Detailed CCN spectral measurements

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Bimodal submicrometer direct aerosol size spectra (Differential Mobility Analyzer; DMA) observed below marine stratus clouds

(Hoppel et al. 1986, 1994;

Clarke et al. 1996, 1998, 1999, 2004, 2013

and many others).

Hoppel minima (70-130 nm)

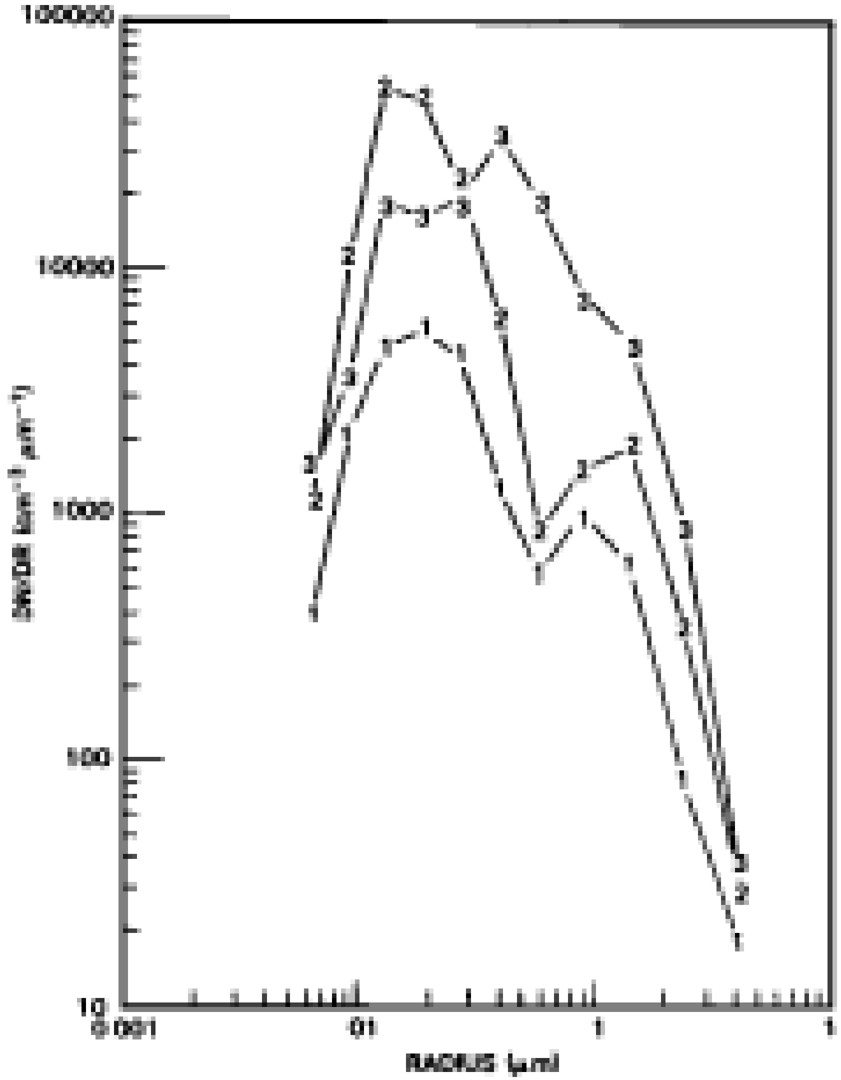


Fig 3. Curve 1 is size distribution before ship was downwind of the islands. Curve 2 was taken downwind of Tenerife. Curve 3 was taken downwind of Grand Canary Island.

Hoppel et a., 1986

Sizes at minima between modes inferred S_{eff} of nearby clouds.

Hoppel minimum diameter converted to S (critical S, S_c) by using solubility/hygroscopicity (κ); particle composition--ammonium sulfate, $\kappa = 0.61$.

Principle: lower S_c particles that nucleated cloud droplets are cloud-processed:

Physical (coalescence among droplets and Brownian capture of interstitial particles)

Chemical (gas-to-particle conversion of sulfate or nitrate) .

Increases soluble content of cloud droplets.
When droplets evaporate, dry particles emerge with **even lower S_c** than CCN that produced cloud droplets.
Unactivated particles remain at same size and S_c.

Size distribution/S_c **gap** at cloud S representes S_{eff} of clouds that processed aerosol.

Hoppel min. > 70 nm diameter and assuming ammonium sulfate so S_{eff} < 0.3% for marine stratus

But Hudson et al. (2010) and Hudson and Noble (2014)

S_{eff} > 1% for $N_{1\%}$ < 300 cm^{-3} in marine stratus

By comparing CCN spectra with droplet concentrations.

Hudson Wojciechowski Squires Twomey Hoppel

September, 1973, Sage Bldg., DRI

Multiple channel DRI CCN reveals bimodality so far in

RICO—Caribbean small cumuli, Dec-Jan. 2004-5;
MASE—Off Central California coast, polluted stratus, July, 2005
PASE—Central Pacific small clouds, Aug-Sep, 2007
POST—Off Central California coast stratus, July-Aug, 2008
ICE-T—Caribbean small cumuli, July, 2011.

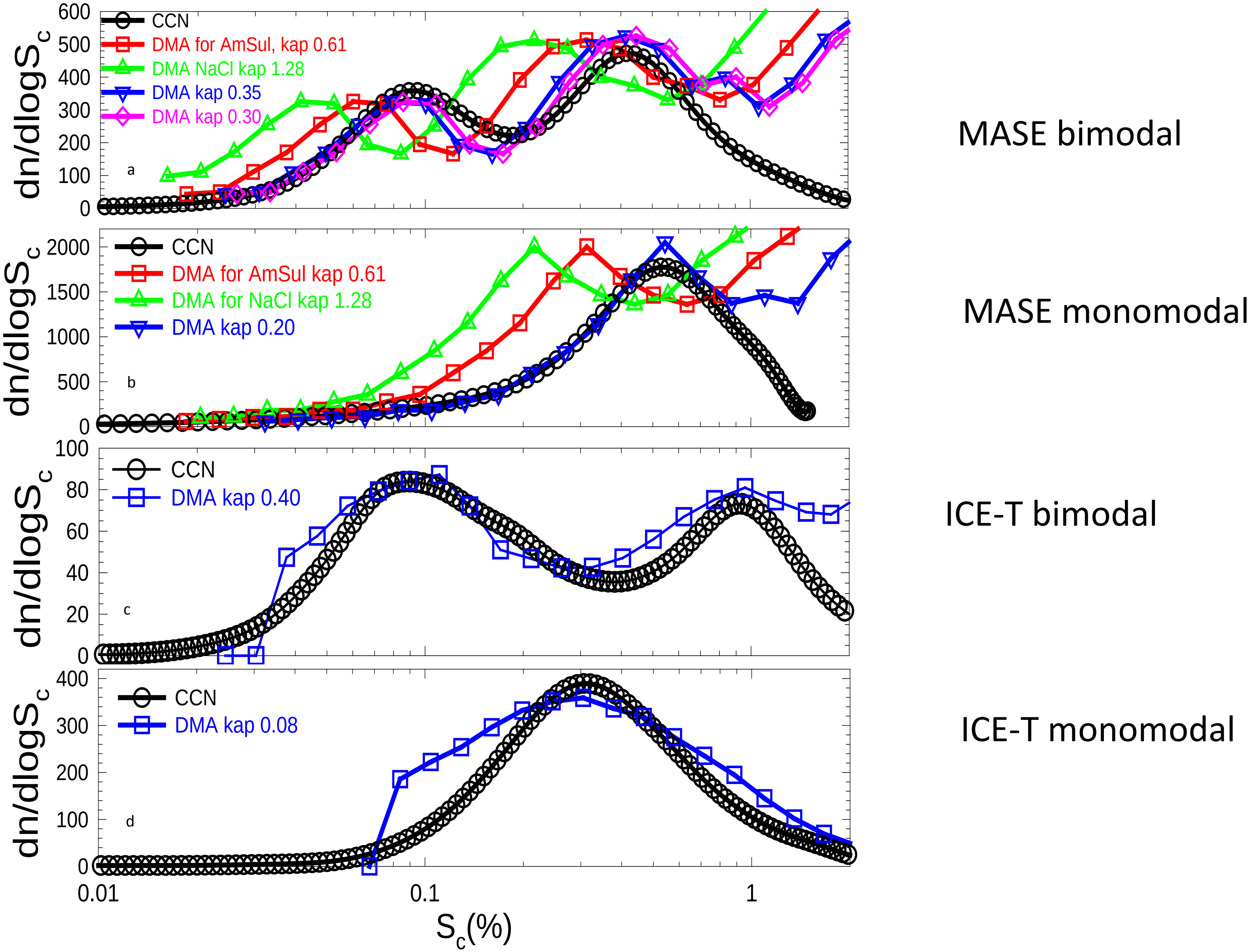
CCN are in terms of S, so κ (particle composition) is not required to estimate S_{eff} .

Simultaneous DMA measurements in MASE and ICE-T.

Compare with CCN when size is transposed to S_c by using specific κ's.

κ that makes DMA spectra agree with CCN spectra is CCN κ.

Consistent DMA-CCN agreement for 227 MASE and 50 ICE-T measurements.



CCN
DMA for AmSul, kap 0.61
DMA NaCl kap 1.28
DMA kap 0.35
DMA kap 0.30
a
DMA for AmSul kap 0.61
DMA for NaCl kap 1.28
DMA kap 0.20
b
DMA kap 0.40
c
DMA kap 0.08
d
dn/dlogS$_c$
S$_c$(%)
MASE bimodal
MASE monomodal
ICE-T bimodal
ICE-T monomodal

Sometimes slightly different κ for each mode.

κ 0.35 for high S_c mode; κ 0.30 for lower S_c cloud-processed mode (Fig. 1a).

Consistent opposite κ differences between modes indicates chemical processing because
Most κ < 0.61 (Amon. Sul.) (Fig. 2, Table)
Chemical processing should move κ toward 0.61.

Higher κ and less bimodality above than below MASE stratus (Table).
Consistent with higher N_{CCN} above cloud and less cloud interaction above than below cloud.

	flt	cas	sec	κ	mode	S_{eff} Hoppel	S_{eff} spec	N_c	$N_{1\%}$
MASE bel	9	135	7509	0.40±0.20	4.80±2.06	0.15±0.07	0.17±0.17	255± 88	703±263
MASE abv	9	92	1657	0.24±0.16	6.00±1.86	0.20±0.16	0.17±0.17	255± 88	1215±521
ICE-T	8	50	6162	0.34±0.22	3.26±2.23	0.52±0.26	1.31±0.54	164± 72	189±100

mode rating 1-8.

1 well-separated bimodal

8 definitely monomodal

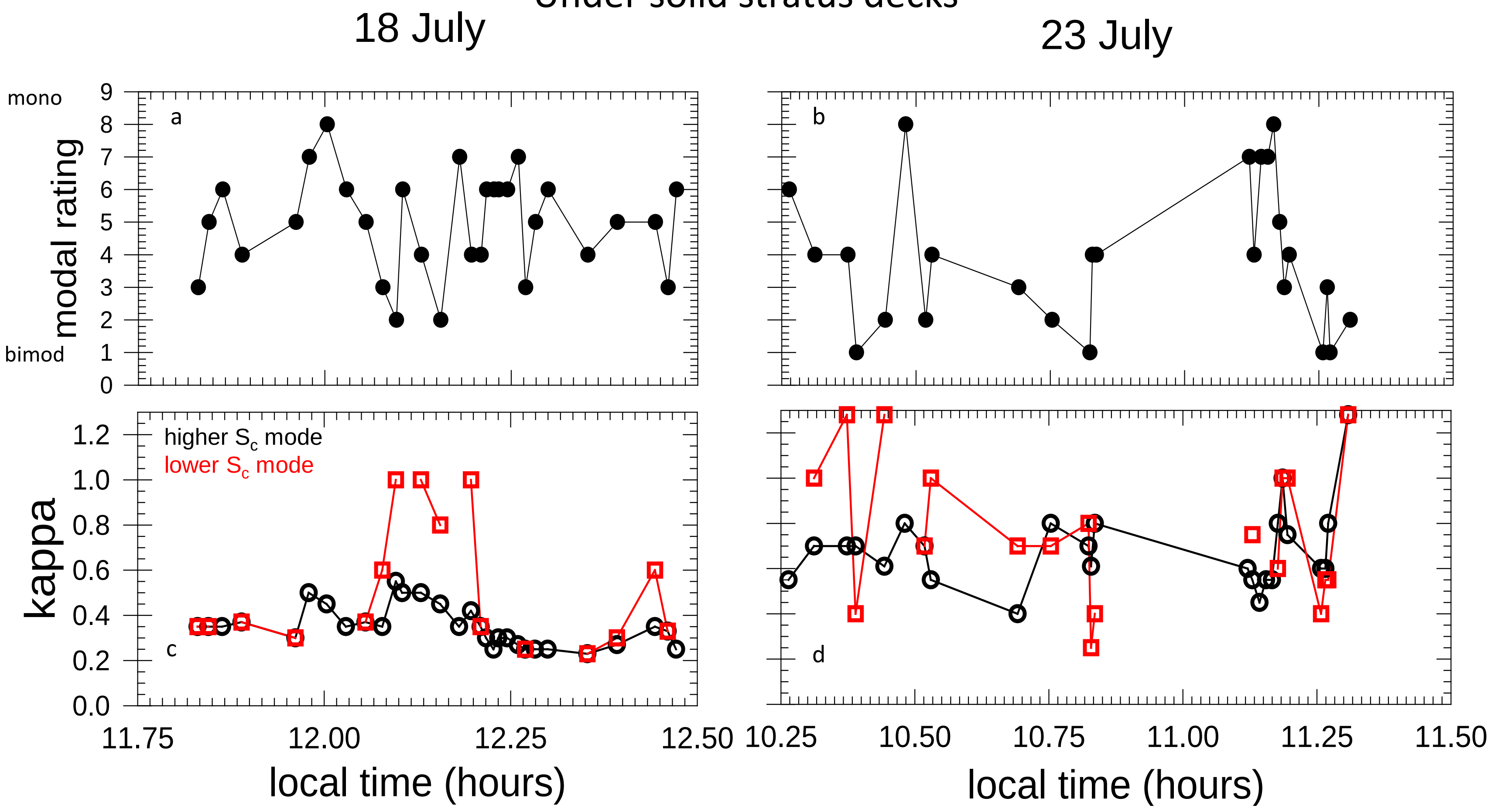
N_c cloud droplet concentration

$N_{1\%}$ CCN concentration at 1% S

S_{eff}spec by comparing CCN spectrum with N_c

MASE, 2005

Under solid stratus decks



Lower S_c mode is cloud processed

Bimodality not universal under ubiquitous MASE stratus (Fig. 2, Table) suggests not well-mixed boundary layer (Fig. 2).

Anthropogenic sulfate and nitrate that enhance cloud processing is another IAE.

Cloud processing alters the lowest S_c CCN; the most likely to form droplets.

Enhanced lowest S_c CCN might deprive higher S_c CCN with higher concentrations from forming cloud droplets. Another counter-IAE.

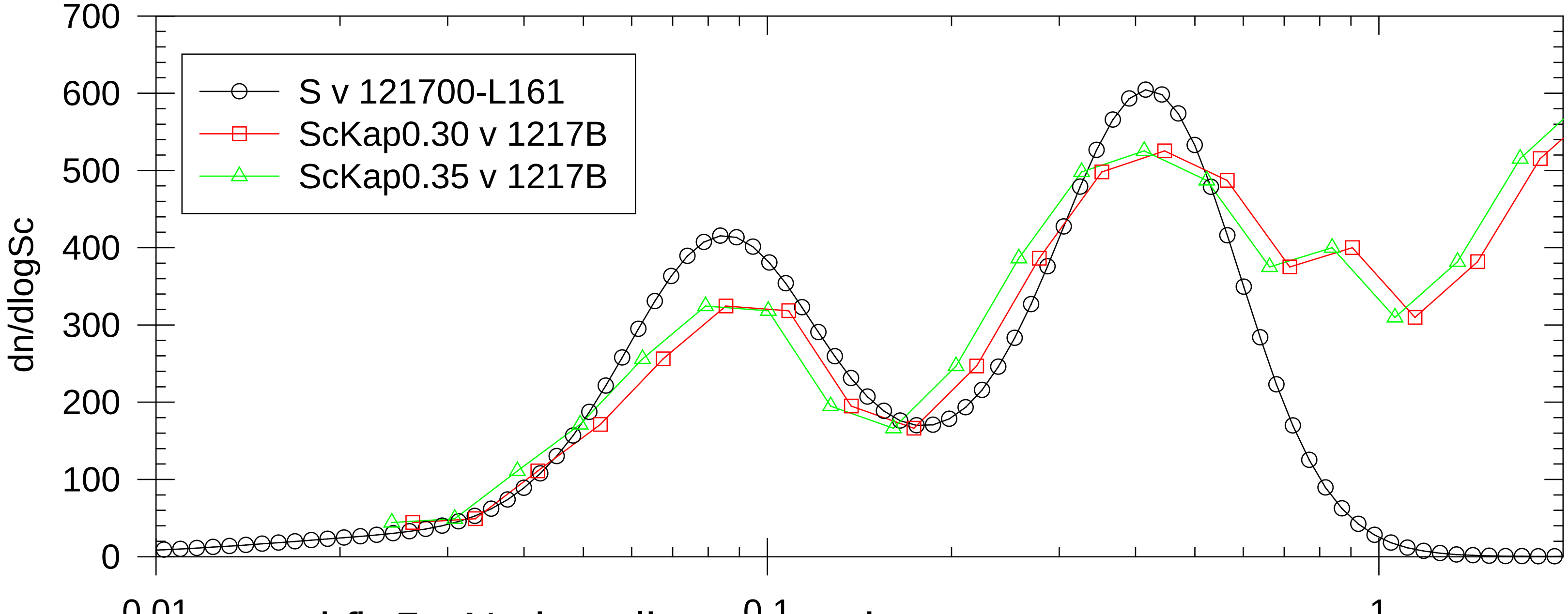
Cloud-processing alters S_{eff}, droplet concentration (N_c), mean diameter (MD), spectral width (σ) and cloud albedo.

S_{eff} differences between methods (Hoppel versus spectrum ;ICE-T, cumuli) may be because only larger droplets participate in cloud processing. Smaller droplets evaporate more easily (Xue and Feingold 2006).

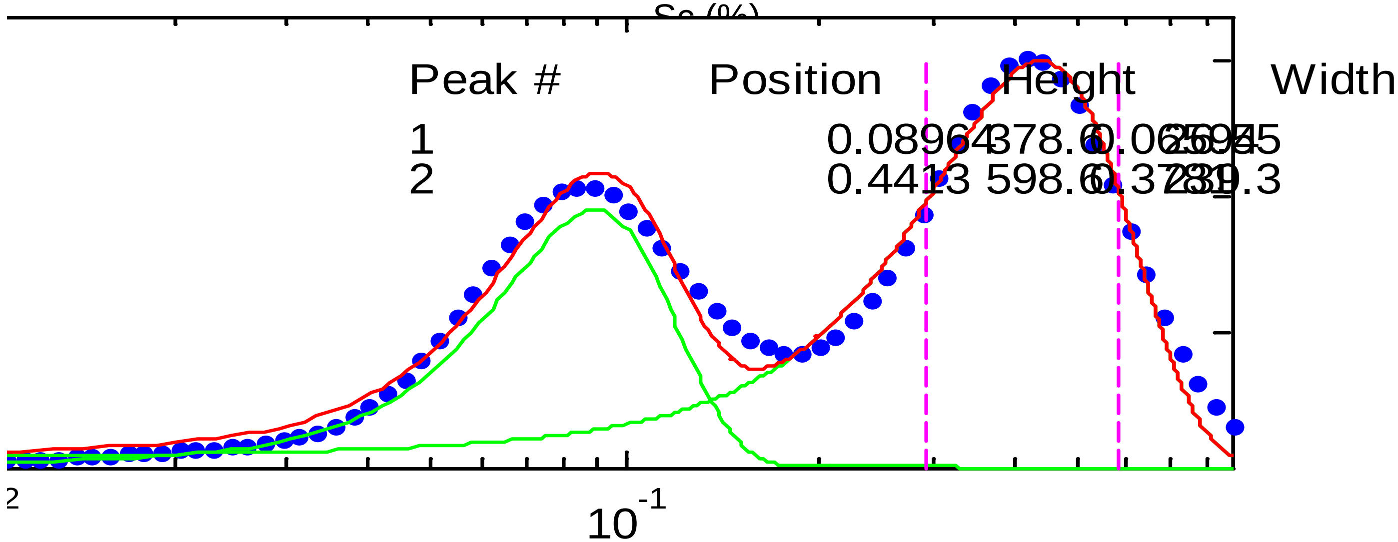
Since modes overlap
(i.e., there are no Hoppel minima with zero concentration),
it might seem reasonable to separate them using Peakfit.

But that analysis really applies to discrete data where the ideal
measurements would be delta functions.
This is unlikely for aerosol modes, which are mixtures,
i.e., cloud processed particles came from clouds with various S_{eff}.

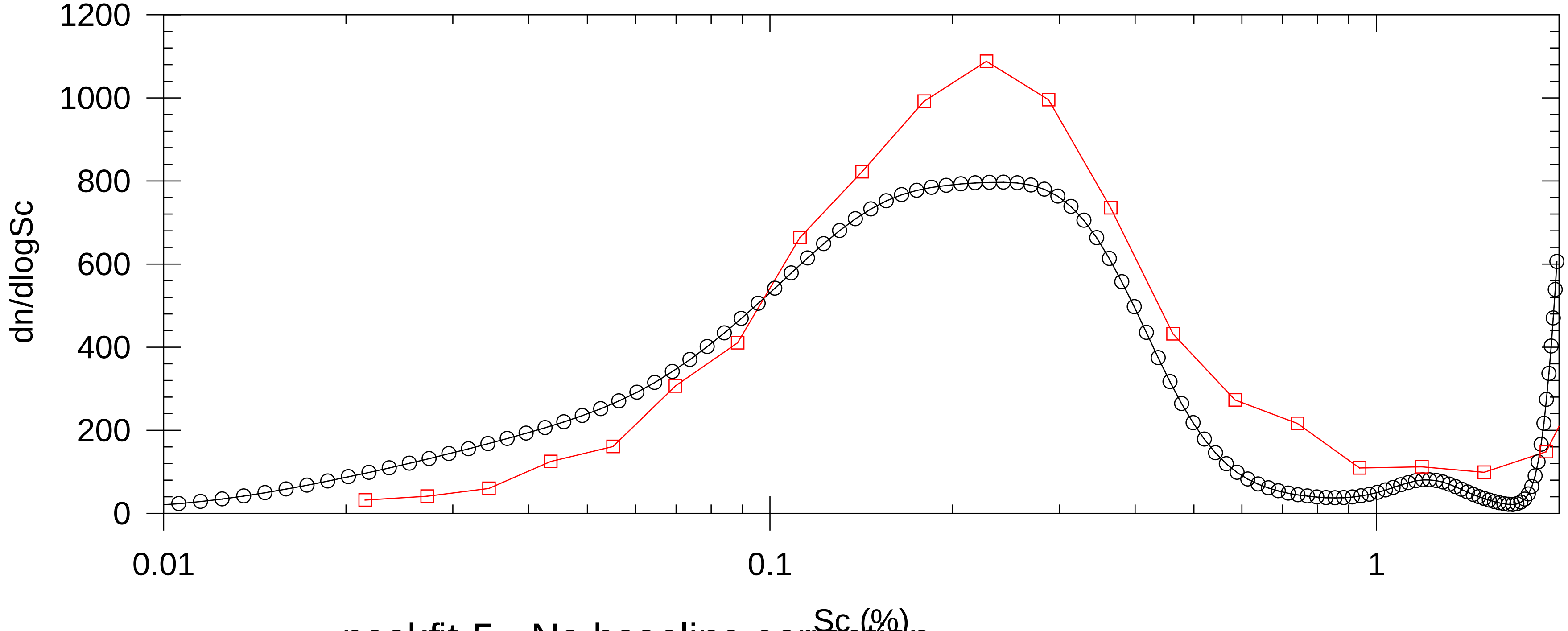
Overlapping modes are not just due to instrumental spreading
but are probably also fundamental properties of the real
aerosol.



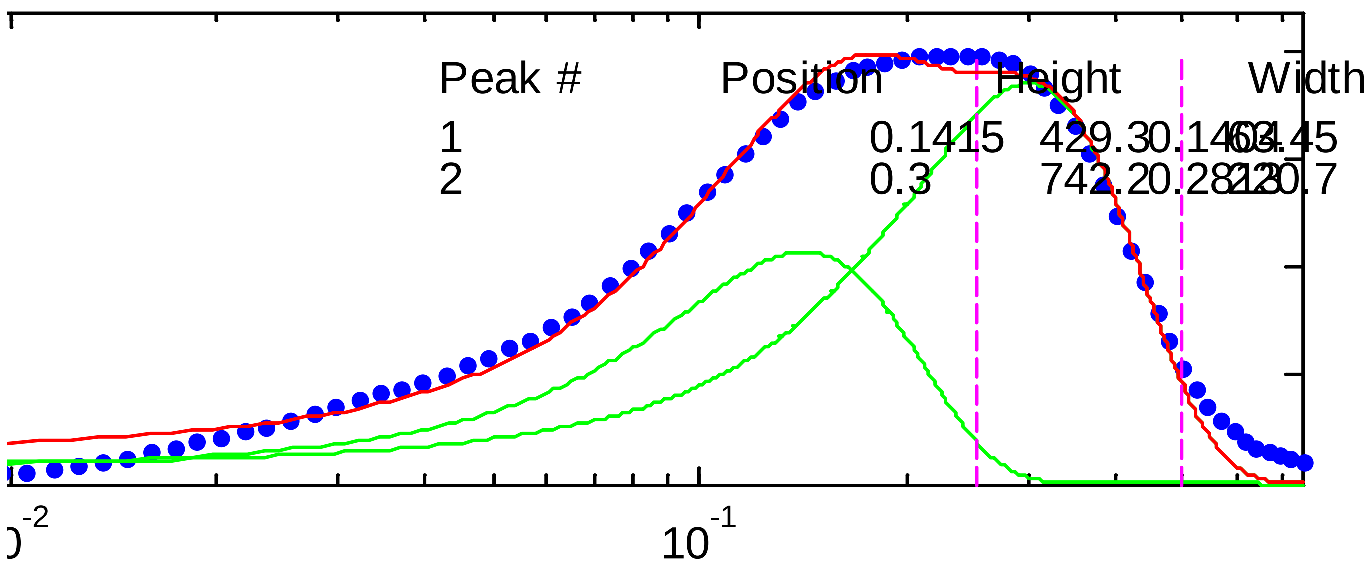
peakfit 5 No baseline correction



= 2 Shape = Gaussian Error = 3.552%



peakfit 5   No baseline correction



Persistent stratus cloud decks were thought to provide more opportunity for cloud processing and more cycles of evaporation and condensation that would enhance bimodal particle distributions.

However, column 6, Table indicates more bimodal distributions in the vicinity of the small cumulus clouds of ICE-T than either above or below clouds in MASE.

Conclusions:

1. DRI CCN spectrometers resolve Hoppel mins.
   Determine cloud S without composition
   With DMA can determine κ, hygroscopicity;
   property relevant for cloud interaction

2. Monomodal as well as bimodal—BL not so well mixed.

3. Even monomodal spectra may be cloud processed

4. Bimodal found in cumulus as well as stratus

5. Cloud processing alters the most important lowest S_c CCN

6. Cloud processing may make other IAEs