

FORM A: GACP ACCOMPLISHMENT REPORT

Name: Philip B. Russell

Institution: NASA Ames Research Center

TITLE: Improved Exploitation of Field Data Sets to Address Aerosol Radiative-Climatic Effects and Development of a Global Aerosol Climatology

ABSTRACT (from proposal): The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) and the second Aerosol Characterization Experiment (ACE-2) gathered extensive data sets on aerosol properties and radiative effects. TARFOX focused on the urban-industrial haze plume flowing from the eastern United States over the western Atlantic Ocean, whereas ACE-2 studied aerosols carried over the eastern Atlantic from both European urban/industrial and African mineral sources. We participated in TARFOX and ACE-2 by playing overall coordinating roles, by making aircraft and ship sunphotometer measurements, by analyzing the data in terms of aerosol-induced radiative flux changes, and by participating in closure studies that test the mutual consistency of a variety of measurements and the models that link them. The sunphotometer data set includes optical depth spectra (380-1020 nm and 380-1556 nm) in horizontal transects and vertical profiles, retrieved aerosol size distributions, water vapor columns and profiles, and ozone columns. Our previous TARFOX funding supported the archival of the optical depth data, their use in the flux-change studies mentioned above, and their comparison to optical depths retrieved from the satellite sensors AVHRR, GOES Imager, and ATSR-2, and from the MODIS Airborne Simulator. Our ACE-2 funding supported analogous archival and studies, with increased emphasis on water vapor retrievals and vertically resolved aerosol and water vapor intercomparisons. Our TARFOX funding was exhausted in mid-FY98, and our ACE-2 funding will be exhausted by the end of FY 1999.

For the funding requested from GACP we propose to conduct additional analyses of the TARFOX, ACE-2, and related data sets, with the goals of greatly increasing their exploitation in aerosol radiative/climatic studies and in the development of a global aerosol climatology. In particular, we propose to: (1) Improve the cloud-screening in the sunphotometer optical depth data set, 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 sources, (3) Develop and test a new, more automated technique for retrieving particle size distributions from optical depth spectra, and apply it to the TARFOX and ACE-2 data sets, (4) Compare results of the new 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 column water vapor and aerosol properties, with the goal of using water vapor information to improve satellite retrievals of aerosol optical depth and radiative forcing. We will use the results of these investigations in collaborative studies with other members of the Aerosol Radiative Forcing Science Team, so as to advance the development of a global aerosol climatology and to reduce uncertainties in aerosol effects on climate.

GOALS:

The goals of this effort 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).

OBJECTIVES:

- (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 the TARFOX and ACE-2 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.

PROGRESS TO DATE (by numbered objective above):

1. Airborne Sunphotometer Optical Depth Data Quality Checks: Data obtained with Ames Airborne Autotracking Sunphotometers (AATS-6 & AATS-14) on ship and aircraft in ACE-2 were cloud-screened and submitted to the ACE-2 Archive. A cloud screen is being developed for the TARFOX AATS data.

2. Mixed Aerosol Optical Modeling: A technique for deriving composite aerosol optical properties by combining lidar, in situ, and sunphotometer data was described by Redemann et al. (1999a). Results are being compared to those obtained by the ELSIE mixing program, by the mixing approach of Collins et al. (1999), and by the flux best-fit method of Russell et al. (1999).

3. Automated Size Distribution Retrievals: A new multimodal technique, based on regularities found in AERONET sky radiation retrievals by Remer et al. (*JGR*, 104, 2223, 1999), was programmed and tested with TARFOX AATS-6 and -14 data. Initial results show the ability to retrieve multimodal size distributions quickly and offer promise of automated retrievals that use any of several specified mixed-aerosol composition models, including models with size-dependent composition. The ability to use size-dependent composition in size-distribution retrievals will be a significant step forward for us.

4. Size Distribution Comparisons and Closure Studies: Comparisons between sunphotometer-retrieved and in-situ-measured size distributions from the Pelican A/C in ACE-2 were described by Schmid et al. (1999). In related closure studies Schmid et al. compared airborne sunphotometer-measured optical depth profiles to vertical integrals of the Collins et al. (1999) results obtained from size spectrometer, nephelometer, and absorption photometer. Their sunphotometer optical depth profiles are also differentiated vertically to yield extinction profiles, which are compared to the profiles mentioned above. In general the aerosol optical depths 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., 1999; Collins et al., 1999; Livingston et al., 1999). 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. But some differences remain. Compared to layer optical depths determined by airborne sunphotometer, humidified nephelometer values were less by $25\% \pm 10\%$, and humidified values from size spectrometers were less by 7% to 30% (Schmid et al., this issue ; Collins et al., this issue).

5. Water Vapor/Aerosol Interactions and Satellite Retrievals: Water vapor column contents and density profiles retrieved from TARFOX and ACE-2 measurements by AATS-6 & -14 were submitted to the LaRC DAAC and the ACE-2 Archive. Comparisons between CWV measured by radiosonde and by AATS-6 on the ship in ACE 2 yielded 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., 1999). Livingston et al., 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.

6. Flux Change/Radiative Forcing Studies (extras not in proposal): Flux changes caused by North Atlantic aerosols were estimated by combining AVHRR aerosol optical depths with aerosol properties determined in TARFOX (Bergstrom and Russell, 1999). This work is also summarized in FORM B HIGHLIGHTS. Vertical profiles of flux changes caused by TARFOX aerosols were described by Redemann et al. (1999b).

FUTURE PLANS: See FORM C.

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FORM B: GACP SIGNIFICANT HIGHLIGHTS: See separate file with color illustration.

FORM C: FUTURE PLANS

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PLANS FOR SECOND YEAR OF GACP: (Listed by numbered objective above)

1. Airborne Sunphotometer Optical Depth Data Quality Checks: During the second year of funding we will test the applicability of several single- and multi-wavelength criteria to filter out data contamination caused by contrails and thin cirrus clouds. Single-wavelength criteria include fine-scale standard deviations and the "smoothness" criterion developed by Smirnov et al. [submitted] for AERONET. Multi-wavelength criteria include variability in the Angstrom parameter and scatter-plot techniques. Each approach will be tested with our airborne sunphotometer data obtained during horizontal transects and vertical profiles in TARFOX. A key goal is to identify occurrences of cirrus clouds, which may have caused some erroneous aerosol optical depth estimates in the TARFOX data set, including apparent increases of optical depth with altitude during aircraft profiles. Among single-wavelength criteria, the fine-scale-standard deviation technique (using 9 measurements in 3 seconds) appears particularly promising, since it has been successfully applied to the ACE-2 data set obtained using AATS-14 aboard the CIRPAS Pelican aircraft. Among multiwavelength techniques, the scatter-plot technique developed by Kent et al. [1993] seems promising, since it has been used successfully to identify cloud occurrences in SAGE II data sets.

2. Mixed Aerosol Optical Modeling: For two TARFOX case studies, the vertical structure of the effective aerosol complex index of refraction has been estimated from a combination of lidar, sunphotometer and in situ particle size distribution data [Redemann et al., 1999b]. The optical properties calculated on the basis of these refractive indices and the in situ particle size distributions will be further compared with independent measurements of, for example, the aerosol single scattering albedo. We plan to apply a similar aerosol refractive index retrieval technique to ACE-2 observations using micropulse lidar data taken at Las Galletas [Powell et al., submitted], aerosol optical depth data obtained using our airborne 14-channel sunphotometer, and in situ particle size distribution measurements. We will utilize the ACE-2 size-distribution measurements of Collins et al. [submitted] and their size-resolved chemical compositions to forward-calculate aerosol optical and radiative properties. The agreement of the forward-calculated optical properties with independent measurements will be an indicator of the validity of the model assumptions regarding the mixing state (internal vs. external) and different effective medium approximations for the aerosol complex index of refraction (e.g., ELSIE, partial molar refractivity, Maxwell-Garnett, etc.). For internally mixed particles, the effects of a shell/core particle morphology on the derived aerosol optical properties, in particular the absorption, will be investigated. The shell/core particle morphology will be used to investigate aerosol humidification factors (i.e., the changes of aerosol scattering and absorption coefficients due to changes in ambient relative humidity).

3. Automated Size Distribution Retrievals: We will use the multimodal retrieval algorithm to calculate aerosol volume-vs-size distributions as a function of altitude (during aircraft vertical profiles) and distance (along horizontal transects) for four-wavelength optical depth spectra from AATS-6 in TARFOX and thirteen-wavelength spectra from AATS-14 in ACE-2. We will also explore whether this technique can be used to identify any dependence of the marine boundary layer volume distribution on air mass origin (urban/marine) by applying the algorithm to aerosol optical depth spectra measured during ACE-2 by the shipboard AATS-6 and the airborne AATS-14 and during TARFOX by the airborne AATS-6. We will also investigate the applicability of the algorithm to AATS-14 African dust optical depth spectra measured during ACE-2. As they become available, the results from Task 2 will be used to adjust the mode-dependent refractive indices used in all retrievals. We will evaluate the feasibility of automating an analogous non-linear least squares fitting procedure to determine whether more than three aerosol size distribution parameters can be retrieved from the thirteen-wavelength AATS-14 data set.

4. Size Distribution Comparisons and Closure Studies: We will extend the comparisons between sunphotometer-retrieved and in-situ-measured size distributions from the Pelican aircraft in ACE-2 (described by Schmid et al.,

1999) considering as many cases as possible from flights in the polluted and non polluted MBL and in the dust and non-dust laden FT. For the sunphotometer retrieval we will switch to a method that allows input of wavelength dependent refractive indices. In this comparison we will also use the inversion method developed in Task 3. This technique can provide a means of incorporating size-dependent refractive indices, and thereby accounting for observed size-dependent particle compositions. We will also compare results of our retrieved size distributions to models used in retrievals from imaging spectrometers on satellites and aircraft. One goal of these comparisons will be to identify potential improvements to the retrieval model size distributions and thereby the accuracy of the imaging spectrometer retrievals of optical depth.

5. Water Vapor/Aerosol Interactions and Satellite Retrievals: Extensive measurements of aerosol hygroscopic growth were performed in TARFOX and ACE-2, using TDMA and humidified nephelometers. Also, hygroscopicity was computed based on size-resolved chemical composition information. Subsequent, often not fully successful, closure studies (see Collins et al.; Livingston et al.; 1999, Schmid et al., 1999) have shown the limitations of measuring or modelling hygroscopicity. In an empirical approach we will correlate sunphotometer-derived extinction with sunphotometer-derived water densities during aircraft spirals flown in TARFOX and ACE-2. We will also consider trajectory studies and large-scale water vapor distributions (obtained from satellites) to investigate aerosol exposure to humidity prior to the sunphotometer measurement. Results of this task will be integrated with those of Task 2 above, in an effort to develop an integrated picture of aerosol hygroscopic growth in relation to chemical composition and optically measured or derived properties. We will also investigate the possibility of using water vapor information to improve satellite retrievals of aerosol optical depth and radiative forcing.

6. Flux Change/ Radiative Forcing Studies (extra not in proposal): We will analyze the solar radiant flux measurements from CIRPAS Pelican aircraft that were made during ACE-2. We will use a similar procedure that was successful in TARFOX (Russell et al 1999) to estimate the effective single scattering albedo of the aerosol. Of particular interest is the case where there is an elevated Saharan dust layer, July 17, 1997. Using the ACE-2 aerosol radiative properties, we will estimate the aerosol impact on the study region in a manner similar to Bergstrom and Russell (1999). We will use AVHRR determined optical depths from Durkee et al (1999) and explore using the ATSR-2 and OCTS satellite measurements to obtain optical depths.

FORM D: GACP BIBLIOGRAPHY

Name: Philip B. Russell

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- Schmid, B, Russell, P. B., J. M. Livingston, S. Gasso, D. Hegg, D. Collins, J. Seinfeld, E. Ostrom, K. Noone, P. Durkee, E. J. Welton, K. Voss, V. N. Kapustin, T. S. Bates, and P. K. Quinn, Clear column closure studies of urban-marine and mineral-dust aerosols using aircraft, ship, satellite and ground-based measurements in ACE-2. ARM Science Team Meeting, San Antonio, Texas, 22-26 March 1999.
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FORM E: GACP METRICS

Name: Philip B. Russell

Institution: NASA Ames Research Center

METRICS (do not include those in progress or planned):

- a. Number of publications (including books, book chapters, and refereed papers). 19.
- b. Number of printed technical reports and non-refereed papers.
- c. Number of oral or poster presentations at professional society meetings and conferences. 18.
- d. Number of advance degree students:
 - i. current
 - ii. graduated: Dr. Jens Redemann (PhD UCLA, June 1999; now working on this task as a scientist at Ames employed by Bay Area Environmental Research Institute).
- e. Number of post-doctorates. 0.
- f. Number and names of proposals:
 - i. submitted
 - ii. accepted
- g. Number of proposals and/or papers that you reviewed. 2 proposals, 3 papers.
- h. Number of committees served on (outside your organization). 2
- i. Number of patents (granted and applications).
- j. Number and names of honors and awards.