The Spatial Distribution and Size Evolution of Particles in Asian Outflow: The Significance of Primary and Secondary Aerosols during ACE-Asia and TRACE-P

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Abstract

During the ACE-Asia and TRACE-P field campaigns aircraft measurements over the Yellow Sea, East China Sea and Sea of Japan revealed widespread secondary aerosol formation in the marine boundary layer. Similar observations were made throughout the day at the Gosan surface site on Jeju Island South Korea and aboard the American research vessel the R/V Ron Brown. Intercomparisons between the surface aerosol platforms and the airborne platforms show excellent agreement. Two post-frontal airmasses were characterized by concentrations of 3 - 12 nm particles as high as 40,000 cm^{-3}, and SO_2 concentrations of 2 - 12 ppbv. Aerosol surface area in the marine boundary layer associated with regional air pollution and in one case, mineral dust, was in excess of 300 μm^2 cm^{-3} and as high as 1200 μm^2 cm^{-3}. Thermodynamic profiles of the regional airmass suggest that steep gradients of aerosol surface area, relative humidity and temperature associated with elevated levels of SO_2 created microphysical environments where new particle production is favoured.

Although nucleation events were not observed directly, evidence for recently formed secondary aerosols were detected up to 48 hours after leaving the Asian landmass and were associated with SO_2 concentrations in excess of 1 ppbv. Molar ratios of $[NH_4^+]$:$(2[SO_4^{2-}] + [NO_3^-])$ in excess of 0.68 and volatility analysis showing partial neutralization of the nucleation and accumulation mode aerosols suggest a ternary nucleation mechanism involving H_2SO_4, H_2O, and NH_3. Growth rates of nucleation mode aerosols of 2.4 and 1.7 nm hr^{-1} were observed over a broad spatial extent during two consecutive flights. Based on the nucleation mode growth rates, the flux rate of vapour to the full size distribution suggests condensation rates of 2.4 +/- 1.2 x 10^6 molecules cm^{-3} s^{-1}. Calculations of the coagulation rate for the nucleation mode particles suggest that most may be scavenged by the primary aerosol in less than 72 hours over the coastal waters of Asia before being transport out toward the Pacific Ocean.