Constraining climate model simulations of aerosol size distributions over the North Pacific and North America using *in-situ* airborne measurements

A DISSERTATION SUBMITTED TO THE GRADUATE DIVISION OF THE UNIVERSITY OF HAWAI'I IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY
IN
OCEANOGRAPHY

MAY 2008

By
Cameron Stuart M^{c}Naughton

Dissertation Committee:

Antony D. Clarke, Chairperson
Barry Huebert
Chris Measures
John N. Porter
Greg Carmichael

Abstract

The effect of aerosols on climate is poorly understood compared to green house gases. Aerosols can scatter and/or absorb solar radiation (the "direct effect") and modify cloud properties (the "indirect effect"), affecting Earth's radiation balance and hydrological cycle. Aerosol lifetimes vary from minutes to weeks in the Earth's atmosphere, so they are heterogeneously distributed in both time and space. Over longer time scales, aerosols can influence climate through sulfur (e.g. CLAW Hypothesis) and iron (e.g. Iron Hypothesis) biogeochemical cycling.

Determination of natural and anthropogenic aerosol effects on past and future climate can only be achieved using global climate models (GCM's). Satellites allow global measurements of the present-day atmosphere, but require calibration/validation by observations *in-situ*. Ground- and ship-based observations are confined to the surface boundary layer which can be decoupled from overlying layers and the free troposphere. Here I use in-situ aircraft measurements from five NASA and NSF airborne field campaigns conducted over the North Pacific and North America between 2001 and 2006 to establish a reduced set of airmass types that are stratified vertically, by source region and according to processes governing their characteristics. For each airmass type the aerosol size distribution, mixing state, optical properties and chemical composition are summarized and discussed.

In this study I found, i) parameterizations of background free troposphere aerosol overestimate extinction by ~50%, minimizing the differences between pre-industrial versus contemporary radiative forcing, ii) meteorological model errors in water vapour mixing

ratio can overwhelm the influence of composition-dependent aerosol hygroscopicity on radiating forcing, iii) aerosol number in convective cloud outflow over North America in summer were reduced by $1/e$ after ~2 days with no detectable increase in aerosol mass or decrease in SO_2, illustrating the need to simulate both mass and number distributions in order to predict aerosol indirect effects, iv) in April 2006 the deposition of Asian pollution and dust to the subtropical Eastern North Pacific could result in a 25% increase in surface ocean nitrogen and a 10-30% increase in iron, SeaWiFS/MODIS satellite retrievals indicate an 8-9% increase in chlorophyll in the same region two weeks after the event.