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Hoppel Minima in Detailed CCN Spectra

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Measurements are presented from two aircraft cloud research projects: Marine Stratus/Stratocumulus Experiment (MASE) off the central California coast and Ice in Clouds Experiment-Tropical (ICE-T) in cumulus clouds of the eastern Caribbean. Both occurred in July, MASE 2005, ICE-T 2011.

DMA dry aerosol spectra below stratus clouds often display bimodality attributed to cloud processing (physical—coalescence and Bownian scavenging or chemical—reactions within droplets) that increase particle sizes and reduce the critical supersaturation, S (S_c) of CCN that had produced 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) but that required particle hygroscopicity (κ) in order to convert size to S_c. Since these Hoppel minima occurred at diameters greater than 70 nm and since ammonium sulfate was assumed, this inferred $S_{eff} < 0.3\%$.

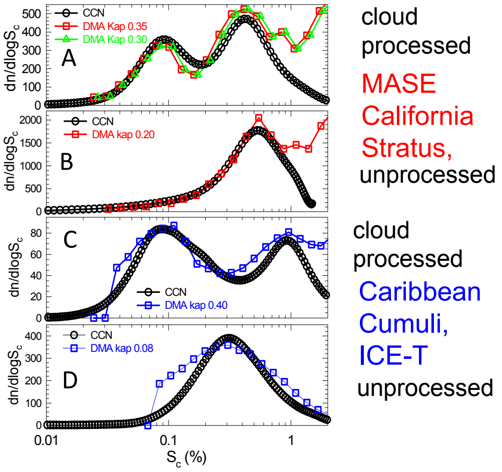

Figure 1. CCN and DMA concentrations against S_c. In a κ 0.30 fits lower S_c mode (cloud-processed); κ 0.35 fits higher S_c mode (unprocessed). Hoppel minimum between modes implies S_{eff} 0.20% for a, 0.40% for c.

When all channels of the DRI CCN spectrometers are plotted, bimodality is seen in six aircraft projects. This provides S_{eff} sans particle composition (κ). In MASE and ICE-T, there were also DMA measurements to compare with CCN spectra by transposing size to S_c by applying various κ. Certain κ produced DMA distributions that agreed with CCN spectra over most of the S range (Figs. 1 and 2). κ that resulted in agreement was then the hygroscopicity of the subject aerosol. DMA--CCN agreement was consistent throughout both projects (Figs. 1 and 2). κ differences between the two modes could distinguish chemical from physical cloud processing; chemistry should push κ of the lower S_c mode (the cloud processed mode) toward κ of ammonium sulfate, 0.61; i.e., which was higher than the typically ambient κ observed in these projects (Table 1).

Spectral modality is quantified on a 1-8 scale. The most bimodal spectra with well separated equal modes are rated 1 (Figs. 1a and c, 2a). Strictly monomodal spectra are rated 8 as in Figs. 1b and d and 2h). Intermediate ratings are for asymmetric or less separated bimodal spectra; e.g., shoulder modes (Fig. 2b-g). Mode ratings up to 4 provided Hoppel minima, S_{eff}. Ratings 7 or 8 did not provide S_{eff}. Ratings 5 or 6 sometimes provided S_{eff}.

	cases	cases S	κ_p	κ_u	mode	S_{eff}Hop (%)	S_{eff}spec	$N_{1\%}$
MASEbelow	135	80	0.50	0.45	4.80	0.16	0.17	703
MASEabove	92	36	0.45	0.26	5.91	0.27	0.17	1215
ICE-T	50	41	0.33	0.35	3.26	0.52	1.30	189

Table 1. MASE data is divided by the stratus; below and above. 2nd column is the number of comparisons, 3rd column is the number of cases that provided Hoppel minima, S_{eff}, 4th column is mean hygroscopicity, κ, of the processed modes (lower S_c), 5th column is mean κ of the corresponding unprocessed modes (higher S_c), 6th column is the mean mode rating, 7th column is mean S_{eff} from Hoppel minima, 8th column is S_{eff} by matching below cloud CCN spectra with mean cloud droplet concentration of the nearest cloud, 9th column is CCN concentration at 1% S.

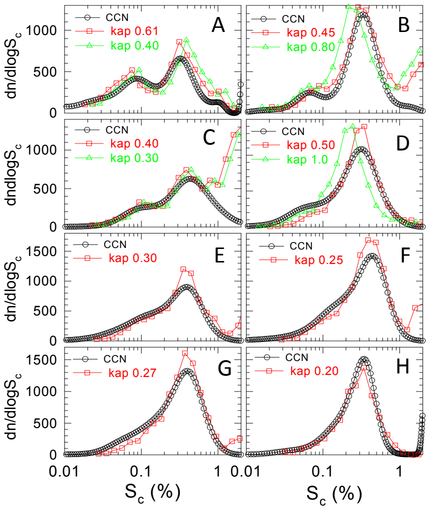

Figure 2. MASE examples of simultaneous CCN and DMA distributions for each of the 8 mode ratings. (a) mode 1, July 23, 11:15:23-11:15:33; (b) mode 2, July 18, 12:09:05-12:09:35; (c) mode 3, July 15, 10:58:30-11:00:12; (d) mode 4, July 18, 12:07:24-12:08:29; (e) mode 5, July 18, 11:57:02-11:58:20; (f) mode 6, July 18, 12:18:47-12:18:09; (g) mode 7, July 18, 11:12:15:21-12:15:47; (h) mode 8, July 19, 11:59:29-12:00:00. DMA κ (kap) are shown in legend.

As expected S_{eff} is higher for the cumulus clouds (ICE-T) where there is disagreement between the two methods. Surprisingly there is more bimodality (more cloud processing) for the cumulus clouds (lower mode rating) than for stratus clouds. Higher κ for the processed mode in MASE suggests chemical processing that is not indicated for ICE-T.

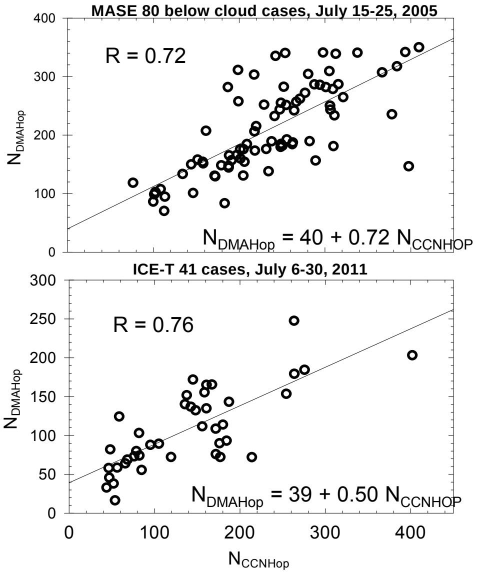

Figure 3. Comparisons of particle concentrations within the cloud processed modes; i.e., left side modes in Fig. 1a and c and 2a-e. Linear regressions are shown.

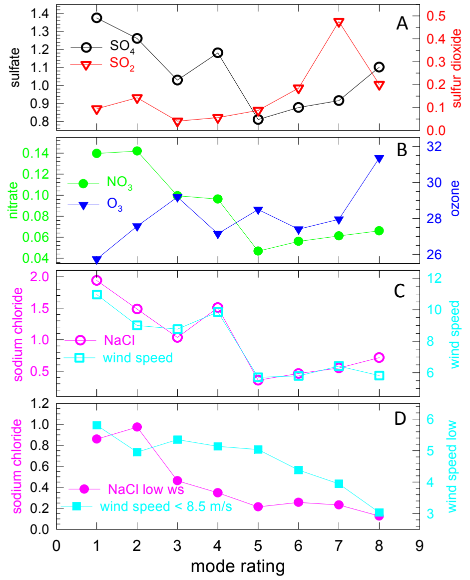

Figure 4. Mean ionic concentrations for each of the modal ratings for all MASE DMA-CCN comparisons. Mean wind speed is shown in c and d. (d) data only for wind speeds < 8.5 m/s.

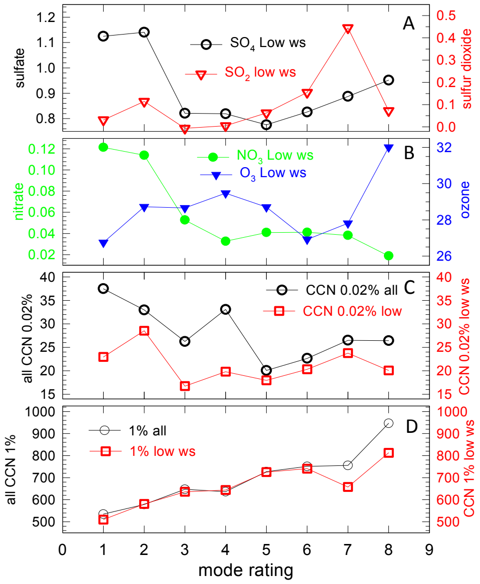

Figure 5. a and b are like Fig. 4a and b except only for data obtained with wind speeds < 8.5 m/s. c and d show CCN concentrations for all data and for data with wind speed < 8.5 m/s.

Figure 3 displays the agreement between the DMA and CCN spectra for the cloud processed mode.

Figure 4a indicates gas-to-particle chemical conversion of sulfur dioxide to sulfate aerosol since SO_4 concentrations are greater for more bimodal spectra (lower mode rating) and SO_2 concentrations are lower. For more monomodal aerosol, higher ratings (less cloud processing), there is less SO_4 and more SO_2. Figure 4b indicates similar nitrate aerosol production and associated ozone depletion, which tracks opposite to nitrate. Figure 4c suggests physical cloud processing since NaCl is an excellent CCN that could coalesce droplets to produce more massive aerosol. However, since NaCl tracks with wind speed this might just be breaking wave sea salt production. **But** Fig. 4d shows that in lighter winds that produce less sea salt and where NaCl does not track with wind speed, there is still greater NaCl at low modal ratings.

Figures 5a and b indicate the same gas-to-particle chemical transformations under light winds.
Figure 5c shows higher concentrations of low S_c CCN for more bimodal spectra whereas Fig. 5d shows lower concentrations of total CCN for more bimodal spectra. This may indicate coalescence among cloud droplets and Brownian scavenging of interstitial particles that reduce total CCN and increase low S_c CCN.

Conclusions:

Aerosol bimodality can be detected in CCN spectra.
Hygroscopicity (κ) can be inferred from DMA comparisons.
Bimodality is not universal even under solid stratus; monomodal and bimodal spectra are intermingled.
Bimodal aerosol/CCN spectra are just as common under cumulus clouds as stratus clouds.
Both chemical and physical cloud processing are indicated.

Cloud processing could buffer the indirect aerosol effect by reducing S_{eff}, which could deprive higher concentrations of high S_c CCN from forming cloud droplets. The extent of cloud processing requires as much attention as CCN sources.

Hoppel et al., 1986: *Geophys. Res. Lett.*, **13 (1)**, 125-128.

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