

Physicochemical transition of long-range transport and its effect on cloud condensation nuclei over the Eastern Pacific Ocean



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Introduction:

The **Intercontinental Chemical Transport Experiment – Phase B (INTEX-B)**, which occurred in April 2006, focused on the transport and transformation of gases and aerosols on transcontinental/intercontinental scales to assess their impact on air quality and climate. The experiment was conducted in Seattle, WA (Figure 1), where we expected to intercept plumes of pollution and dust transported from the Asian continent across the Pacific Ocean.

During this experiment, we observed variety of aerosol layers during vertical and horizontal profiles, ranging from aged aerosols from major Asian dust storms to ultrafine particles from recent nucleation events. Aerosol layers observed during the flights were found in thin stratified layers ranging from altitudes of 1000 meters to 7000 meters and thicknesses from 100 to 1000 meters. Size resolved chemical measurements indicate cloud processed aerosol in the boundary layer and both aged and ultrafine layers aloft. Aerosols undergoing cloud processing activate close to the $(\text{NH}_4)_2\text{SO}_4$ limit (activation index near unity); while those with anthropogenic origins show lower activation indices which points to an insoluble or hydrophobic core (i.e., biomass burning or dust) that has been aged by deposition of a water-soluble material. A comparison of the aerosol size distributions and chemistry 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.

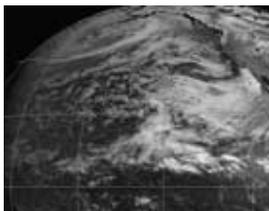


Figure 1: GOES satellite image of Eastern Pacific Ocean at 18:00 UTC 8 May 2006.

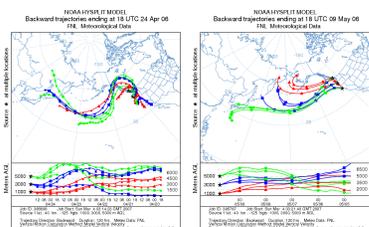


Figure 2: Back-trajectories for RF3 and RF10.

Examples of back trajectories that show recirculation of anthropogenic aerosols from North America over the eastern Pacific Ocean and Asian long-range transport during the INTEX. Figure 2 clearly illustrate that aerosols and gases can traverse the Pacific Ocean in four to six days. Back trajectories were computed using HYSPLIT [Draxler and Hess, 1997]

Marine boundary layer aerosol have relatively low concentrations and often exhibit cloud-processed size distributions which are identified by a trough between 60 and 80 nm – [Hoppel et al., 1986]. The North American continental aerosol was identified by back trajectories which pass over the western coast of Canada and United States and carry regional anthropogenic emissions back over the ocean. These air masses are exclusively within the marine boundary layer. Long-range transport are generally above the boundary layer and are identified by air masses with Asian origins.

Table 1. Summary of C-130 missions for INTEX. Aerosol classifications are separated into the following categories: M – marine boundary layer; NA_c & NA_o – North American continental & ocean; LR – long-range transport

Flight number	Day of year	Launch date/time (UTC)	Duration (hh:mm)	Aerosol classification	CCN data ¹ S _c (%)
2	111	21 April, 17:04	7:45	NA _c	0.2
3	114	24 April, 17:59	5:00	NA _c , LR	0.2
4	116	26 April, 17:19	7:15	NA _c	0.2
5	118	28 April, 17:01	7:45	M, NA _c , LR	0.2
6	121	1 May	n/a	n/a	n/a
7	123	3 May, 17:05	8:30	NA _c , LR	0.2
8	125	5 May	n/a	n/a	n/a
9	128	8 May, 16:07	8:00	M, NA _c , NA _o , LR	0.2
10	129	9 May, 17:29	7:30	NA _c , NA _o , LR	0.2
11	131	11 May, 16:35	7:30	M, NA _c , LR	0.2

¹ Continuous-flow streamwise thermal-gradient CCN instrument [Roberts and Nenes, 2005]

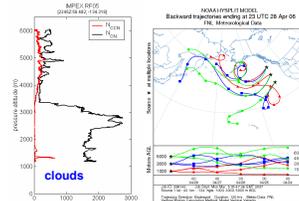


Figure 3: Vertical profiles of CN and CCN and back trajectories during RF05 INTEX.

Research flight 5 was the first flight during INTEX that transected long-range transport of Asian aerosols. Back-trajectories show that air masses aloft (z = 5000 m) traversed the Pacific Ocean while lower trajectories (z < 3500 m) recirculated in a low pressure region off the Alaskan coast. A vertical profile (Figure 3) of the CCN and CN during the flight shows a strong gradient in aerosol concentrations and a superposition of seven thin layers on top of a 2km thick layer. Size distributions (personal communication, University of Hawaii) below 3500 m showed only particles less than 40 nm diameter indicating a recent nucleation event. There had been clouds and precipitation a day or two before sampling, so the CCN below 3500 m were probably removed from wet deposition. As the vertical profile occurred near a frontal system, the source of the thinner sub-layers may be from the Arctic recirculation pool. Further chemical analysis will determine its origins.

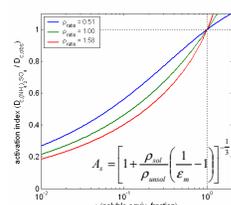


Figure 4: Activation index (As) as a function of equivalent soluble mass fraction for a range of expected density ratios.

The CIFEX study [Roberts et al., 2006] highlighted the importance of chemical effects on CCN measurements and introduces a CCN activation index as a method of classifying the efficiency of an aerosol to serve as CCN relative to an ammonium sulfate particle. The density ratio of the water-soluble and insoluble material is dependent on the aerosol chemistry, but can be constrained by known aerosol species.

To address the CCN activation and the ability of different aerosol classifications to serve as CCN, we introduce a CCN activation index (A_s). The activation index offers a relativistic comparison of the ability of a particle to serve as CCN by comparing an experimentally-determined critical diameter (D_{pc}) to that of ammonium sulfate for a given supersaturation (i.e., $D_{pc, (NH_4)_2SO_4} / D_{pc, obs}$). The CCN activation index provides a useful proxy when aerosol size distributions are known, but detailed size-resolved chemistry has not been measured. Airborne measurements during INTEX provide detailed characterization of the physicochemical properties of aerosol which allow an independent assessment of their mixing states and ability to act as cloud condensation nuclei (CCN).

Acknowledgements: We thank the C-130 crew for their efforts prior to and during the experiment. This experiment was supported by the National Science Foundation.

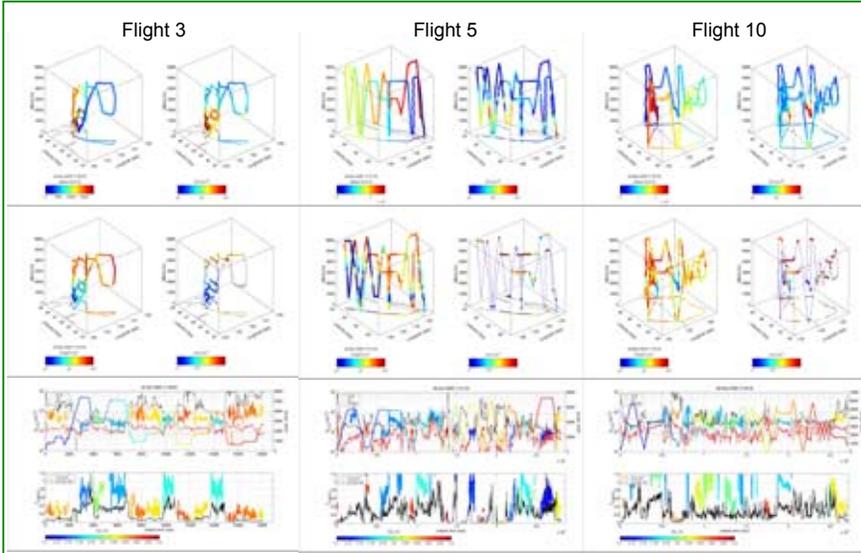


Figure 5: Vertical profiles and time series for total aerosol concentrations (N_{CN}), concentrations of particles > 0.3 μm , and $CCN_{2.0}$.

Comparing size distributions and aerosol chemistry to measured CCN concentrations provides insight on the evolution of these aerosols. Aerosols originating from natural marine sources activate close to the $(\text{NH}_4)_2\text{SO}_4$ 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**.

Summary:

An **airborne experiment** over the Eastern Pacific Ocean measured multiple aerosol layers and **highlighted the influence of aerosol properties on CCN concentrations**. The 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 contrast, **aged dust/biomass burning aerosols scatter light efficiently and serve as CCN**; however, in some cases, they seem to have been **transported across the Pacific without undergoing cloud processing**. In previous experiments, CCN measurements indicate that a significant mass fraction of aged aerosols was insoluble or hydrophobic. The profile seems to show that horizontal mixing during long-range transport may be more significant than vertical mixing, which has important implications on the influence of CCN on the aerosol indirect effect. These layers may have a greater impact on the direct effect during transport until vigorous mixing brings the aerosol into the boundary layer.

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