

Recent Particle Formation and Aerosol Variability Near Southern Ocean Low Clouds

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Additional Supporting Information (Files uploaded separately)

Captions for Movie S1

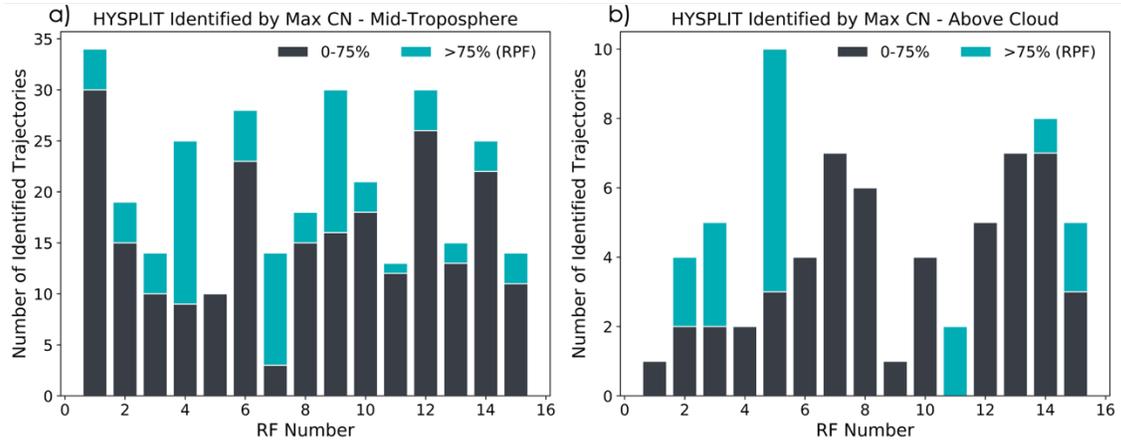


Figure S1. Number of RPF (blue) vs non-RPF identified HYSPLIT trajectories by research flight for trajectories initiated in the (a) mid-troposphere and (b) above cloud.

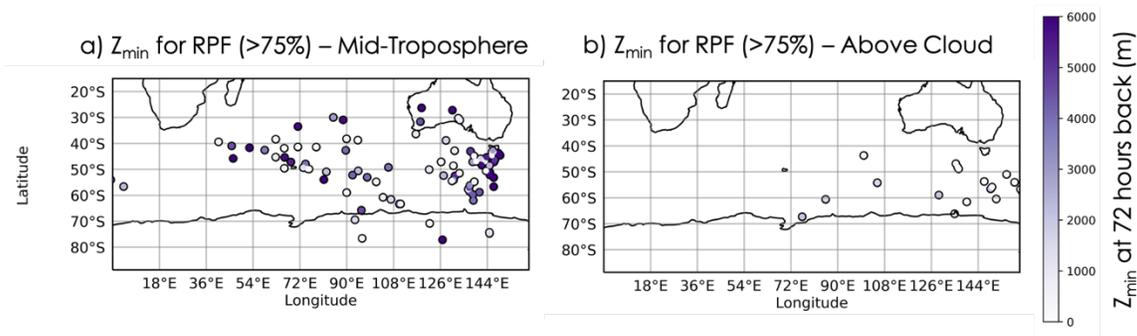


Figure S2. Location and altitude of minimum height that occurs over full 72 hours for RPF identified trajectories in (a) mid-troposphere and (b) above cloud.

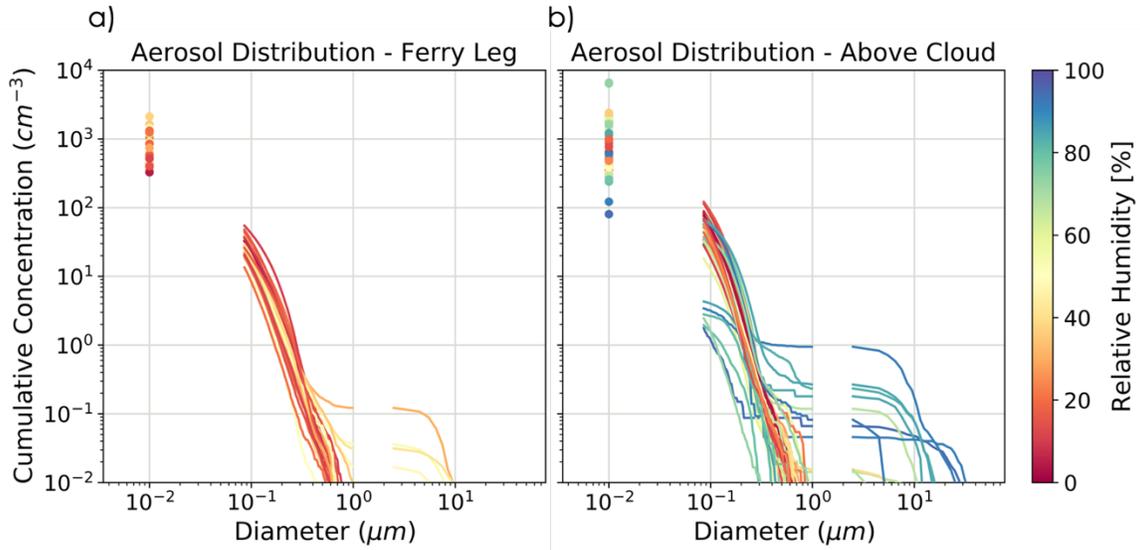


Figure S3. Median cumulative size distributions for each level leg in the mid-troposphere (a) and above cloud (b) sampled during the campaign. Each line is colored by the median relative humidity to indicate where near cloud contamination may be occurring. Note that concentrations are not adjusted for standard temperature and pressure.

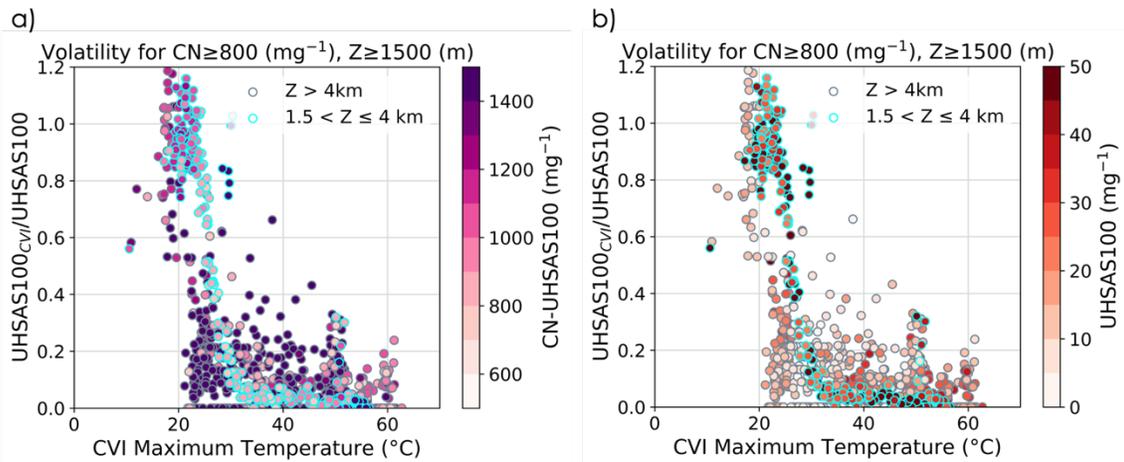


Figure S4. Volatility curves from CVI analysis as in Figure 7 but for the small number of accumulation mode particles captured in these Aitken-dominated samples. Accumulation mode volatilization is presented as ratio between UHSAS100 and UHSAS100_{CVI} versus the maximum temperature of the CVI instrument. Points are shown for $CN \geq 800$ mg^{-1} and limited to free troposphere samples ($Z \geq 1.5$ km). Outline colors denote altitude of sample: mid-troposphere (gray) and above cloud (blue). Points are colored to estimate the number of particles in the (a, CN-UHSAS100) Aitken mode (generally more in the mid-troposphere) and (b, UHSAS100) accumulation mode. Note that accumulation number in these Aitken-

dominated samples are rarely more than 30 mg^{-1} in mid-troposphere and increase only to $\sim 50 \text{ mg}^{-1}$ above cloud.

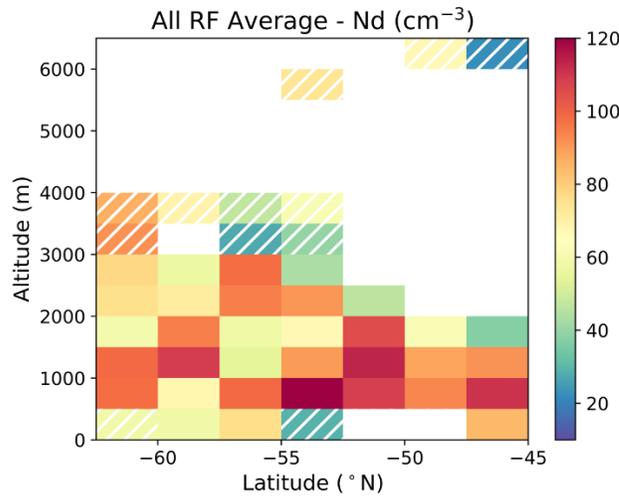


Figure S5. All flight average composite of binned flight medians ($500 \text{ m} \times 2.5^{\circ}$ boxes) for cloud droplet number concentration as in Figure 8d but using volume units (cm^{-3}) instead of standard temperature and pressure corrected units (mg^{-1}). Bins where 2 or less flights sampled are hatched to indicate reliability of sampling.

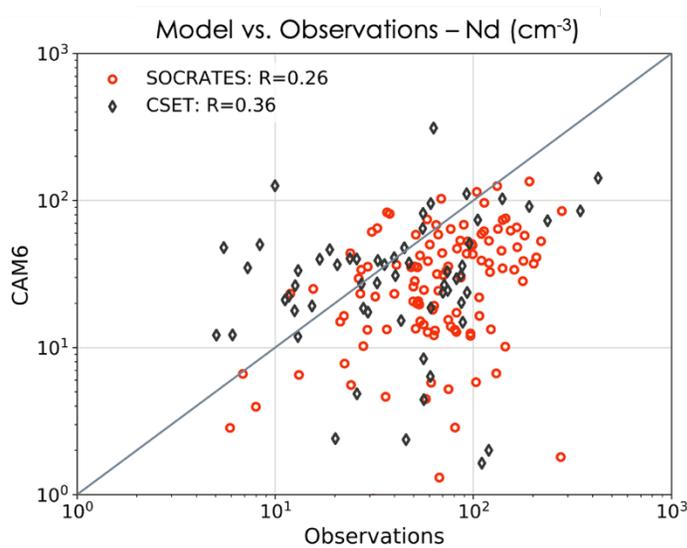


Figure S6 Comparison of N_d between CAM6 and observations from all CSET (gray) and SOCRATES (orange) flights. Each point used in the scatter represents a $500 \text{ m} \times 2.5^{\circ}$ bin mean value for a given flight during the campaign. A reference 1:1 line is included along with correlation coefficients computed for the linear relationship between CAM6 and observations.

CAM6 over produces precipitation-depleted clouds ($N_d \leq 10 \text{ cm}^{-3}$) relative to SOCRATES observations and are not collocated with actual observations for either CSET or SOCRATES.

SOCRATES CAM6 Aerosol Composition

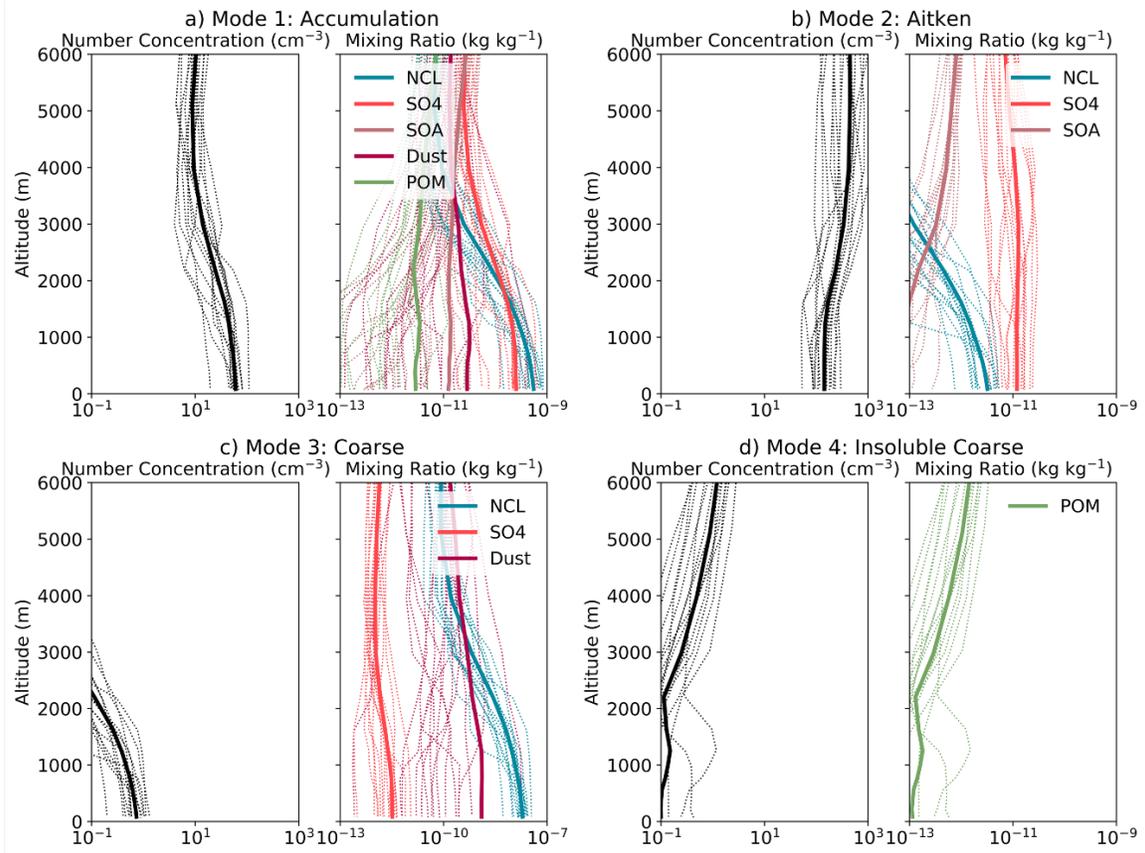


Figure S7 Aerosol Number Concentrations (left) and Mixing Ratios (right) for nudged SOCRATES CAM6 Aerosol Modes: Accumulation (a), Aitken (b), Coarse (c), and Insoluble Coarse (d). All levels and extent of model data extracted along the Latitude-Longitude flight path is

included to understand overall model behavior. Dashed lines are individual flight means and solid line is campaign average tendency.

CSET CAM6 Aerosol Composition

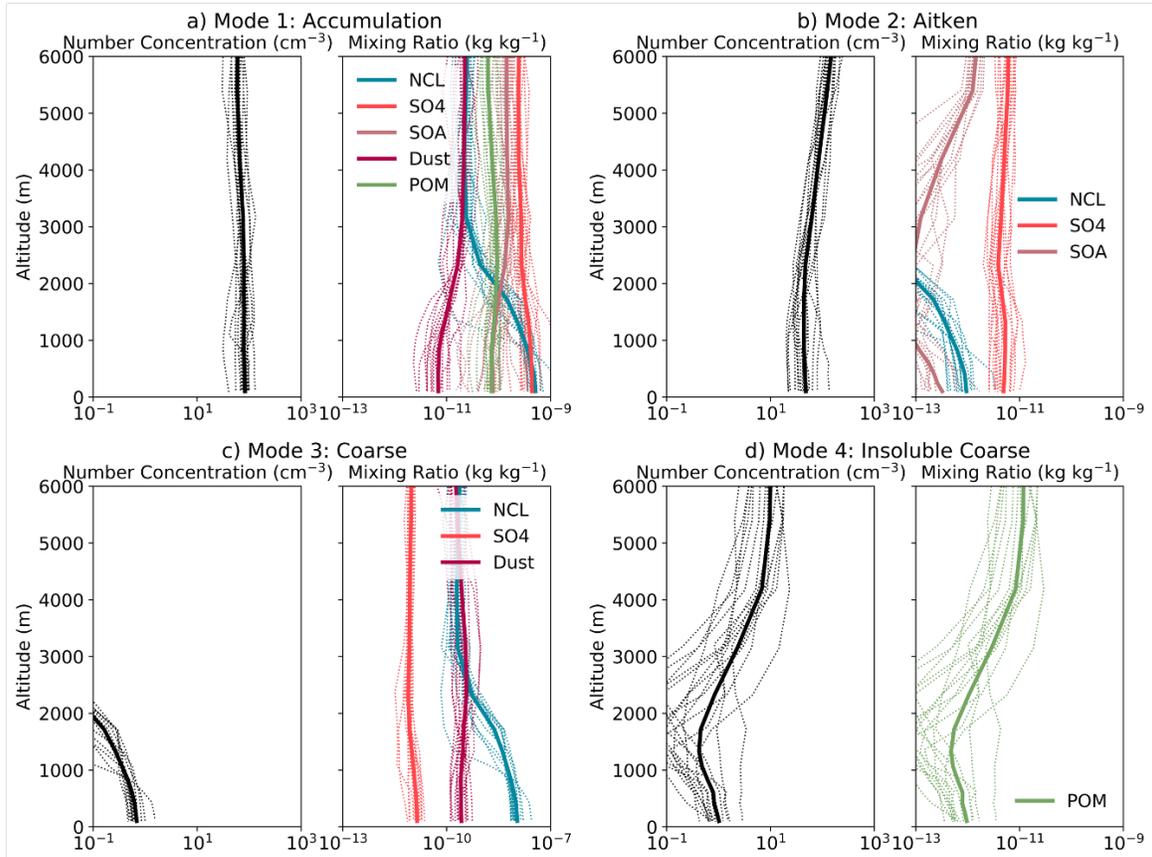


Figure S8 As in Figure S5 but for nudged CSET CAM6 simulations.

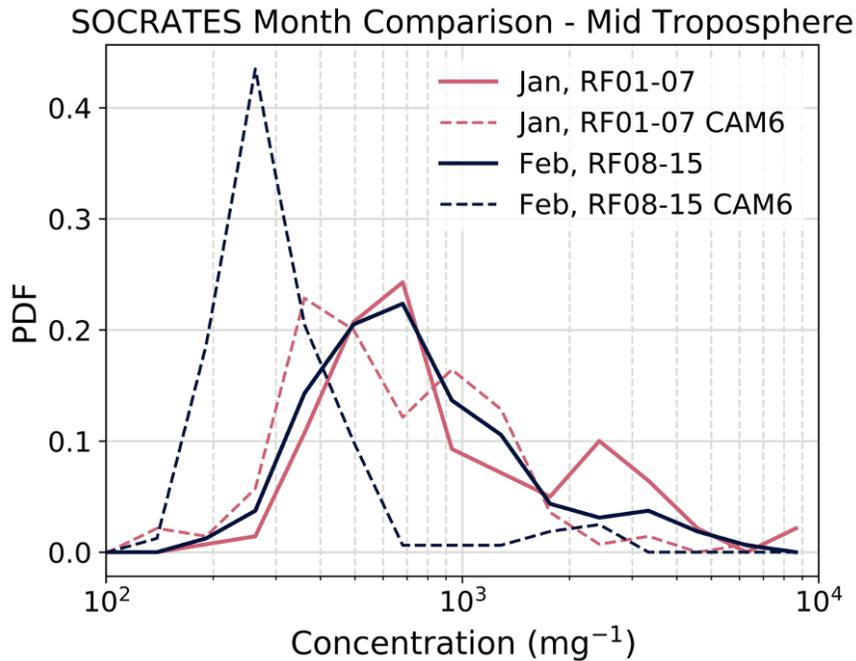


Figure S9 Observed CN number concentration in the mid-troposphere for January (RF01-07) and February (RF08-15) samples are found to have similar PDFs, suggesting statistically consistent CN concentrations across these months. CAM6 matched CN has distinctly different PDFs for January and February, with February producing much lower number concentrations than January and far too low relative to observations. Both CAM6 month PDFs under produce high concentrations of aerosol (CN ≥ 2000 mg⁻¹) relative to observations.

Movie S1. Synoptic scale patterns influencing mid-tropospheric RPF identified air masses in the 72-hour period before sampling by RF07 (black line from Tasmania). ERA5 reanalysis maps include 700 hPa vertical velocity (colors) with a 700 hPa geopotential height contour of 2.9 km for reference (black contour). RPF trajectories (gray lines) with air mass locations (circles) colored by their altitude (white to purple, as in the ascent profiles in Figure 5a). Ascent of the first set of trajectories at ~60-hr occurs off the tip of Africa while ascent of the ~36-hr trajectories occurs off the coast of Antarctica, both driven by the advance of a warm-conveyor belt towards the south east (i.e. along the height contour).