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Indications of Cloud Processing from Cloud Remote Sensing and Surface Aerosol Size Spectra

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Three cloud processes cause bimodal particle size distributions by increasing the mass of dissolved material within cloud droplets, which upon evaporation are larger and thus size-separated from particles that did not produce 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. The larger size mode (> 100 nm) that has usually been cloud processed is the accumulation mode while the cloud unprocessed smaller mode (< 100 nm) is the Aitken mode.

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

Conversely clear skies were implicated as the cause of Aitken particles (~25-100 nm) that grow from nucleation mode particles (<~25 nm) due to photochemistry when clouds do not block the sun.

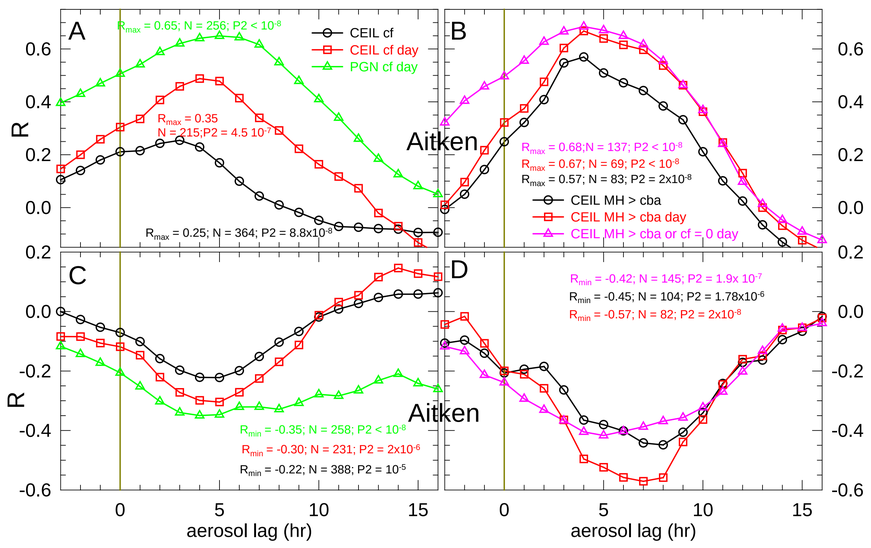

Figure. 1. **For Aitken particles 25-100 nm diameter.** A legend applies to C. B legend applies to D. Correlation coefficients, R, of ceilometer (CEIL) cloud fraction, cf, (**A**) and (**B**) with mean particle diameter, mpd, and (**C**) and (**D**) cf with concentration for aerosol lags of cf measurements at zero hour. R_{max} is maximum R, R_{min} is minimum R, N is number of hours, P2 is two-tailed probability. For (A) and (C) black is all CEIL data, red is CEIL daytime (07-19h), green is pyranometer gridded network (PGN). For (B) and (D) black is CEIL when mixing height, MH (Liu and Liang, 2010) is greater than cloud base altitude, cba . Red is these daytime cases. Pink is for MH > cba or cf = 0 for daytime only.

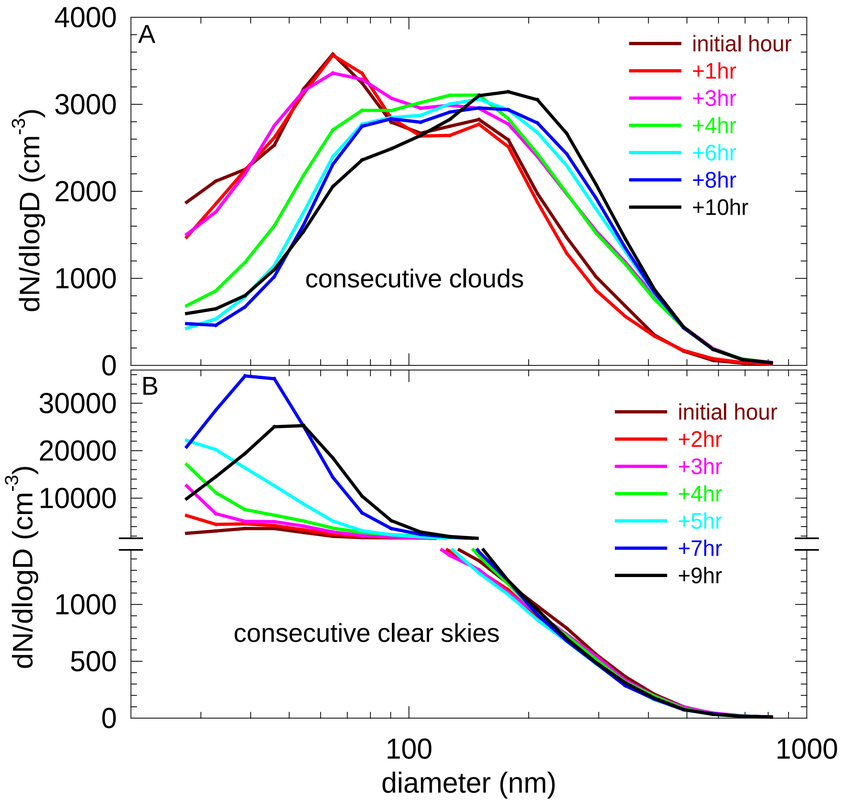

Fig. 4A shows how hours of consecutive cloudiness move Aitken mode particles to the accumulation mode by cloud processing. And further expand and enlarge the accumulation mode.

Fig. 4B shows the buildup of the Aitken mode after several consecutive hours of clear skies; as well as the enlargement of the Aitken mode. What there is of the accumulation mode erodes.

Figure 4. Mean particle size distributions at the initial onset of 7+ consecutive hours of **A**) ceilometer cf > 0.75 when MH > cba and the majority of hours are daytime; and **B**) daytime ceilometer cf = 0.. Hours following onset are denoted in legend.

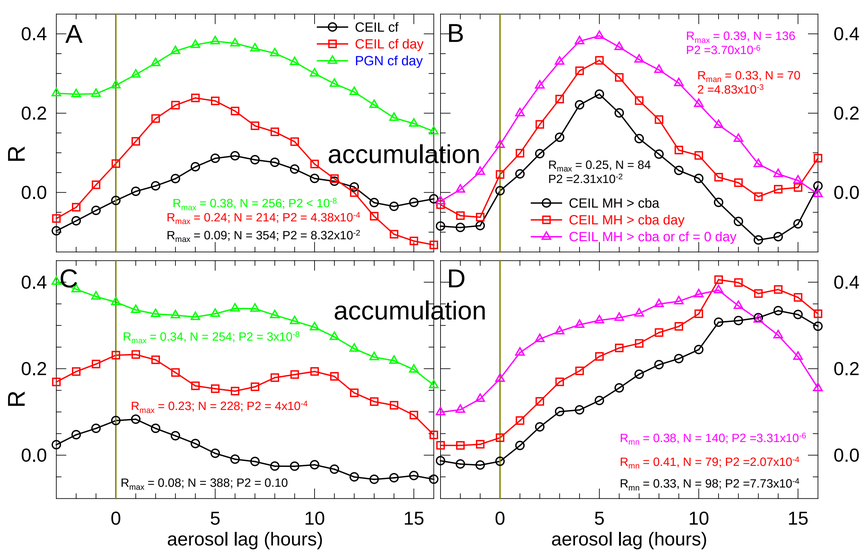

Figure. 2. As Fig. A but only for **accumulation particles (diameter > 100 nm)**.

Figs. 5 (black) and 6A chart the initial increase of both particle sizes to clouds within the mixed layer, especially when overcast (Fig. 5 red, Fig. 6B). As in Figs. 1 and 2 concentrations go in opposite directions; accumulation particles (N_L) increase while Aitkens (N_s) decrease.

When cloud base is above the mixed layer or there are no clouds (Fig. 5 green, Fig. 6C) cloud processing cannot be observed at the surface so photochemistry dominates. Thus, both particle sizes (smpd and Lmpd) and N_L decrease while N_s increase, especially when only no cloud cases (Fig. 5 blue, Fig. 6D) are considered.

A and B panels of Figs. 1 and 2 indicate that greater cloudiness causes larger Aitken and accumulation mode particles. Aerosol response lags cloudiness mainly due to transport time to the surface. Fig. 1C and D show reductions of Aitken concentrations in response to cloudiness. This is ostensibly because clouds block solar radiation that causes photochemical production of small particles that grow into the Aitken range. Thus clear skies decrease Aitken sizes while increasing Aitken concentrations. Fig. 2C and D indicate greater accumulation concentrations with cloudiness. Thus, cloud processing increases accumulation sizes and concentrations.

Black traces in all panels of Figs. 1 and 2 for all hours of the day are much lower than red data for daytime hours because at night cloud base is more often decoupled from the surface. PGN (green) shows higher Rs because of its greater sky coverage than CEIL. Red and pink of panels C and D show even higher Rs for conditions when cloud base is within the mixed layer.

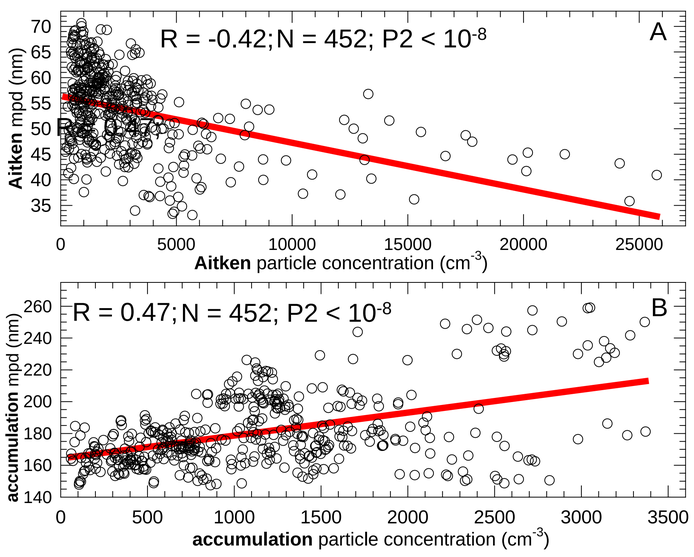

Figure 3. Concentration-mean particle diameter (mpd) relationships for Aitken and accumulation particles.

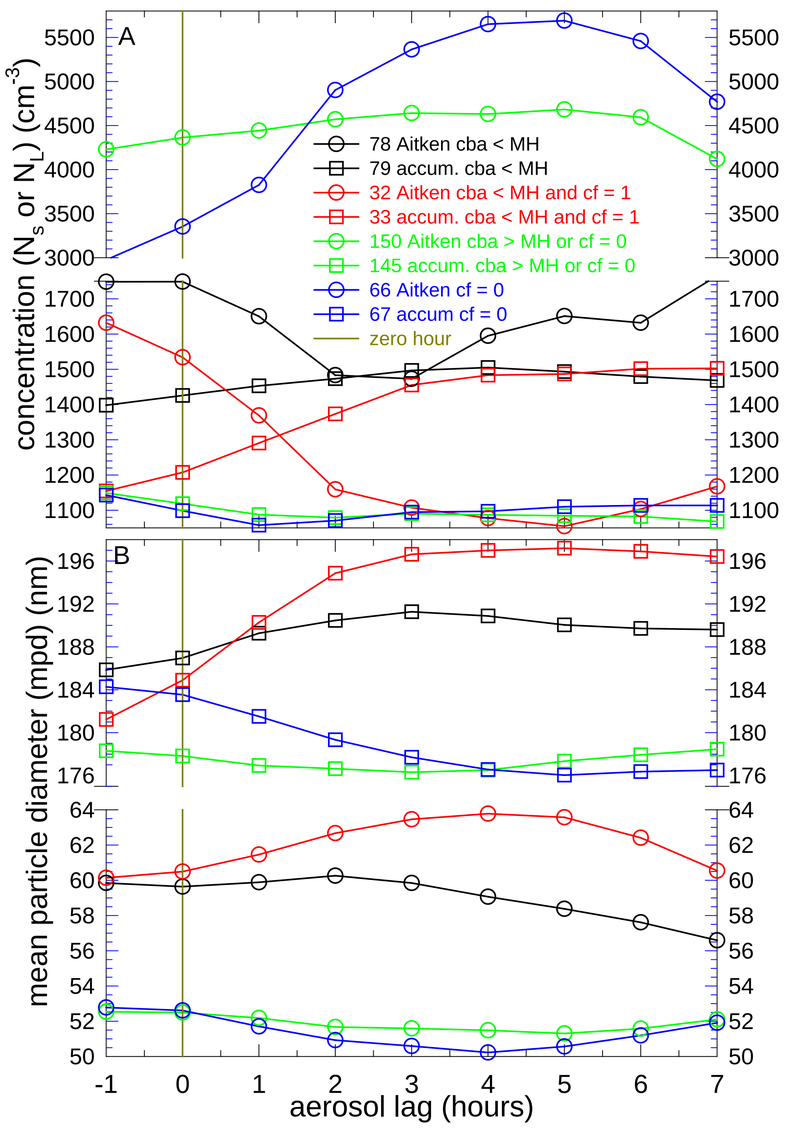

Figure 5. Mean response of Aitken and accumulation concentrations (**A**) and mean particle diameters (**B**) to cases of various relationships among mixing height (MH), ceilometer cloud base altitude (cba) and cloud fraction (cf). Effects of cloud processing at the surface should be most obvious when cba is within (<) MH, especially for complete overcast (cf = 1). Effects of photochemical particle production should be most obvious when cba > MH or cf = 0, especially for only cf = 0 cases. Legend A applies to B. Legend shows number of hour cases.

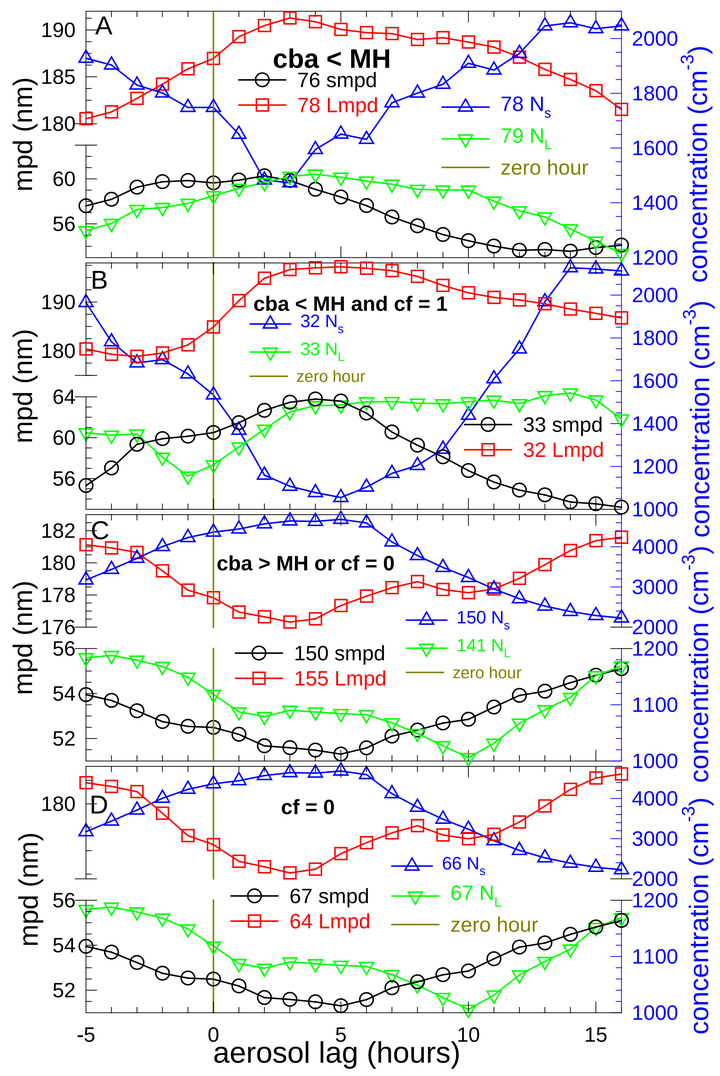

Figure 6. Same data as Fig. 5. (**A**) Cloud processing effects. (**B**) More intense cloud processing effects. (**C**) Photochemical effects. (**D**) More intense photochemical effects. Smpd is Aitken mpd, Lmpd is accumulation mpd. N_s is Aitken concentration. N_L is accumulation concentration. Legend shows number of hour cases.

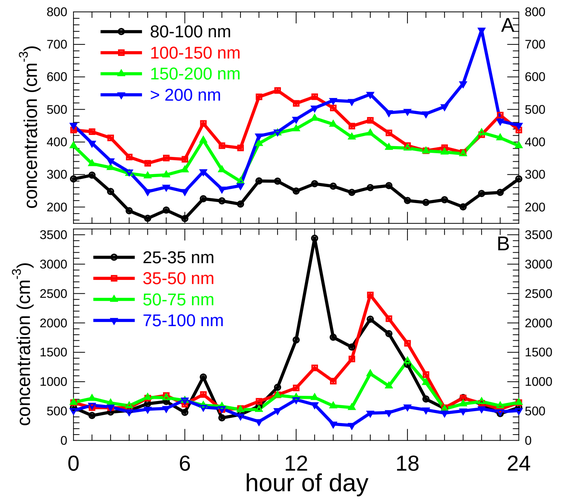

Figure 7 (**A**) Diurnals of large particle concentrations (mostly accumulation) when CEIL cf 5-hour running mean 4 hours prior to aerosol exceeds mean cf, 0.225. (**B**) Diurnals of small particle concentrations, Aitken, when CEIL cf = 0.

Summary

Positive correlations of Aitken (smpd) and accumulation (Lmpd) sizes and accumulation concentrations (N_L) with cloud fraction (cf) and negative correlations of Aitken concentrations (N_S) with cf (Figs. 1-3) are consistent with cloud processing and photochemical production of nucleation mode particles (~< 25 nm). Since cloud processing increases mpd it reduces N_S and increases N_L. Photochemistry initially decreases smpd and increases N_S. Photochemistry can occur at many altitudes.

Effects of cloud processing at the surface are best observed when cloud base altitude (cba) is below the mixing height (MH), which is mostly during daylight hours. Photochemistry effects are prominent when cloud processing effects are minimal, i.e., during daylight when cba exceeds MH or especially when cf = 0.

Figs. 4 and 7 show the evolution of the two modes in response to cloudiness and uncloudiness.

Figs. 5 and 6 show how mpds and concentrations respond to four categories of cba, MH and cf.

Conclusions:

1. Effects of cloud processing and photochemistry were readily observed at the SGP surface in May 2003.

2. Cloud processing made most accumulation mode particles. This may be so in many seasons and in many environments (Kerminen and Wexler, 1995).

3. Conversion of Aitken particles to accumulation particles in the presence of high accumulation mode particle concentrations indicates that CCN are not restricted to the accumulation mode (diameter > 100 nm).

4. Furthermore, the best way to produce accumulation mode particles is by cloud nucleation, which readily occurred at SGP in May, 2003.

Kerminen and Wexler (1995), Growth laws for atmospheric aerosol particles: an examination of the bimodality of the accumulation mode**, Atmos. Envion. 29**, 3263-32975.
Liu and Liang (2010), Observed diurnal cycle climatology of planetary boundary layer height, **J. Climate, 53**, 5790-5809.