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

JAMES G. HUDSON and STEPHEN NOBLE

Desert Research Institute, University of Nevada, Reno
Reno, Nevada 89512-1095 USA
hudson@dri.edu
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.** Effective cloud supersaturation ($S_{\text{eff}}$) against CCN concentration at 1% S ($N_{1\%}$) black POST; red MASE. $S_{\text{eff}}$ is the S for which nearby below cloud $N_{\text{CCN}}(S)$ equals mean droplet concentration ($N_c$). Linear regression lines are shown.
Fig. A. (a) Mean $N_c$ versus $\sigma_w$ 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

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 $S$, ($S_{\text{eff}}$) (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 $S_{\text{eff}}$ sans particle composition ($\kappa$).
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, $S_{\text{eff}}$. Ratings 7 and 8 did not provide $S_{\text{eff}}$. Ratings 5 and 6 sometimes provided $S_{\text{eff}}$. 
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.
<table>
<thead>
<tr>
<th>cld</th>
<th>cases</th>
<th>mode</th>
<th>$S_{\text{eff}}S(%)$</th>
<th>$S_{\text{eff}}H(%)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MASE</td>
<td>St</td>
<td>80</td>
<td>4.80</td>
<td>0.23</td>
</tr>
<tr>
<td>ICE-T</td>
<td>Cu</td>
<td>80</td>
<td>2.93</td>
<td>1.03</td>
</tr>
</tbody>
</table>

**Table 1.** 2\textsuperscript{nd} column is the number of cases that provided Hoppel minima, 3\textsuperscript{rd} column is mean modal rating, 4\textsuperscript{th} column is mean traditional $S_{\text{eff}}$ by matching below cloud CCN spectra with mean cloud droplet concentration, $N_c$, of the nearest cloud, 5\textsuperscript{th} column is mean $S_{\text{eff}}$ from Hoppel minima.
Figure 5. As Fig. 4 but ratio of $S_{\text{eff}}H$ to $S_{\text{eff}}S$ for MASE (black) and ICE-T (red); data from Fig. 5.
\[ S_{\text{eff}H} < S_{\text{eff}S} \]

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 \( S_{\text{eff}} \) 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. Nc 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 $S_m$, $S_{effH}$ (%) (bold) and mean of $N_{CCN}M$, $N_{CCN}U$ and $N_{CCN}P$ are shown (cm$^{-3}$).
\[ \text{total} = \text{unprocessed} + \text{processed} \]

\[ N_{\text{tot}} < N_{\text{monomodal}} \quad \text{and} \quad N_{\text{tot}} \quad \text{decreases with bimodality for both projects implies physical processing} \]

\[ \text{MASE:} \quad N_{\text{unp}} \quad \text{decreases with bimodality Implies Brownian capture of highest } S_c, \text{ which are most mobile} \]

\[ N_{\text{proc}} \quad \text{increases with bimodality implies chemistry and Brownian} \]

\[ S_{\text{unp}} = S_{\text{cmono}} \]

\[ \text{ICE-T:} \quad S_{\text{unp}} > S_{\text{cmono}} > S_{\text{cproc}} \]

\[ S_{\text{unp}} \quad \text{increases with bimodality implies coalescence removing the lower } S_{\text{unp}}; \text{ i.e., largest droplets in that mode} \]

**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).
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.

**Acknowledgements.** MASE was supported by DOE grants DE-FG02-05ER63999 and DE-SC0009162. ICE-T was supported by NSF grant AGS-1035230,
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