Final Progress Report to Global Aerosol Climatology Program (GACP)
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Improved Exploitation of Field Data Sets to Address Aerosol Radiative-Climatic Effects and Development of a Global Aerosol Climatology

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Goals and Objectives (from February 1998 proposal)

The goals of this research are to (A) improve understanding of aerosol radiative forcing of climate and (B) help guide the development of an aerosol climatology. Our proposed approach is to improve the exploitation of data already acquired in two multiplatform field experiments, the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) and the second Aerosol Characterization Experiment (ACE-2), plus other experiments when appropriate. The objectives are given by the following task specifications:

(1) Improve the cloud-screening in the TARFOX and ACE-2 airborne and shipborne sunphotometer optical depth data sets, perform more general quality checks, and archive reprocessed data as necessary,

(2) Investigate the question of the best aerosol optical models (e.g., complex refractive indices, internal vs. external mixtures, shapes) to account for observed aerosol compositions (e.g., water, carbonaceous material, sulfates, minerals) and for observed relationships between measured size distributions and measured optical coefficients (e.g., extinction, backscatter, total scatter, absorption),

(3) Develop and test a new, more automated technique for retrieving particle size distributions from optical depth spectra, and apply it to field-measured data sets,

(4) Compare results of the new size distribution retrievals to those of previous retrievals, to size distributions measured in situ, and to models used in retrievals from imaging spectrometers on satellites and aircraft, and

(5) Investigate the relationship between water vapor and aerosol properties, with the goal of using water vapor information to improve satellite retrievals of aerosol optical depth and radiative forcing.

3rd Year Progress Report

Four papers (three journal, one proceedings) were accepted in Year 3, including two for the GACP issue of *J. Atmos. Sci*. Here we give a one-sentence synopsis of each paper; more extensive results are described in the following section in the context of related work from the full three-year study.

Bergstrom et al. (2001) compares the wavelength dependence of aerosol absorption measured in TARFOX with theoretical predictions for small black carbon particles, finding good agreement. Russell et al. (2001) compares aerosol absorption derived by diverse techniques in TARFOX and ACE-2, noting that absorption from irradiance measurements is larger than from other techniques. Redemann et al. (2001) describes use of a core/shell model of internally mixed soot/sulfate particles to study changes in particle size, light scattering and absorption caused by humidity changes. Redemann et al. (2000) present a method to estimate aerosol radiative effects from two-wavelength lidar measurements; they show that using concurrent optical depth measurements significantly reduces uncertainty in derived aerosol forcing.

A funding augmentation helped support measurements by the 14-channel Ames Airborne Tracking Sunphotometer (AATS-14) in the Asian Aerosol Characterization Experiment (ACE-Asia, March-May 2001). 19 flights on the Twin Otter studied properties and radiative effects of desert dust, urban pollution, and other aerosols. Included were measurements to validate aerosol retrievals by sensors on EOS Terra and other satellites.

Summary of Results Obtained During the Three-Year Period of GACP:

Results are grouped by the five Tasks listed above, plus a sixth section for results beyond the scope of these five Tasks. All told, our three-year GACP funding helped support 26 peer-reviewed journal papers authored by our team members (13 first-authored, 13 co-authored) and 18 conference abstracts/proceedings first-authored by our team members (see GACP Bibliography below).

Task 1. Airborne Sunphotometer Data Quality Checks: Data obtained with Ames Airborne Tracking Sunphotometers (AATS-6 & -14) on ship and aircraft in ACE-2 were cloud-screened and submitted to the ACE-2 Archive in Year 1. A new cloud screen was developed and applied to the AATS-6 data set from TARFOX in Year 2. The cloud-screened TARFOX data set was archived at the Langley DAAC. The screened ACE-2 and TARFOX data are available at http://geo.arc.nasa.gov/sgg/ACE-2/ACE-2_Website_Data/Data/ and http://eosweb.larc.nasa.gov/PRODOCS/tarfox/table_tarfox.html, respectively.

Task 2. Mixed Aerosol Optical Modeling: This task provided support for four journal publications. Redemann et al. (2000a) developed a technique for deriving composite aerosol optical properties by combining lidar, sunphotometer, and in situ particle size distribution data. They used this technique to estimate the vertical structure of the effective aerosol complex index of refraction for two TARFOX case studies. A companion paper, Redemann et al. (2000b), used the combined aerosol fundamental properties thus retrieved to estimate the vertical structure of the single scattering albedo (SSA). These SSA profiles were compared to independent estimates using in situ measurements of aerosol scattering and absorption. The vertical profiles of the aerosol SSA were shown to agree well within the error bars of the two methods. (See also Task 6 below for the radiative forcing results in Redemann et al. (2000b).)

Redemann et al. (2001) used a core/shell model of internally mixed, single sulfate particles with soot cores to study the changes in particle size, light scattering and absorption that accompany changes in ambient relative humidity. The sulfate species considered were sulfuric acid, ammonium sulfate and ammonium bisulfate. Absorption humidification factors were quantified for a range of realistic, dry particle size distributions.

In a paper accepted for the GACP issue of *J. Atmos. Sci*., Bergstrom et al. (2001) compared the wavelength dependence of aerosol absorption measured in TARFOX with theoretical predictions for small black carbon particles. TARFOX-measured aerosol absorption varies as λ^{-1} for wavelengths λ between 0.4 and 1.0 μm, a result predicted by theory for wavelength-independent refractive index. For typical size distributions of sulfate-soot particle mixturess, this absorption wavelength dependence implies that aerosol single scattering albedo decreases with wavelength.

Task 3. Automated Size Distribution Retrievals: A new multimodal technique, based on regularities found in AERONET sky radiation retrievals for the US East coast by Remer et al. (1999) and Remer and Kaufman; (1998), was programmed and tested with AATS-6 and -14 data from TARFOX and ACE-2. These tests demonstrated the ability to retrieve multimodal size distributions quickly, offering the promise of automated retrievals. A major advantage of this technique is that it can incorporate size-dependent refractive indices, thereby accounting for observed size-dependent particle compositions. Multimodal parameter evolutions along horizontal transects and vertical profiles have now been retrieved. For an ACE-2 boundary layer case, Schmid et al. (2000b) compared retrievals by the multimodal technique to retrievals by constrained linear inversion and to in situ measurements by optical particle counters. The three methods agreed well for the optically efficient part of the accumulation mode. However, the multimodal technique placed the large-particle mode at smaller radii than the in situ measurements—possibly indicating a need to tune the multimodal technique to the Eastern Atlantic location of the ACE-Asia measurements.

<u>Task 4. Size Distribution Comparisons and Closure Studies</u>: This task supported the use of our airborne sunphotometer data in a variety of closure studies published in the ACE-2 special issue of *Tellus* and in the second TARFOX special issue of *J. Geophys. Res.*

In the ACE-2 *Tellus* issue Schmid et al. (2000a) compared sunphotometer-retrieved and in-situ-measured size distributions from the Pelican A/C. In related closure studies they compared airborne sunphotometer-measured optical depth profiles to vertical integrals of the Collins et al. (2000) results obtained from size spectrometer, nephelometer, and absorption photometer. Schmid et al. (2000a) also differentiated sunphotometer optical depth profiles vertically to yield extinction profiles, which they compared to the profiles mentioned above. In general the aerosol optical depths or extinctions derived from in situ scattering, absorption and/or size distribution profiles measured on the Pelican aircraft were less than airborne sunphotometer values (Schmid et al., 2000a; Collins et al., 2000; Livingston et al., 2000). Agreement was improved by accounting for inlet aerodynamic size selection, inadvertent and intended evaporation, and optical sizing calibration, all as a function of size-resolved composition. However, corrections for these processes have relatively large error bars, and significant differences remain. Compared to layer optical depths determined by airborne sunphotometer, values from absorption photometer and humidified nephelometer differed by +15% to –44%, and humidified values from size spectrometers differed by +20% to -40%.

Livingston et al. (2000) reported shipboard sunphotometer measurements of aerosol optical depth (AOD) spectra and column water vapor (CWV). AODs inferred from shipboard aerosol lidar backscatter measurements during one day were consistent with those measured by the shipboard sunphotometer, but the uncertainties associated with deriving optical depth from the shipboard lidar data were large (~factor 2) because of the need to assume an extinction-to-backscatter ratio that differed for maritime and continental-influenced aerosols. . The wavelength dependence of shipboard sunphotometer AODs was compared with the corresponding dependence of aerosol extinction calculated from shipboard measurements of aerosol size distribution and of total scattering measured by a shipboard integrating nephelometer. Results of these comparisons were highly variable, illustrating the great difficulty of deriving column values from point measurements. Livingston et al. (2000) also performed column closure tests between AODs measured by sunphotometer and computed by combining shipboard particle size distribution measurements with models of hygroscopic growth and radiosonde humidity profiles; results are described under Task 5 below.

Welton et al. (2000) presented micropulse lidar measurements of upslope aerosols and African dust layers over Izaña on Tenerife (Table 2g). Comparison to an airborne sunphotometer profile (Schmid et al., 2000) within the dust layer yielded differences of ±0.02 or less at all altitudes (~2500-3800 m asl), over which optical depth decreased from 0.22 to 0.05.

Durkee et al. (2000) compared aerosol optical depth (AOD) values retrieved from AVHRR satellite retrievals to measurements by AATS-6, AATS-14, and other sunphotometers in ACE-2. They showed that the presence of African dust caused negative errors in AVHRR-retrieved AOD. For all cases, including dusty and dust-free conditions, the AOD standard error of estimate was 0.025 for 630 nm wavelength and 0.023 for 860 nm wavelength.

Flamant et al. (2000) reported airborne lidar measurements and closure studies for a European pollution outbreak sampled by the ARAT aircraft. The lidar mapped vertical profiles of the pollution plume and the marine boundary layer aerosol as the plume was carried from the coast of Portugal near Sagres over the ship and beyond over the Atlantic Ocean. Values of aerosol optical depth (AOD) derived from the lidar ranged from 0.055 to 0.10. When compared to ship sunphotometer measurements, differences were 0.02 or less, within the combined lidar and sunphotometer uncertainties. In contrast, AODs derived from METEOSAT radiances exceeded the lidar values by 0.01 to 0.08, with the largest differences in the area where the pollution plume contributed most to column optical depth. Flamant et al. suggest that the difference may be caused by large uncertainties associated with the Meteosat sensitivity for small AODs or by the presence of thin scattered clouds.

Russell and Heintzenberg (2000) gave an overview of the ACE-2 Clear Sky Column Closure Experiment (CLEARCOLUMN), including results from the above studies and many others. They found a wide range in the degree of agreement resulting from closure tests. In general, the smallest discrepancies were found in comparisons among (1) different techniques to measure an optical property of the ambient, unperturbed aerosol (e.g., optical depth, extinction, or backscatter by sunphotometer, lidar, and/or satellite) or (2) different techniques to measure an aerosol that had passed through a common sampling process (e.g., nephelometer and size spectrometer measurements with the same or similar inlets, humidities and temperatures). Typically, larger discrepancies were found between techniques that measure the ambient, unperturbed aerosol and those that must reconstruct the ambient aerosol by accounting for (a) processes that occur during sampling (e.g., aerodynamic selection, evaporation of water and other volatile material) or (b) calibrations that depend on aerosol characteristics (e.g., size-dependent density or refractive index). A primary reason for the discrepancies in such cases is the lack of validated hygroscopic growth models covering the necessary range of particle sizes and compositions. Other common reasons include (1) using analysis or retrieval techniques that assume aerosol properties (e.g., density, single scattering albedo, shape) that do not apply in all cases and (2) using surface measurements to estimate column properties.

In the second TARFOX special issue of *J. Geophys. Res.*, Ferrare et al. (2000a) compared aerosol optical thickness (AOT) and precipitable water vapor (PWV) measurements derived from ground and airborne lidars and Sun photometers during TARFOX (Tropospheric Aerosol Radiative Forcing Observational Experiment). Aerosol extinction profiles and estimates of AOT were derived from the lidar measurements using a value for the aerosol extinction/backscatter ratio Sa=60 sr. The lidar measurements of AOT were found to be generally within 10-15% of the AOT measured by the NASA Ames Airborne Tracking Sunphotometer (AATS-6). Ferrare et al. (2000b) examined aerosol extinction and optical thickness from the Lidar Atmospheric Sensing Experiment (LASE) on the ER-2 aircraft, AATS-6 on the C-131A, and other measurements during TARFOX. The LASE profiles of AOT were found to be about 10% higher than those derived from the airborne Sun photometer, which in turn were about 10% higher than those derived from the airborne in-situ measurements. These differences are generally within the error estimates of the various measurements.

Hartley et al. (2000) compared ambient aerosol optical depths (AOD) derived from airborne in situ measurements of aerosol properties and from AATS-6 measurements in TARFOX. At wavelength 450 nm the AATS-6 measurements of AOD exceeded in situ measurements by 12%±5% on average. At wavelength 550 nm the aerosol single scattering albedo derived from nephelometer and absorption photometer measurements was 0.95±0.03.

This task also supported final revisions, typesetting, and page charges for two papers in the first TARFOX issue of *J. Geophys. Res.* (Russell et al., 1999a,b).

A new ACE-2 closure result with the University of Arizona Micro-Pulse Lidar was published in conference proceedings [Schmid et al., 2000b].

<u>Task 5. Water Vapor/Aerosol Interactions and Satellite Retrievals</u>: We submitted to the LaRC DAAC and the ACE-2 Archive the water vapor column contents and density profiles retrieved from TARFOX and ACE-2 measurements by AATS-6 & -14. Livingston et al. (2000) compared column water vapor measured by radiosonde and by AATS-6 on the ship in ACE 2, finding good agreement, with an rms difference of 0.09 g cm^{-2} in 7 samples having a CWV range of 1.6 to 3.2 g cm^{-2}. Livingston et al. (2000) also performed column closure tests between AODs measured by sunphotometer and computed by combining shipboard particle size distribution measurements with models of hygroscopic growth and radiosonde humidity profiles (using the assumption that dry particle size distribution and composition were independent of height in the boundary layer). These closure tests often produced big discrepancies, in large part because of their great

sensitivity to models of hygroscopic growth, which vary considerably and have not been validated over the necessary range of particle size/composition distributions.

This task supported the updating of our water vapor retrieval algorithms to include the latest spectroscopy [e.g., Ingold et al., 2000; Schmid et al., 2001d]. Results of water vapor intercomparisons were presented and published [Schmid et al., 2000c,d,e, 2001]. Optical depths we measured in the 1997 Atmospheric Radiation Measurement Water Vapor Intensive Observation Period (ARM WVIOP) were used to study the relationship between water vapor and model-measurement differences in solar irradiance at the ground [Pilewskie et al., 2000].

Task 6. Results Beyond the Scope of the Five Proposed Tasks

6A. Flux Change/Radiative Forcing Studies: Bergstrom and Russell (1999) estimated solar radiative flux changes caused by aerosols over the mid–latitude North Atlantic by combining optical depths from AVHRR measurements with aerosol intensive properties determined in TARFOX. They found cloud–free, 24–hour average flux changes at the tropopause ranging from –9 W/m^2 near the eastern US coast for summer-average optical depths to –1 W/m^2 in the mid–Atlantic for winter-average optical depths. Cloud–free North Atlantic regional-seasonal averages range from –5.1 W/m^2 in summer to –1.7 W/m^2 in winter, with an annual average of –3.5 W/m^2. The cloud-free summer and annual-average values exceed in magnitude the radiative forcing by anthropogenic greenhouse gases, but are opposite in sign. Cloud effects, estimated from ISCCP data, reduce the regional annual average to –0.8 W/m^2. These values are for a moderately absorbing TARFOX aerosol (ω(0.55 µm) = 0.9); values for a nonabsorbing aerosol are ~30% more negative. Bergstrom and Russell (1999) also compared their results to a variety of other calculations of aerosol radiative effects.

Redemann et al. (2000b) used the Fu-Liou broadband radiative transfer model to calculate vertical profiles of flux changes caused by TARFOX aerosols. The aerosol optical properties were calculated from independent vertically-resolved estimates of the complex aerosol indices of refraction in two to three distinct vertical layers, using profiles of *in situ* particle size distributions measured aboard the University of Washington research aircraft. Aerosol single-scattering albedos at 450 nm thus determined range from 0.9 to 0.985, while the asymmetry factor varies from 0.6 to 0.8. The instantaneous shortwave aerosol radiative forcings derived from the optical properties of the aerosols are of the order of -36 W m^{-2} at the top of the atmosphere and about –56 W m^{-2} at the surface for both case studies.

Redemann et al. (2000c) presented a method to estimate aerosol radiative effects from two-wavelength lidar measurements. They found that diurnally averaged aerosol-induced radiative flux changes calculated from the estimated aerosol properties can differ greatly from true radiative forcing. However, using concurrent optical depth measurements reduces this uncertainty significantly.

6B. Single Scattering Albedo Comparison. In a second paper accepted for the GACP issue of *J. Atmos. Sci*., Russell et al. (2001) compared aerosol single scattering albedos ω determined in TARFOX and ACE-2 by a variety of techniques. The techniques included fitting of calculated to measured radiative fluxes; retrievals of ω from skylight radiances; best fits of complex refractive index to profiles of backscatter, extinction, and size distribution; and in situ measurements of scattering and absorption at the surface and aloft. They show that both TARFOX and ACE-2 found a fairly wide range of values for ω at midvisible wavelengths (~550 nm), with $0.85 \leq \omega_{midvis} \leq 0.99$ for the marine aerosol impacted by continental pollution. Frequency distributions of ω could usually be approximated by lognormals in ω_{max}-ω, with some occurrence of bimodality, suggesting the influence of different aerosol sources or processing. In both TARFOX and ACE-2, closure tests between measured and calculated radiative fluxes yielded best-fit values of ω_{midvis} of 0.90±0.04 for the polluted boundary layer. Although these results have the virtue of describing the column aerosol unperturbed by sampling, they are subject to questions about representativeness and other uncertainties (e.g., pyranometer thermal offsets, unknown gas absorption). The other techniques gave larger values for ω_{midvis} for the polluted boundary layer, with a typical result of ω_{midvis} = 0.95±0.04. Russell et al. (2001) conclude that current uncertainties in ω are large in terms of climate effects, and that more tests are needed of the consistency among different methods and of humidity effects on ω.

6C. Measurements in Field Studies A funding augmentation in Year 2 helped support instrument preparation and measurements in the Puerto Rico Dust Experiment (PRIDE, June-July 2000) and the South African Regional Science Initiative (SAFARI, August-September 2000). These experiments study the radiative effects of desert dust, biomass smokes, and other aerosols. They include major validation studies of aerosol retrievals by sensors on EOS Terra and other satellites. The 6-channel Ames Airborne Tracking Sunphotometer (AATS-6) made 21 flights on a Navajo in PRIDE, and the 14-channel version (ATS-14) made 24 flights on the University of Washington CV-580 in SAFARI-2000.

A second funding augmentation in Year 3 helped support measurements by AATS-14 in the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia, March-May 2001). 19 flights on the Twin Otter studied properties and radiative effects of desert dust, urban pollution, and other aerosols. Included were measurements to validate aerosol retrievals by sensors on EOS Terra and other satellites.

Data analyses for our measurements in PRIDE, SAFARI 2000 and ACE-Asia are supported by our Earth Observing System Inter-Disciplinary Science (EOS-IDS) task. Hence, presentations and publications resulting from our PRIDE, SAFARI 2000 and ACE-Asia analyses are not included in this report.

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