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**Remote Marine Aerosol: A Characterization of Physical, Chemical
and Optical Properties and their Relation to Radiative transfer
in the Troposphere**

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BACKGROUND

Recently, and because of the potential for aerosol to impact radiative transfer and climate, concern has been expressed over the ability to properly model aerosol optical properties on global scales. These uncertainties have prompted the creation of several optical closure experiments throughout the world (ACE1, ACE2, TARFOX) which seek to test and validate our ability to model the aerosol optical properties from measured aerosol properties. One objective of these experiments is to obtain both in-situ and vertical closure of the aerosol optical properties by comparison with various measurement methods. Modeling the aerosol optical properties requires calculation of the scattering coefficient (with MIE theory) from the aerosol size dependant composition. This is often limited by the uncertainty in aerosol chemical composition as a function of size and in response to changes in relative humidity. If these aerosol properties can be determined, and the vertical structure of aerosol and RH are known then the total column optical properties can be calculated and compared to either in-situ column extinction measurement and/or to satellite retrievals of optical depth in order to assess "closure" between measured and modeled properties. Such "closure" is achieved only when agreement is found within the uncertainties associated with the various measurement and modeling steps inherent to this procedure and when these uncertainties are sufficiently small to yield results of practical value.

OBJECTIVES

Our research effort is focused on improving our understanding of aerosol properties needed for optical models for remote marine regions. This includes in-situ and vertical column optical closure and involves a redundancy of approaches to measure and model optical properties that must be self consistent. The model is based upon measured in-situ aerosol properties and will be tested and constrained by the vertically measured spectral differential optical depth of the marine boundary layer, MBL. Both measured and modeled column optical properties for the boundary layer, when added to the free-troposphere and stratospheric optical depth, will be used to establish spectral optical depth over the entire atmospheric column for comparison to and validation of satellite derived radiances (AVHRR).

INSTRUMENTATION

Cape Kumukahi Coastal Measurements

- 1) Laser Optical Particle counter (PMS - LAS-x) provides 256 channels of size resolution from 0.15-7.0 μ m. Thermal analysis is used to differentiate species such as sulfuric acid, ammonium sulfate and sea-salt.
- 2) Shadowband Photometer, 7-wavelength differential aerosol optical depth measurements between the coast and Mauna Loa Observatory (MLO) (3,500m alt.). The difference yields the optical depth of the MBL constrains the effective MBL size distribution.
- 3) The new TSI 3-wavelength total and backscatter nephelometer was finally operational in 1995. We added a Berner impactor (50% cut at 1 μ m) to the unit that can be put in the airstream to obtain total and submicron aerosol scattering and backscattering coefficients at three wavelengths.
- 4) Condensation nucleus counters (TSI 3760) provide an indication of total aerosol number and help identify background conditions and perturbed conditions (eg. volcanic haze, pollution).
- 5) Aethalometer (McGee Scientific) provides the aerosol absorption coefficient and an estimate of pollution from absorbing combustion soot.
- 6) Differential Mobility Analyzer - size distribution from 0.1 to 0.5 μ m.
- 7) Meteorological sensors

Light Aircraft Measurements (Cessna 172)

- 1) Mini-optical particle counters (MET 1 237A and 237H) that together describe the size distribution from 0.1 to 10 μ m.
- 2) Mini-nephelometer - Radiance Research single wavelength (530 nm). We have fabricated a low pressure drop impactor for it that can be used to also obtain total and submicron scattering coefficients.
- 3) CN counter (TSI-3760) - for gauging rapid variability in aerosol concentrations.
- 4) GPS - for logging our 3-D position continuously.
- 5) Four wavelength hand held photometers for differential optical depth determination.

SUMMARY OF 1995 KUMUKAHI EXPERIMENT

Measurements were made at Kumukahi, Hawaii (Jan-Feb, 1995) on the most eastern tip on the upwind coast of the Big Island of Hawaii. It is exposed to clean Pacific air from the northeast trades and the clearest skies due to reduced orographic lifting along this coast. Our measurements include the aerosol size distribution from 0.02 - 7.5 μm diameter (custom optical particle counter, OPC and differential mobility analyzer, DMA), the aerosol volatility (used to infer the aerosol composition), the aerosol scattering coefficient (TSI three wavelength Nephelometer), the aerosol absorption coefficient (Aethalometer), differential aerosol optical depth at 7 wavelengths (measured at Mauna Loa Observatory-MLO at 3,500 m alt. and the coast) and satellite derived aerosol optical depth. Our preliminary 1994 measurement phase established a measurement methodology, tested and calibrated instruments, evaluated the sampling site and established intercomparison procedures with MLO. Recent activities expanded our characterization the marine boundary layer (MBL) optical depth and aerosol properties and completed the development and deployment of a light aircraft (Cessna) measurement capability suitable for the in-situ characterization of aerosol physical and optical properties. During the 2 month deployment a near continuous record was established for aerosol physical and optical properties at the coastal site. This baseline instrumentation was also used for intercalibration of the smaller portable instrumentation that comprised our aircraft package.

Eleven 2-3 hour flights were made during this period for both test and measurement purposes. When possible flights and vertical profiles were flown upwind of our ground station in order to allow comparison of the in-situ vertical profiles with our MBL differential optical depth measurements and with AVHRR aerosol optical depth retrievals over the ocean. The combination of operational instrumentation, satisfactory weather conditions, aircraft availability coincident with AVHRR images and well defined aerosol fields were present on four of these flights that allowed for complete vertical profiles through the trade wind inversion. Two were obtained for typical "clean" marine conditions and two were for a MBL impacted by elevated volcanic sulfate haze. The latter was planned for in order to assess the optical influence of a "pure" well defined sulfate haze on the MBL column optical properties since this volcanic haze has many of the same properties as continental sulfate pollution haze.

ACE-1 FIELD PHASE - NCAR C-130 - TASMANIA

During November-December 1995 we commenced the third phase of our program. This was aboard the NCAR C-130 as part of the ACE-1 program. Our NASA objectives include column radiative closure based upon the same or similar instrumentation and approach described above. These were carried out at various locations along the flight track from Alaska to Australia including our primary base site out of Hobart Tasmania. This objective includes intercomparisons (column closure) measurements in conjunction with column extinction measurements using the Total Diffuse and Direct Radiometer (TDDR-F. Valero, NASA/Scripps) aboard the aircraft. This was supplemented by extensive in-situ aerosol probe data and aerosol chemistry as well as satellite derived optical depths. These missions were one of the objectives of the ACE-1 program and about 20% of the 33 flights include closure

opportunities. Table 1 is a summary of the ACE-1 flight missions. Dedicated "column closure" missions were made in the predominately sulfuric acid plume from Kilauea downwind of the Hawaiian Islands. Direct coordination with satellites allowed comparison with satellite derived properties. We also carried out dedicated "closure" missions in several "clean" air regions near Tasmania including one case where virtually only sea-salt aerosol was present.

ACTIVITIES 1996-1997

Our final efforts under this proposal have focussed upon the analysis of the radiative closure efforts mentioned above. Some delay has been encountered while waiting for some of the differential optical depth analysis to be completed by other ACE-1 investigators. We have also worked on refinements to satellite retrieval algorithms. We have now completed:

- 1) Local closure between the OPC and DMA with our single and 3 wavelength nephelometer. This closure focused on aerosol of known composition (lab generated) and later on ambient aerosol.
- 2) Analysis of aircraft vertical profiles during our experiment using our new integrated light-aircraft package (nephelometer, particle counters, CN counter, GPS).
- 3) The field data was analyzed and tested against the measured spectral differential optical depth.
- 4) The optical model and sunphotometer data was compared to satellite retrievals in order to test and refine retrieval algorithms.
- 5) The above approach was applied to the NCAR C-130 aircraft measurement program during ACE-1. This included closure studies with the TDDR (F. Valero, NASA/Scripps). Cases of elevated sulfate, clean marine background and low sulfate with high sea-salt were evaluated.
- 6) Uncertainty analysis of closure approach.

PAPERS BASED UPON THIS WORK

- * Papers on clean background radiative closure are presently under preparation for JGR based upon the above work.

THESIS BASED ON THIS WORK

- * **Measured and Modeled Aerosol Radiative Properties in the Marine Boundary Layer**
Masters Thesis by Xinli Li, University of Hawaii, Honolulu, Hi, November 1997.

PRESENTATIONS BASED UPON THIS RESEARCH

- * Clarke A.D. and R. Schnell, Black Carbon Variability in the Pacific Free troposphere and Marine Boundary Layer, 5th International Carbon Conference, Berkeley, CA., Aug. 22-26, 1994.
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- * Clarke, A.D. Thermal volatility techniques and their application to current problems in atmospheric chemistry - Gordon Conference on Atmospheric Chemistry, R.I., June 1995
- * Clarke A. D., X. Li, J. Porter and M. Litchy, Radiative Properties of Marine Aerosol: Column Closure in Clean and Perturbed Marine Boundary Layer, American Association for Aerosol research, Orlando, 1996
- * Litchy, M. and A.D. Clarke, Light-Aircraft Portable Sampling Package for Measuring Differential Aerosol Radiative Properties in the Pacific Marine Boundary Layer, paper 1C2, Annual Conference of American Association for Aerosol Research, Orlando, Oct, 1996.
- * Clarke, A.D., X. Li, J. Porter and M. Litchy, A Preliminary Investigation of Column Radiative Closure During ACE-1, Fall AGU, 1996.
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- * Clarke, A.D., Column Radiative Closure in the Clean and Perturbed Marine Boundary Layer, Spring AGU, Baltimore, 1997
- * A. D. Clarke, Xinli Li, J. Porter, Radiative Column Closure in the Marine Boundary Layer During Ace-1: Instrumentation and Implementation Issues, AAAR, Oct. 1997