

GACP Third Year Progress Report for NAG5-8118

A. Accomplishment Report

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TITLE: **A Preliminary Aerosol Climatology for the Pacific Boundary Layer and Free Troposphere**

3rd Year Progress Report

Extensive aerosol data collected during the past decade around the Pacific Basin provide a preliminary assessment of aerosol microphysics for this region and cycling of aerosol in the troposphere. Our aircraft measurements include aerosol horizontal and vertical profiles of concentrations, size distributions, optical properties (light scattering and light absorption) and chemistry. The data characterize aerosol concentrations and properties over all sizes of primary interest to atmospheric chemistry and aerosol physics (i.e. 0.003 to 20 μm) and address topics of aerosol nucleation, transport processes, evolution, entrainment and aerosol radiative effects. Thermal analysis of size distributions obtained from OPC (optical particle spectrometers), differential mobility analyzers (DMA) and condensation nuclei counters (CN) allowed inference of aerosol physico-chemistry, state of mixing, and aerosol with continental vs. "clean" characteristics.

The first year of our projects involved assembling, editing and merging data from diverse aircraft experiments over the past decade (see below). This was followed in the second year by analysis that led to publications and presentations described below on particle production and cycling the Pacific troposphere (Part 1). In this third year we are expanding on this work with a related paper (Part 2) emphasizing size distributions and optics. Portions of these papers have also been presented in numerous meetings as outlined below. We also have our papers and other materials available on our Web Page (<http://pali.soest.hawaii.edu/>).

Some long-term observations results were reviewed in a presentation made by A. Clarke at the **AMS 81st Meeting, Albuquerque, Jan. 2001: "Aerosol Climatology of the Pacific: Production, Transport, Evolution and Mixing Evident in Two Decades of Aerosol Measurements"**. The processes related to the formation and evolution of aerosol, as well as aerosol radiative effects and optical depth, have been explored. The tropical free troposphere is characterized by low aerosol mass but high number and often volatile (at 300C) aerosol, which is naturally formed from sulfuric acid (nucleation) at very high numbers near cloud edges associated with ITCZ convection but appear to grow in size during subsidence. Many vertical profiles and horizontal legs of aerosol

concentration, size distribution and light-scattering in the marine boundary layer (MBL) and the free troposphere, collected during a number of experiments, were included in these studies.

In “**A Pacific Aerosol Survey - Part 1: A Decade of Data on Particle Production, Transport, Evolution and Mixing in the Troposphere**“ [1] we presented an integration of the data obtained during the past decade around the Pacific Basin. These include aircraft based data collected as part of numerous field experiments supported by NASA, NSF and NOAA (GLOBE, ACE1, PEM-Tropics A&B). These experiments provided the extensive aerosol data, including vertical profiles which were used to characterize vertical structure from 70S to 70N. The *in-situ* data is placed in the context of meteorological regimes over the Pacific as well as processes associated with particle formation, growth and evolution and include dust, pollution, sea-salt, sulfates and clean cloud-processed air. The Pacific free troposphere was shown to have aerosol predominately sourced from natural nucleation of aerosol from cloud outflow and from long-range transport of continental emissions. Nuclei formed in the tropics appear to spread in the FT between about 20S and 35N perhaps as a result of the Hadley circulation, while greater continental contributions to FT aerosol is evident above 35N. The data for mid-latitude FT shows fewer number but larger aerosol as compared to tropics. These can often contain a non-volatile core indicative of black carbon and also a combustion derived aerosol (North Pacific springtime). The FT in the subtropics tends to exhibit frequent and marked transitions and mixing between the clean and continental aerosol types. In this paper we have placed greater emphasis on new particle production and evolution and its relation to large scale aerosol features in the troposphere.

In a subsequent paper currently in preparation (**Part 2**) we will place greater emphasis on the total size distribution, its physio-chemical properties and links to aerosol optical properties including greater emphasis on continental sources. The extensive field campaigns of the last decade allowed for a noticeable progress in the characterization of the aerosol transport. Measurements have revealed numerous cases of plumes undergoing long range transport. These observations undermine the notion of “a global background aerosol”. Instead, a dynamic system of independent sources and sinks exists that tends to result in aerosol “rivers”, “layers” and “regions” in the troposphere with specific microphysical and chemical characteristics. In a presentation made by **V.Kapustin at European Aerosol Conference, Leipzig** [7], a two examples of transport were provided, active over 5,000 to 10,000km both in the MBL and in the FT followed by entrainment into the MBL, that reveal the diverse ways such transport may occur.

The extended characterization of the aerosol layers is presented in a paper by Moore et al. [3]. The tropical Pacific region had not been sampled in great detail in the past and had been assumed to be representative of natural background conditions since it is remote from any significant anthropogenic sources. However, aerosol and gas measurements during the NASA Pacific Exploratory Mission – Tropics (PEMT), supported by remote sensing and modeling efforts, revealed the presence of “rivers” of continental outflow propagating into the remote marine atmosphere. The data on both the coarse and fine mode aerosol during the PEM-Tropics A and B field campaigns provided assessment of the spatial variability in aerosol parameters in continental plumes encountered over the Pacific Ocean. Most plumes were encountered in the Southern Hemisphere during PEMT A while the opposite was observed during PEMT B. The bulk of these plumes were found to be located in the free

troposphere (FT) in the 2 to 6 km range. The data suggest a variety of anthropogenic and natural sources for the plumes and show that aerosol size distributions varied considerably from one plume to another. The radiatively important single scatter albedo (ω) ranged from ~0.88 for the PEMT A (biomass) FT plumes to ~0.94 for the PEMT B (dust and pollution) and to 0.96 (pollution and sea salt) within the Marine Boundary Layer (MBL). The vertical profiles of the plumes showed the vertical extent, layering, and evidence of vertical mixing. Vertical profiles often revealed more concentrated plumes aloft and much lower values of ω than in the underlying marine boundary layer

A detailed study of aerosol transport was also discussed, among other topics, in the survey of a decade-long airborne measurements over remote oceans, presented by **A. Clarke** at **8th Scientific Assembly of IAMAS, Innsbruck [5]**, and in the presentation made by **V. Kapustin** at **8th European Symposium on the Physico-Chemical Behavior of Atmospheric Pollutants, Turin [8]**. The data from over 8 major international aircraft programs were accumulated in the past decade that provide insight into the issues of how the aerosol constituents are transported and distributed in the atmosphere and whether there are regional or other characteristics that should be considered.

In the case study paper by Clarke et al., [2], a new Chemical Transport Model (CTM) was used to predict aerosol and gas concentrations and the aerosol optical effects along the flight path. The measurements were conducted near Hawaii on April 1999 during PEMT B and characterized aerosol microphysics, inferred chemistry, optical properties and gases in several extensive dust and pollution plumes, also detected by satellites, which had 10,000km trajectories back to sources in Asia. The CTM model includes aerosol, meteorological fields, dynamics, gas and particle source emissions, a chemistry component and assimilated satellite data. The predicted “river-like” plume structures, as well as contributions of dust and sulfate to aerosol concentrations and optical properties, are in good agreement with the results of flight measurements. Observations and model results confirmed that this aerosol was being entrained into the marine boundary layer between Hawaii and California where it can be expected to modify the type and concentration of cloud condensation nuclei. These observations emphasize the complexity of long range transport and the potential of emerging CTM models to deal with related issues on a global scale.

Recent Experiments In Radiative Column Closure From Aircraft were presented by **A. Clarke** at **8th Scientific Assembly of JAMAS, Innsbruck [6]**. In order to link satellite radiances to the aerosol species responsible a number of experiments have carried out so-called closure studies whereby an over-determined set of measurements is used to establish uncertainties in key parameters. Certain aerosol characteristics such as, for example, scattering extinction may be derived from different types of data, with subsequent comparison. In this fashion measurements of column optical depth can be interpreted in terms of the physics and chemistry of the aerosol contributing to the column burden. In different times or places the optical properties may be dominated by such things as sulfates, dusts, soot and organic carbon, water or more commonly various combinations of these. We describe such column closure measurements from several recent aircraft experiments that were dominated by very different aerosol types. Size distributions, composition, state of mixing (internal/external), growth in response to humidity variations, etc. are included in this assessment.

Summary of three-year period results

The data from over 8 major international aircraft programs, including recent INDOEX (1999), PEMT(1999), SEAS(2000) experiments were accumulated in order to establish microphysical, optical and climatological characteristics of the MBL aerosol that can be linked to known meteorological regimes and long range transport. Though having diverse goals, these experiments included extensive data on aerosol size distributions, optical properties (light scattering and light absorption) and chemistry. Many vertical profiles available from these data were processed to develop improved models of aerosol size, properties, evolution and vertical structure over different regions. In “**A Pacific Aerosol Survey - Part 1**” we focused mostly on particle formation and integral aerosol characteristics. “**Part 2**” is now in preparation where emphasis will be placed upon the time and space variations of particle size distribution, mass and optical properties.

We have our papers and some other materials available on our web site for the Hawaii Group for Environmental Aerosol Research [HiGEAR] to be found at <http://pali.soest.hawaii.edu/>

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