SIO 217D: Atmospheric and Climate Sciences IV

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As part of the First Aerosol Characterization Experiment (ACE-1) campaign, airborne measurements of the marine aerosol size distribution were conducted off the Tasmanian coast via a radially-classified aerosol detector. On 6 November 1995, the instrument measured fine (nucleation, Aitken, accumulation) mode atmospheric particles spanning 0.005 to 0.15 μm diameter. These small particle sizes suggests their presence is likely the result of chemical processes, e.g., photochemical oxidation of sulfur-containing compounds that lead to the formation of sulfate particles. The average of the first 10 distributions observed possesses a single mode at a diameter slightly larger than 0.01 μm. Back trajectories converging upon the sampling location and time suggest the sampled air mass to be of maritime polar (mP) origin, mostly from the coastal regions of Wilkes Land, East Antarctica. Large-scale descent prevails during this period, indicating significant influence from a high pressure zone, or post-frontal subsidence during the $\sim$4 day period shown.

Stratiform clouds are a persistent feature over the marine boundary layer (MBL) of the Southern Ocean, and moreover, are common under conditions of weak synoptic descent. At this level of the atmosphere (1-2 km above mean sea level), droplets comprising a boundary layer cloud scavenge interstitial aerosol through particle-droplet collisions. To determine how the aerosol distribution would be influenced by such a process, here the characteristic lifetime of such atmospheric particles is computed as a function of size. The evolution of a population of aerosol particles via interstitial scavenging by cloud droplets in the MBL is described by,

$$\frac{\partial n(D_p,t)}{\partial t} = -n(D_p,t)\int_0^\infty K(D_p,x)n(x,t)dx, \tag{1}$$

as discussed in *Seinfeld and Pandis*, 2006. In equation (1), $n(D_p,t)$ is the aerosol particle size distribution, $n_d(x,t)$ is the cloud droplet size distribution, x being the cloud droplet diameter, and $K(D_p,x)$ is a collection coefficient describing the efficiency by which cloud droplets of size x scavenge particles with diameter D_p. The integral in (1) defines the scavenging coefficient Λ, whose evaluation is the foremost task in solving for aerosol lifetimes as a function of particle size in a boundary layer cloud. If time dependence of the cloud droplet distribution is negligible (e.g., collision-coalescence droplet growth does not occur, droplet sizes unaffected by scavenging, etc.), then (1) admits a simple exponentially decaying solution in time, with a scavenging coefficient solely dependent on particle size.

Harvey et al. [1991] find a high degree of similarity in fine mode aerosol composition and distribution made from Ross Island, Antarctica and observations from lower latitudes of the southwest Pacific. Aerosol scavenging calculations are therefore performed here with parameters typical of MBL clouds from the Antarctic coast, for a range of cloud droplet radii between 7-15 μm ($\sim$9-13 μm observed in coastal stratus over the Ross Sea [*Saxena and Ruggiero*, 1990]). Assuming that the cloudy MBL consists of a uniformly distributed population of droplets of fixed radius, with a number concentration $N_0 = 100$ cm^{-3}, the integral in (1) reduces to

$$\int_0^\infty K(D_p,x)n(x)dx = N_0 K(D_p, 22\mu m). \tag{2}$$

Aerosol lifetimes can then be computed, with the following expression

$$K = \frac{2kT}{3\mu}\frac{22\mu m}{D_p}, \tag{3}$$

valid in the limit of large cloud droplets compared to the size of particles scavenged [*Seinfeld and Pandis*, 2006]. Results indicate that for fixed cloud particle radius, small aerosol particles tend to be scavenged more quickly, therefore having a shorter lifetime than larger ones. Put another way, for a given aerosol particle size, larger cloud droplets are more efficient particle scavengers. If interstitial scavenging were the only process influencing the aerosol particle size distribution, the temporal dependence of the average particle size distribution should reveal the most rapid decrease at smaller particle sizes. This temporal behavior, however, is not manifested via further averaging of subsequently collected size distributions, indicating that these samples were in all likelihood not collected within a MBL stratiform cloud. Within half of a day, a signature of deposition is most evident, which is perhaps reasonable given the superimposed descending large-scale motion. By one day, however, a signature of cloud processing becomes apparent, with the formation of a second mode within the accumulation regime.

References

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