Aerosol Bimodality:
The Accumulation Mode is Mainly due to Cloud Processing

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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).

Surface aerosol modality measurements with an Scanning Mobility Particle Sizer SMPS at SGP during A-IOP of May 2003 were compared with remotely-sensed cloud fractions (CF) and cloud base altitudes (CBA). When these correlations were time adjusted to account for aerosol movement from clouds to surface, clouds were consistently implicated as the source of aerosol bimodality.

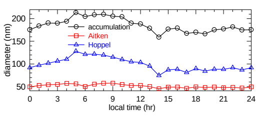

Figure 1. Mean diurnal trends for mean diameters of SMPS modes and Hoppel minima that separates the two modes.

Fig. 1 shows clear separations between the sizes of the aerosol modes, and the Hoppel minima between them.

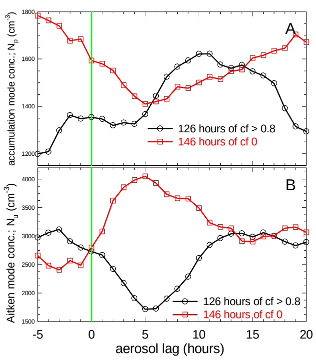

Figure 2. Surface particle concentrations within the two aerosol modes as a function of time after the specific ceilometer cloud fractions (CF) have been remotely observed at time zero (green line). Accumulation mode is the larger sized mode (>~ 100 nm) that is mostly a result of cloud processing, N_p. Aitken mode is the smaller particles (<~ 100 nm) that individually are unaffected by cloud processing; N_u. However, the Aitken mode is affected by the removal of some Aitken particles to the accumulation mode by cloud processing.

Fig. 2A shows that clouds convert Aitken particles to accumulation mode particles. Fig. 2B shows that absence of clouds reduces the accumulation mode and increases Aitken particles.

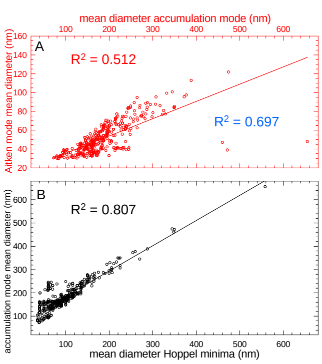

Figure 3. Relationships between hourly mean diameters of (a) unprocessed/Aitken and processed/accumulation, (b) Hoppel minima that separate the two modes and Aitken. Blue coefficient of determination exclude the 3 outliers. This reduces the number of hours from 428 to 425. All two-tailed probabilities (P2) of these regressions are < 10^{-8}.

High coefficients of determination in Fig. 3 indicate that the two aerosol modes are simultaneously altered. Thus, the same process altered the mean diameters of both modes; i.e., cloud processing. Numerous observations have linked cloud processing to aerosol bimodality.

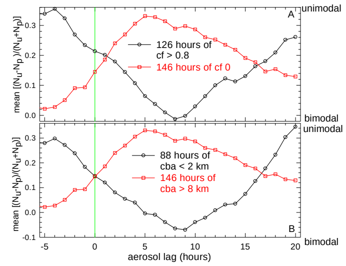

Figure 4. Mean normalized particle modalities (Aitken-accumulation concentrations divided by total particle concentration) as a function of time after cloud measurements within the extreme ranges noted in the legends for (a) ceilometer cloud fraction (CF) and (b) ceilometer cloud base altitude (CBA). Green lines mark the time of the cloud measurements. Lower normalized is bimodal.

Large CF (0.8) and low CBA (< 2 km) should have greater potentials toward particle bimodality, black circles. Smaller CF and higher CBA red squares should and do tend toward particle unimodality. In both panels of Fig. 4 the black data shifts toward lower normalized (more bimodal) between zero hour at green vertical lines, when the specified clouds were observed, to minimum values a few hours later.

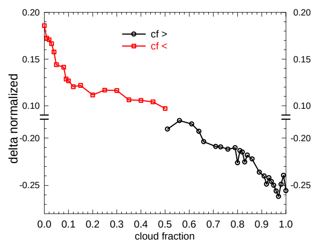

Figure 5. Differences between mean normalized at 0 hour from mean normalized at lag hour of minimal normalized (black) for all cases with CF greater than the abscissa designation; this indicates progressively greater bimodality for progressively higher CF. Red are the differences between mean normalized at 0 hours from mean normalized at lag hour of maximal normalized for all CF less than abscissa; this indicates progressively greater unimodality for progressively lower CF.

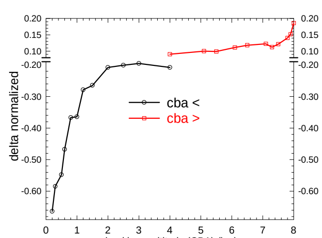

Figure 6. As Fig. 5 but CBA. Low CBA (black) is analogous to high CF as both have potential for bimodal aerosol whereas high CBA (red) is analogous to low CF as both have potential for unimodal aerosol.

Figures 5 and 6 push the CF and CBA extremes further than Figs. 4. Fig. 5 displays progressively higher (black data) and progressively lower (red data) CF to show that the differences between normalized at zero hour and at maxima (black data) and minima (red data) lags after zero hour are progressively greater for plots such as Fig. 4 for cumulatively higher CF (black) and cumulatively lower CF (red). For example, as higher CF than 0.8 are considered in figs. such as Fig. 4 minimal values of normalized are progressively lower. This means that the aerosol becomes progressively more bimodal for progressively greater CF (black data). On the other side of Fig. 5 as progressively lower CF are considered (red data) maximal normalized shows progressively greater differences from normalized at zero hour. This means that at progressively lower CF there are progressively greater shifts toward unimodal aerosol. For instance, in Fig. 4 normalized black goes from 0.2137 at zero hour (green line) to a minimum of -0.0125 at 8 hours after 0.8 CF is observed. This -0.2262 delta normalized is displayed in Fig. 5 at CF 0.8. Likewise, red in Fig. 4 shows 0.1452 at zero hour and achieves maximum normalized of 0.3310 at 5 hours. This +0.1858 delta normalized is plotted in Fig. 14A at 0 CF in Fig. 5.

Figure 6 shows analogously similar CBA trends for progressively lower CBA (black data) and progressively higher CBA (red data). Shifts toward bimodality for progressively lower CBA (black) are similar to Fig. 5 black for higher CF. Shifts toward unimodality for progressively higher CBA (red) are similar to Fig. 5 red for lower CF.

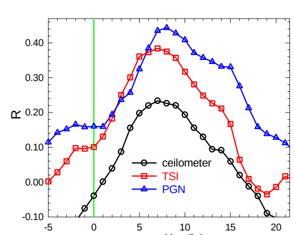

Figure 7. Correlation coefficients (R) between hour averages of SMPS modality with cloud fractions from ceilometer, total sky imager, and pyranometer gridded network against time lag of aerosol data from the cloud data. The ceilometer is an active sensor with minimal sky coverage; just straight overhead. TSI is a hemisphere dome and PGN covers an area equivalent to 80% of Oklahoma.

Inst	period	N	R	lag	P2
CEIL	21 days	385	0.25	7	3.81(-6)
TSI	21 days	274	0.38	7	1.00(-8)
PGN	21 days	244	0.44	8	1.00(-8)
CEIL	mean of daily averages	19.3	0.51	9.45	8.80(-2)
CEIL	mean of daily averages; 5-hr running mean	14.4	0.67	10.6	3.37(-2)
TSI	mean of daily averages	12.8	0.73	13.3	3.81(-2)
TSI	mean of daily averages; 5-hr running mean	12.8	0.79	9.8	4.35(-2)
PGN	mean of daily averages	11.5	0.82	10.8	2.43(-2)
PGN	mean of daily averages; 5-hr running mean	8.4	0.88	10.7	1.49(-2)

Table 1. First 3 rows characterize Fig. 7. N is number of hours, R is maximum correlation coefficient. Lag is the hour at maximum R. P2 is the two-tailed probability of the regression at maximum R. Destructive interference due to variations of the time lags for the various days during the project reduced R. Thus, plots of data within each day showed higher R values. Thus means of the daily averaged Rs are a factor of two higher than corresponding Rs from the entire 21-day project. 5-hour running means of the cloud data also make higher Rs. Rs are progressively higher for the instruments that have greater sky coverage.

Hoppel, W.A., J.W. Fitzgerald, and R.E. Larson (1985), Aerosol size distributions in air masses advecting off the East Coast of the United States, J. Geophys. Res., 90, 2365-2379.

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Instruments with greater sky coverage yield better representations of regional cloudiness that causes bimodal aerosol due to clouds.

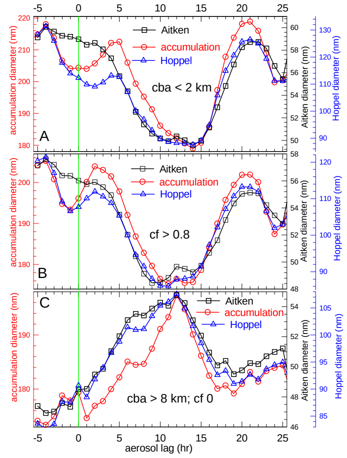

Figure 8. As Fig. 4 except showing mean diameters of the two modes and Hoppel minima in lagged response to ceilometer CBA and CF.

Similar responses of both modal diameters and Hoppel minima diameters to cloudiness.
Lower CBA or higher CF reduce the mean modal sizes in Fig. 8A and B whereas
higher CBA or lower CF make greater mean modal particle sizes in Fig. 8C.
Since cloud processing would move the larger of the Aitken particles to the accumulation mode this would reduce the mean size of the remaining yet unprocessed Aitken mode.
Promotion of these marginal CCN particles would occur to smaller sizes of the accumulation mode. This then reduces the mean size of the accumulation mode.
Apparently any tendency of the preexisting accumulation mode particles to grow larger by further cloud processing is less than the effect of newly promoted Aitken particles added to the low end of the accumulation mode.

The coordination of the two mode mean sizes in Figs. 3 and 8 and the opposite responses of the concentrations within the two modes to clouds in Fig. 2 verifies a single mechanism simultaneously affecting both modal sizes,
i.e., cloud processing.

Conclusion:

At SGP in May 2003 cloud processing made most accumulation mode particles while it simultaneously altered the Aitken mode.

It is possible that in many environments much of the accumulation mode is caused by cloud processing (Kerminen and Wexler, 1995).

Particle size distribution measurements and cloud measurements at SGP and other ARM sites could test this hypothesis.

Kerminen and Wexler (1995), Growth laws for atmospheric aerosol particles: an examination of the bimodality of the accumulation mode, Atmos. Envion. 29, 3263-32975.