemical processing.

Cloud-processing of CCN spectra is as important as CCN sources; it alters CCN concentrations, spectra, cloud supersaturations, kappa, N_c, droplet sizes, droplet spectral width and consequently cloud albedo (1st IAE) and precipitation/cloud lifetime (2nd IAE). These changes could be additional or reverse IAE; i.e., buffering.

Since marine stratus are the main IAE drivers, cloud processing appears to be a net enhancement of IAE. However, the improvement of CCN (lower S_c and greater kappa) by cloud processing may trump anthropogenic CCN from making subsequent clouds and thus thwart IAE. If so then the anthropogenic contributions to trace gases that cause chemical processing take on greater importance. The extent of cloud processing needs to be assessed.

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