Geophysical Research Abstracts, Vol. 7, 09986, 2005
SRef-ID: 1607-7962/gra/EGU05-A-09986

Cloud condensation nuclei over the Eastern Pacific Ocean: A case study

G. Roberts (1), G. Mauger (1), O. Lariviere (1), E. Wilcox (2), V. Ramanathan (1)

(1) Scripps Institution of Oceanography, California, USA (greg@fiji.ucsd.edu / +1 858-822-1662), (2) Geophysical Fluid Dynamics Laboratory, New Jersey, USA.

Introduction:

The Cloud Indirect Forcing Experiment (CIFEX), in April 2004, focused on the measurement of aerosol and cloud properties to assess the impact of long-range transport on aerosol-cloud interactions. The experiment was conducted in Northern California, where we expected to intercept plumes of pollution and dust transported from the Asian continent across the Pacific Ocean. During the CIFEX experiment, we observed a variety of aerosol layers in vertical and horizontal profiles, ranging from aged aerosols associated with two major Asian dust storms to ultrafine particles from recent nucleation events. Aerosols were found in thin stratified layers ranging from altitudes of 1000 to 7000 m and thicknesses from 100 to 1000 m. Aerosol size distributions indicated cloud processed aerosol in some of the layers, while other layers were clearly due to secondary particle formation with over 90% of the number concentration smaller than 20 nm diameter. In this analysis, we focus on flights where we observed a nucleation event and transition in aerosol properties that have a large effect on cloud condensation nuclei (CCN). A comparison of the aerosol size distributions to the measured CCN concentration provides insight to the chemical evolution of the aerosols and their potential effect on aerosol/cloud interactions during long-range transport.

Instrumentation:

Airborne measurements included total number concentration, aerosol size distributions, aerosol back scattering, absorbing carbon and CCN. The CCN instruments were of different designs: the streamwise continuous-flow device operated at 0.2% supersaturation (S) at 1 sample per second (Roberts and Nenes, 2005), while the static thermal-

gradient instrument cycled through 0.2%, 0.4%, and 0.6% S once a minute. This was the first airborne comparison of streamwise and static CCN chambers, which highlighted the importance of fast measurements for aircraft experiments. Also, we used the Chemical Weather Forecast System (CFORS; Uno, 2003), a chemical transport model, to plan flight missions.

Results and discussion:

During a particular flight on 14 April 2004, particle concentrations at the beginning of the layer averaged 630 cm^{-3}; CCN concentrations (at 2% S) were also high at 350 cm^{-3}. The remote free tropospheric aerosol concentrations averaged 250 cm^{-3}. The aerosol concentrations suddenly doubled, then gradually increased to 2300 cm^{-3} while CCN concentrations (at 2% S) tapered to nearly zero. During this leg, we have identified three sections of interest based on different aerosol size distributions: i. accumulation mode; ii. bimodal distribution; and iii. ultra-fine aerosols. The accumulation mode aerosol at the beginning of the leg was characterized by a large median diameter near 150 mm; no small particles indicated aged aerosol. The lack of a Hoppel minimum suggests the parcel had not undergone cloud processing, in spite of an activation ratio (N_{CCN}/N_{CN}) of 0.5 at 0.2% S. The accumulation mode quickly transformed to a bi-modal distribution with a large ultra-fine component and fewer accumulation mode aerosols as the relative humidity increased and sparse cloud cover changed to a precipitating stratus deck. The large number of particles < 20 nm implies a recent nucleation event. Similar peak diameters between the distributions suggest mixing between two airmasses or nucleation in a diluted aged-aerosol plume. By the end of the leg, the accumulation mode completely disappeared and the particles became too small to serve as CCN (at 0.2% S); yet, they could grow to CCN size in a few days (Raes et al., 2000).

Comparing size distributions to measured CCN concentrations provides insight on the chemistry and evolution of these aerosols. The limiting cases of an ammonium bisulfate (NH_4HSO_4) and insoluble aerosol activate at 87 and 1000 nm at 0.2% S, respectively. Our measurements show a critical diameter of ca. 150 nm – an internally-mixed equivalent of a 20% NH_4HSO_4 core (by mass) with an 80% insoluble core. This estimate does not include surface effects or slightly soluble compounds. Aerosols originating from natural marine sources activate close to the NH_4HSO_4 limit (i.e., VanReken et al., 2003); hence, the reduced efficiency by which the larger aerosols activate points to an insoluble or hydrophobic core (i.e., biomass burning or dust) that has been aged by deposition of a soluble salt (i.e., SO_4).

This flight leg also provided an opportunity to compare the performance of the static and streamwise CCN instruments. While both instruments showed the decreasing

CCN concentration, the streamwise CCN instrument fully captured the changes in aerosol properties on a time scale necessary for airborne measurements.

Summary:

An airborne experiment over the Eastern Pacific Ocean measured multiple aerosol layers and highlighted the influence of aerosol properties on CCN concentrations. The streamwise and static CCN chambers show consistent results; nevertheless, the streamwise CCN instrument captures the effects of rapidly changing aerosol size distributions, emphasizing the importance of fast measurements for aircraft studies. The CFORS chemical transport model shows that dust and anthropogenic emissions stretch from the Asia across the Pacific Ocean. Vertical profiles near Northern California's coast reveal multiple layers of newly formed and aged aerosol. Our measurements show that new particle formation, characterized by high concentrations of particles $<$ 20 nm diameter, frequently occurs in thin stratified layers between 1000 and 7000 m altitude. However, due to their small size, the ultra-fine aerosols neither effectively scatter light nor immediately serve as CCN (at 0.2% S). In contract, aged dust/biomass burning aerosols scatter light efficiently and serve as CCN (N_{CCN}/N_{CN} = of 0.5; 0.2% S); however, in some cases, they have been transported across the Pacific without undergoing cloud processing. CCN measurements indicate that a significant mass fraction of aged aerosols was insoluble or hydrophobic as their critical diameter is ca. 150 nm (compared to 87 nm for an NH_4HSO_4 aerosol). Unless these layers are mixed into the boundary layer, these results suggest that such aerosol layers contribute more to the direct effect than the indirect effect during long-range transport.

References

Raes, F., T. Bates, F. McGovern, and M. VanLiedekerke (2000). The 2nd Aerosol Characterization Experiment (ACE-2): general overview and main results, Tellus, 52: 111-125.

Roberts, G., and A. Nenes (2005). A continuous-flow streamwise thermal-gradient CCNchamber for airborne measurements, Aerosol Science and Technology: in press.

Uno, I. (2003). Regional chemical weather forecasting system CFORS: Model descriptions and analysis of surface observations at Japanese island stations during the ACE-Asia experiment, J. Geophys. Res., 30: Art no. 1640.

VanReken, T.M., T.A. Rissman, G.C. Roberts, V. Varutbangkul, H.H. Jonsson, R.C. Flagan, and J.H. Seinfeld (2003). Toward aerosol/cloud condensation nuclei (CCN) closure during CRYSTAL-FACE, J. Geophys. Res., 108: Art. No. 4633.