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7A.1 NORTH ATLANTIC AEROSOL PROPERTIES AND DIRECT RADIATIVE EFFECTS: KEY RESULTS FROM TARFOX AND ACE-2

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1. INTRODUCTION

Aerosol effects on atmospheric radiative fluxes provide a forcing function that can change the climate in potentially significant ways. This aerosol radiative forcing is a major source of uncertainty in understanding the observed climate change of the past century and in predicting future climate. To help reduce this uncertainty, the International Global Atmospheric Chemistry Project (IGAC) has endorsed a series of multiplatform aerosol field campaigns. The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) and the second Aerosol Characterization Experiment (ACE-2) were the first IGAC campaigns to address the impact of anthropogenic aerosols.

Both TARFOX and 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 [*Russell et al.,* 1999a], whereas ACE-2 studied aerosols carried over the eastern Atlantic from both European urban/industrial and African mineral sources [*Raes et al.*, 1999].

These aerosols often have a marked influence on the top-of-atmosphere radiances measured by satellites, as illustrated in Figure 1. Shown there are contours of aerosol optical depth derived from radiances measured by the AVHRR sensor on the NOAA-11 satellite. The contours readily show that aerosols originating in North America, Europe, and Africa impact the radiative properties of air over the North Atlantic. However, the accurate derivation of flux changes, or radiative forcing, from the satellite-measured radiances or retrieved optical depths remains a difficult challenge. In this paper we summarize key initial results from TARFOX and, to a lesser extent, ACE-2, with a focus on those results that allow an improved assessment of the flux changes caused by North Atlantic aerosols at middle latitudes.

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2. TARFOX MEASUREMENTS AND CALCULATIONS

TARFOX made coordinated measurements in July 1996 from four satellites (GOES-8, NOAA-14, ERS-2, LANDSAT), four aircraft (ER-2, C-130, C-131A, and a modified Cessna), land sites, and ships. Included were measurements of aerosol-induced flux changes, made simultaneously with measurements of the chemical, physical, and optical properties of the aerosols causing those changes.

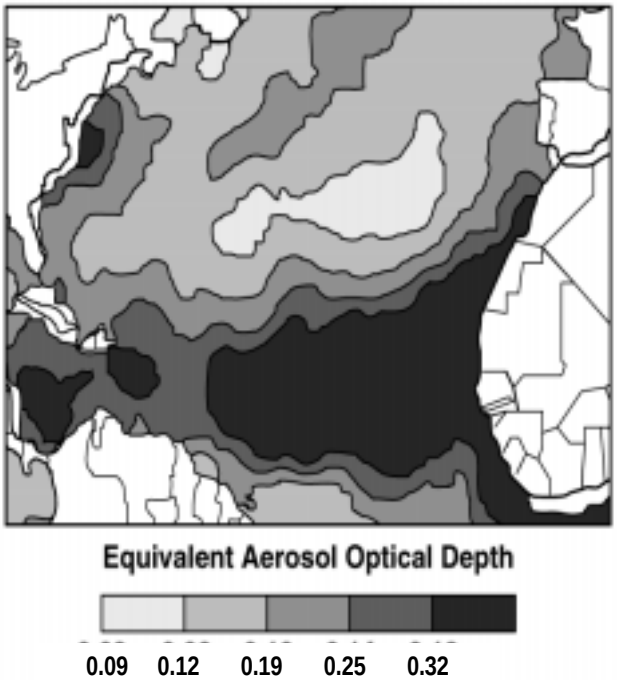

Figure 1. June/July/August map of aerosol optical depth derived from NOAA/AVHRR satellite reflectance data over the oceans [*Husar et al.*, 1997]

Key initial results from TARFOX include:

- Carbonaceous material (organic and inorganic) and sulfates accounted for essentially all (i.e., >90%) of the independently derived total dry aerosol mass in samples obtained at altitudes between 0.2 and 3.8 km [*Hegg et al.*, 1997].
- Averaged over the aerosol layer depth (typically ~3 km), carbonaceous material contributed 50% of dry aerosol mass and 66% of dry aerosol light scattering [*Novakov et al.*, 1997; *Hegg et al.,* 1997]. Both these fractions increased with altitude; near the surface carbonaceous mass fractions were ~10-35%, similar to some other results for the eastern U.S. and Canada [e.g., Trijonis, 1982; EPA, 1997; Andrews et al., 1998; McInnes et al., 1998; Turpin et al., 1998].
- The mean midvisible single-scattering albedo of the dry aerosol was 0.90, with a range of 0.76 to 0.98 for individual samples and 0.84 to 0.96 for vertical columns [*Hegg et al.,* 1997].
- The aerosol scattering humidification factor (ratio of light scattering at 80% relative humidity (RH) to that at 30% RH) at wavelength 550 nm averaged 2.1. It was larger (averaging 2.3) in westerly airflows (i.e., those from the continent) than in northerly and southerly airflows (which averaged 1.8). Values decreased with altitude and increased with wavelength [*Kotchenruther et al.*, 1999].
- Aerosol optical depths derived from in situ scattering and absorption profiles (using measured scattering humidification factors) averaged ~70-90% of airborne sunphotometer values [*Hegg et al.,* 1997; *Ross et al.,* 1998].
- Chemical apportionment of aerosol optical depths showed the dominant importance of water condensed on aerosol, with carbonaceous material second, and sulfate a close third [*Hegg et al.,* 1997].
- Remarkable year-to-year consistency was observed in the characteristics of the aerosol in the TARFOX region during episodes of heavy haze; at high aerosol loading the aerosol phase function at back scattering angles varied by less than 7-10% over a 5-year period including 1996, the TARFOX year [*Remer et al.*, 1999].
- Agreement was obtained between aerosol optical thickness magnitudes and wavelength dependences obtained by the MODIS Airborne Simulator (MAS) on the ER-2 and by sunphotometers on the C-131A and the surface [*Tanre et al.*, 1999].
- First validation of a dual-view algorithm that retrieves two-wavelength aerosol optical depth over land and ocean from radiances measured by the Along-Track Scanning Radiometer-2 (ATSR-2) on the European ERS-2 Satellite [*Veefkind et al.,* 1998, 1999].
- Agreement between shortwave radiative flux changes measured from aircraft and those calculated from measured aerosol properties, provided the calculations use midvisible single scattering albedos for the ambient (humidified) aerosol of ~0.89 to 0.93 [*Hignett et al.,* 1999; *Russell et al.*, 1999b]. An example of this agreement is shown in Figure 2. These single scattering albedo values are consistent with the independent measurements of the dry TARFOX aerosol particles by *Hegg et al.* [1997] and *Novakov et al.* [1997], cited above.

3. ACE-2 MEASUREMENTS AND PRELIMINARY CALCULATIONS

The ACE-2 intensive observation campaign was conducted in June-July 1997. Early results have been presented at special sessions of the IGAC conference in Seattle (August 1998) and the Fifth International Aerosol Conference in Edinburgh (September 1998); some are included in papers at this Conference [e.g., *Schmid et al.,* 1999]. These results show some similarities between ACE-2 and TARFOX results for the polluted marine boundary layer aerosol, as well as some significant differences. An example of a significant difference is:

- ACE-2 measurements typically yield smaller organic aerosol mass fractions than were obtained in TARFOX and in the other measurements on the U.S. and Canadian East coasts cited above.

Examples of similarities include:

- Sunphotometer-derived aerosol optical depths usually exceeded values derived from aircraft-measured profiles of scattering and absorption coefficients, humidification factor, and/or size distribution [e.g., *Schmid et al.,* 1999].
- Comparisons (e.g., *Durkee et al.*, 1998; Livingston et al., 1998; Schmid et al., 1999) between optical depths measured in ACE 2 with Ames sunphotometers and retrieved from satellite radiances show better agreement when African dust is absent than when dust is present. For example, four comparisons made with no dust present yielded a mean difference (AVHRR minus sunphotometer) for AVHRR Channel 1 (wavelength 640 nm) of 0.015, with standard deviation 0.030. For AVHRR Channel 2 (wavelength 840 nm) the mean difference was 0.025, with standard deviation 0.020. In contrast, for the single comparison to date with dust present, the differences were –0.03 and –0.08 in Channels 1 and

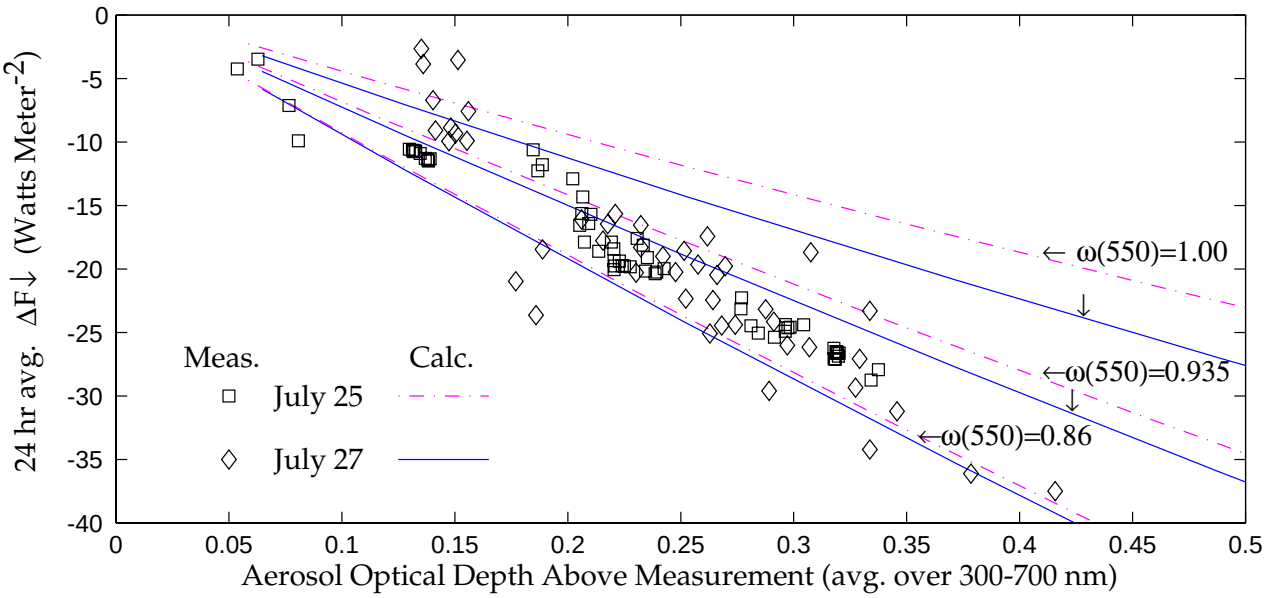

Figure 2. Comparison between aerosol-induced changes in shortwave downward flux determined from TARFOX C-130 measurements (data points) and calculations (curves) for size distributions retrieved from sunphotometer optical depth spectra for two days.

2, respectively (i.e., AVHRR values with dust present were less than sunphotometer values).

We have applied to ACE-2 data the methods used to compute the TARFOX flux changes shown in Figure 2. The results are preliminary, in that they use literature values for refractive indices of polluted marine boundary-layer and soil-dust aerosols, rather than values based on the ACE-2 size-resolved chemical composition measurements. However, these refractive indices from the literature do yield midvisible single-scattering albedos (~0.94) that are in the range measured for ACE-2 aerosols, both in the polluted marine boundary layer and in elevated African dust layers.

The preliminary calculations, which use ACE-2 measured spectra of aerosol optical depth (AOD), yield radiative flux sensitivity (ΔFlux/ΔAOD) values for ACE-2 aerosols (boundary layer and African dust) over the ocean that are similar to values for TARFOX boundary layer aerosols (e.g., $\Delta F\downarrow_{24\text{-hr}}$/ AOD(550 nm)≈-70 W m^{-2}).

4. NORTH ATLANTIC FLUX-CHANGE CALCULATIONS

Noting this similarity in flux sensitivities, especially for the polluted boundary-layer aerosols in TARFOX and ACE-2, we have used the aerosol optical model that yielded the agreement in Figure 2 to estimate shortwave radiative flux changes caused by aerosols over the North Atlantic Ocean [*Bergstrom and Russell*, 1999]. We exclude African dust from our calculations, primarily by restricting calculations to latitudes north of 25 N. Our calculations use the North Atlantic AOD values obtained over a two-year period from AVHRR radiances by *Husar et al.* [1997; cf. Figure 1]. We compute the net flux change at the tropopause for cloud-free and cloud-containing conditions for the winter, spring, summer and fall seasons as well as the annual average.

The local, cloud-free effects using the TARFOX derived aerosol properties range from –9 W/m^2 near the eastern US coastline in the summer to –1 W/m^2 in the mid-Atlantic during winter. The North Atlantic cloud-free regional averages range from –1.7 W/m^2 in the winter to – 5.1 W/m^2 in the summer. The cloud-free regional annual average is –3.5 W/m^2. The effect of changing the aerosol single-scattering albedo from our TARFOX-derived value of 0.9 (moderately absorbing) to 1.0 (no absorption) is to increase the flux change by about 30%. We used the ISCCP cloud data to compute the effect of clouds in the region. Inclusion of the cloudiness reduced the regional annual net flux change to –0.8 W/m^2 for the TARFOX aerosol and –1.1 W/m^2 for the non-absorbing aerosol.

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