North American, Asian, and Indian haze: Similar regional impacts on climate?

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[1] Pollution plumes recur seasonally downwind of the Indian subcontinent and Asian continent due to industrial and vehicular emissions, biomass burning, and wind-blown dust. These plumes have been well documented by field campaigns and satellite observations and the environmental implications of the "Asian Brown Cloud" have been widely publicized in a recently released UNEP report [*UNEP and C*^{4}, 2002]. Recent field experiments, however, demonstrate that the U.S. pollution plume can be as intense (in terms of aerosol mass concentration, aerosol optical depth, and ozone mixing ratio) as those downwind of India and Asia affecting regional climate along the U.S. East Coast. The use of identical sampling protocols in these experiments has been key in eliminating sampling biases and making the data directly comparable. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 1610 Global Change: Atmosphere (0315, 0325).
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1. Introduction

[2] Global distributions of aerosol optical depth (AOD) derived from satellite observations of backscattered radiation indicate persistent seasonal aerosol plumes downwind from many continental regions [*Husar et al.*, 1997]. In March through May a plume extends from Asia eastward across the North Pacific Ocean. During the winter monsoon period (December through April), a plume is located off the southwest coast of India over the Indian Ocean. In the summer months of June through August an aerosol plume originates from the mid-Atlantic states of the United States and extends across the North Atlantic Ocean. Although satellite observations show the seasonal regularity and relative magnitude of these plumes in terms of backscattered radiation they do not provide information about the aerosol chemical composition, optical properties, or mass loading in the boundary layer. Such information can be obtained from in situ measurements and is required to fully understand the regional climate effects of the haze. In situ measurements made during recent intensive field campaigns have characterized boundary layer aerosol properties in plumes emanating from the Indian subcontinent (INDOEX), Asia (ACE Asia), and the eastern U.S. (NEAQS).

2. Measurements

[3] During INDOEX, ACE Asia, and NEAQS, identical sampling protocols were used to measure submicron aerosol mass, mass concentrations of the major aerosol chemical components, aerosol scattering and absorption coefficients, AOD at a wavelength of 500 nm, and O_3 [*Quinn et al.*, 2002]. All in situ aerosol measurements were made at a constant RH of 55 ± 5% to maintain constant instrumental size cuts despite variations in ambient RH. The ambient RH encountered over all experiments ranged from 35 to nearly 100%. Here, submicron refers to all particles with aerodynamic diameters $\leq$1.1 um at 55% RH. We focus on submicron particulates as they are of the size range most likely to be transported over long distances and have the greatest radiative impact on climate in the visible wavelengths.

[4] Sampling for these three experiments was done onboard the NOAA Research Vessel *Ronald H. Brown*. The use of the same sampling platform as well as identical inlets, sampling humidities, aerosol samplers, and analysis methods makes the data from these three experiments directly comparable and, hence, avoids the introduction of sampling biases. We also include data from TARFOX, which took place off the U.S. East Coast, as further evidence of the intensity of the North American plume, even though different sampling protocols were used [*Hegg et al.*, 1997; *Novakov et al.*, 1997]. TARFOX was an aircraft study; we include only data collected at altitudes less than or equal to 300 m. The aerosol properties and ozone mixing ratios that are reported here for all experiments represent regional values since the platforms were moving within the study region rather than located at a fixed site.

[5] For INDOEX, ACE Asia, and NEAQS, sampling periods were categorized according to air mass history based on trajectories calculated for an arrival height of 100 and 500 m and an arrival position corresponding to the ship's location [*Draxler*, 1992]. ACE Asia samples were divided into three categories: Marine, Polluted, and Polluted with Dust. "Marine" encompasses the ship's transit from Hawaii to Japan when sampled air masses had not been in contact with land for 3 or more days prior to being sampled. It is included as a background reference. "Polluted" includes time periods when air masses had passed over Japan, Korea, and/or China less than 2 days before reaching the ship and had not passed over dust source regions. "Polluted with Dust" is similar to "Polluted" but has the additional constraint that the sampled air masses had passed

over dust source regions in northern China and/or Mongolia prior to reaching the ship. The ship did not encounter pure dust during ACE Asia. Using the same strategy, INDOEX sampling periods were categorized as "Arabia" or "India." Only one category is included for NEAQS; it includes time periods when trajectories indicated offshore flow from the region between New York and New Hampshire to the Atlantic. The TARFOX data are from periods of offshore flow as indicated by calculated air mass trajectories.

3. Results and Discussion

[6] It is recognized that the degree of variability presented here reflects only that which was measured during these short-lived campaigns. However, results from the 1999 INDOEX field campaign are expected to be representative of other years based on satellite data for 1996 to 2000 [*Ramanathan et al.*, 2001]. Seasonally (January to March) and spatially (0° to 30°N) averaged AOD for these five years varied by less than 10% of the 1999 values. Similarly, the summer mean values of AOD measured at Greenbelt, MD, a site in the mid-Atlantic region of the U.S., varied by less than 15% between 1993 and 1996 indicating that the 1996 TARFOX values were representative of other years [*Remer et al.*, 1999]. In addition, the regional mean AOD measured during NEAQS was within 18% of regional mean summer values derived from 5 Aeronet sites along the mid-Atlantic coast in 1993 and 1996 [*Remer et al.*, 1999]. Dust storms from northern China and Mongolia have increased noticeably in frequency and intensity during the past three years (http://www.usembassy-china.org.cn/sandt/estnews051702.htm). The dust storm sampled aboard the *Ronald H. Brown* during ACE Asia was a major event that extended across the Pacific, North America, and the Atlantic [*Huebert et al.*, 2003]. Hence, the ACE Asia time period may represent an upper bound in dust emissions.

[7] The distance of the measurements from the haze source may also lead to variability in the compared data sets. During NEAQS, measurements were made 20 to 100 km downwind of the eastern shore of the U.S. During ACE Asia measurements were made 100 to 400 km from the upwind shore and during INDOEX, 900 to 1400 km. In all instances, back trajectories indicated that the travel time from the upwind shore to the measurement platform was less than 2 days. The global estimate of the average lifetime of sulfate aerosol is about 5 days [*Langer and Rodhe*, 1991]. During INDOEX, the aerosol lifetime was estimated to be 7 to 8 days due to a lack of wet deposition [*Rasch et al.*, 2001]. Similary dry conditions during NEAQS and ACE Asia are expected to have led to equivalent lifetimes. Given short transport times relative to the aerosol lifetime and assuming minimal dilution of the air from mixing across the marine boundary layer inversion, the concentration of particulates in the plume should remain fairly constant 100 to 1400 km from shore. The uniformity of the aerosol concentration downwind of India is confirmed by satellite-derived average AOD for a two week period during INDOEX [*Collins et al.*, 2001]. Values ranged from 0.35 to 0.4 for the region extending westward from the coast of India to 68°E longitude. This region encompassed the cruise track of *Ronald H. Brown.*

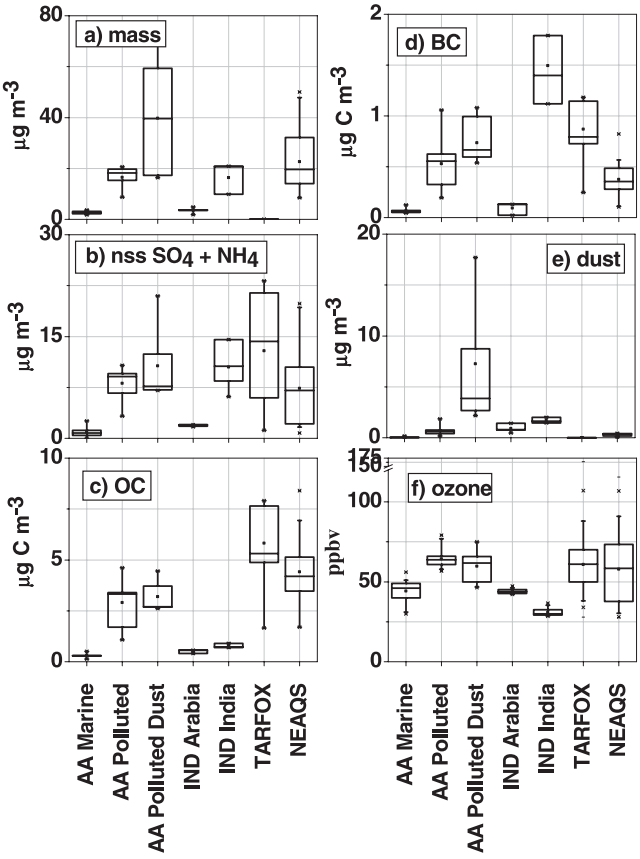

Figure 1. Concentrations of submicron aerosol mass, aerosol species, and ozone (1100 to 1700 local time). Horizontal lines denote the 25th, 50th, and 75th percentiles. Error bars denote 5th and 95th percentiles and symbols above and below the ends of the error bars indicate the 0th and 1st and the 99th and 100th percentiles. The solid square in the box denotes the mean.

[8] Concentrations of submicron aerosol mass and the dominant chemical components are shown in Figure 1 for INDOEX, ACE Asia, TARFOX, and NEAQS. The height of the marine boundary layer was highly variable within and between regions ranging from 0.25 to 3.5 km. Elevated ozone and particulate concentrations occurred irrespective of boundary layer height.

[9] Mean submicron aerosol mass concentrations were highest for the ACE Asia Polluted with Dust case (Figure 1a) where dust made up 22% of the fine mass (Figure 2). The NEAQS mean submicron mass concentration was about 40% higher than the ACE Asia Polluted and the INDOEX India cases. Fine particulate mass was not directly measured during TARFOX.

[10] The TARFOX mean sulfate aerosol (non-sea salt (nss) $SO_4^=$ + NH_4^+) concentration was about 20% higher than the INDOEX India and ACE Asia Polluted with Dust cases and 60% higher than the ACE Asia Polluted case (Figure 1b). The NEAQS mean concentration was 10 to 30% lower than these INDOEX and ACE Asia cases. However, NEAQS concentrations extended to higher levels by 40 to 90%.

[11] Even more striking differences are seen in the organic aerosol data. The TARFOX mean organic carbon (OC)

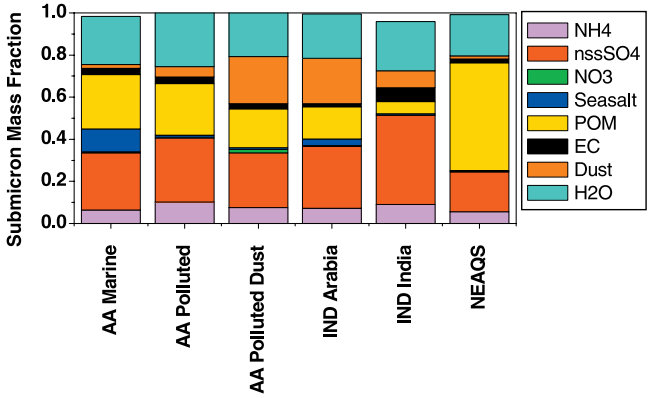

Figure 2. Average submicron mass fractions (MF) of the aerosol chemical species for ACE Asia, INDOEX, and NEAQS. MF are based on the gravimetrically-determined aerosol mass and concentrations of the chemical species. The MF of H_2O is that calculated to be associated with the ionic chemical species at 55% RH. POM is estimated by multiplying the measured concentration of OC by a factor of 1.6 to 2.1 to account for associated H and O [*Turpin and Lim*, 2001].

concentration (reported as μg C m^{-3}) was 1 to 7 times that measured in ACE Asia and INDOEX (Figure 1c) and the NEAQS mean concentration was 1.5 to 6 times higher. Particulate organic matter (POM) made up a larger fraction of the submicron aerosol mass during NEAQS (mean and 1σ standard deviation of 51 ± 19%; range of 18 to 80%) than during INDOEX (6 to 15%) or ACE Asia (18 to 24%) (Figure 2). POM was estimated to make up, on average, 50% of the dry aerosol mass during TARFOX [*Novakov et al.*, 1997].

[12] Mean BC concentrations were highest in the INDOEX India case and dust concentrations were highest in the ACE Asia Polluted with Dust case (Figures 1d and 1e). As a result, BC and dust have a greater influence on aerosol climate effects downwind of India and Asia, respectively, than downwind of the U.S. east coast.

[13] Ozone concentrations are compared for the 1100 to 1700 local time period when concentrations are expected to be highest (Figure 1f). Mean ozone concentrations were within 10% for TARFOX, NEAQS, and both ACE Asia Polluted cases. However, TARFOX and NEAQS concentrations extended to values that are a factor of 2 to 1.5 higher, respectively, than the ACE Asia Polluted cases. Low ozone concentrations off the Indian coast have been attributed to a high initial VOC/NO_x ratio and/or the absorption of ultraviolet radiation by absorbing aerosol [*Stehr et al.*, 2002].

[14] In terms of AOD, the eastern U. S. haze also can be as intense as the haze downwind of Asia and the Indian subcontinent (Figure 3). The mean AOD from TARFOX was within 3% of the two ACE Asia Polluted cases and 15 to 50% higher than both INDOEX cases. The NEAQS mean AOD was within 10% of both INDOEX cases but 20 to 25% lower than the ACE Asia Polluted cases. Maximum NEAQS values were 20 to 40% higher than the ACE Asia Polluted cases, however, and a factor of 2 higher than the INDOEX cases.

[15] As for the Asian and Indian subcontinent plumes, the aerosol plume emanating from the eastern U.S. has regional climate implications. Aerosol single scattering albedo (SSA) is the ratio of scattering by the aerosol to extinction (where extinction is a sum of the scattering and absorption by the aerosol) and thus indicates the absorptivity of the aerosol. SSA was derived from aerosol scattering and absorption coefficients measured at 55% RH. Using the simultaneously measured response of scattering by the aerosol to changes in relative humidity or f(RH) [*Carrico et al.*, 2003], the SSA was adjusted to ambient RH. SSA is shown for all experimental categories in Figure 3. The mean SSA and range of values observed during TARFOX, NEAQS, and ACE Asia were similar (≈0.94 ± 0.03). These values are higher than the INDOEX India case (0.88 ± 0.02) due to the higher mass fraction of BC in the INDOEX aerosol (Figure 2). Depending on the vertical distribution of SSA (the values in Figure 3 only apply to the lowest 300 m), the cloud fraction, and the surface albedo, this difference may lead to regional differences in the sign of the top of the atmosphere (TOA) forcing. It has been estimated that aerosols have a net negative (cooling) versus positive (warming) TOA forcing for SSA greater than 0.95 and less than 0.85, respectively [*Ramanathan et al.*, 2001].

[16] Similar diurnally averaged clear-sky surface forcings for an AOD of 0.3 have been reported for the aerosol plumes measured off the U.S. east coast (−19 to −23 W m^2 [*Russell et al.*, 1999]), the Indian subcontinent (−20 ± 4 W m^2 [*Ramanathan et al.*, 2001]), and Asia (−20 to −25 W m^2 [*Bergstrom et al.*, 2002]). These large negative surface forcings indicate a loss of shortwave radiation within the plume and a cooling of the underlying surface. Hence, the direct clear sky radiative effect of aerosols appears to be as large for the eastern U.S. plume as for plumes emanating from Asia and the Indian subcontinent. The mechanism of the forcing may not be the same in all regions, however, due to different SSA.

4. Conclusions

[17] The pollution plume downwind of the east coast of the U.S. is comparable in aerosol mass loading, aerosol

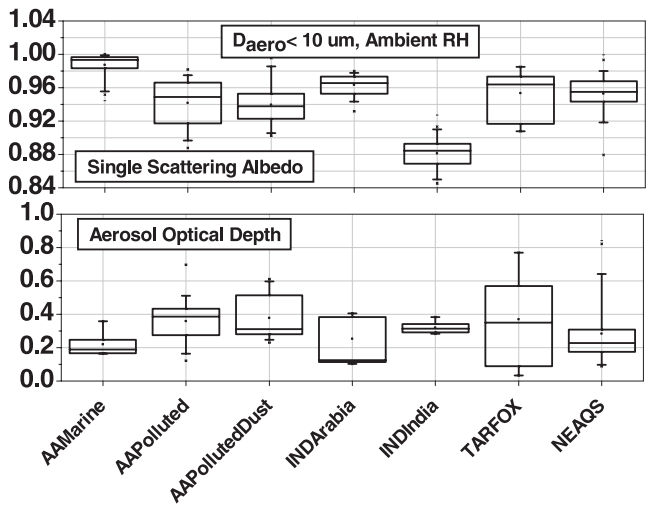

Figure 3. AOD (500 nm) and SSA (550 nm, ambient RH) for all experiments. Percentile information is the same as for Figure 1.

optical depth, and ozone mixing ratios to plumes downwind of the Indian subcontinent and Asia and, hence, deserves similar attention in determining its climate impacts. The aerosol chemical composition within the eastern U.S. plume is unique, however, with a mass fraction of POM (18 to 80%) higher than that observed in the Asian plumes (6 to 24%) (Figure 2). For all experiments, the OC analysis method that was used only quantifies total organic carbon and does not identify individual organic species. Hence, it is not possible from these data to determine if the organic material was of biogenic and/or anthropogenic origin. Speciation of the organics is an essential next step in determining sources, climate impacts, and possible strategies for reducing emissions. Even though the data presented here do not show a direct link to health effects, the high aerosol mass and ozone concentrations within the eastern U.S. plume indicate that impacts on health may also exist. Speciation of the organics also will help in determining their toxicity and, therefore, health effects.

[18] To aid policy actions, year-round measurements of ozone and aerosol properties close to the coast of New England and the mid-Atlantic states both at the surface and aloft are needed to assess the variability and long term trends of the pollution haze. Shorter-term intensive field experiments are needed to link the emissions of precursor species with the transport, transformation, and fate of ozone and aerosols and thereby improve our capabilities to forecast atmospheric species concentrations under different emission scenarios.

[19] The pollution plumes that are clearly discernable downwind of continents can undergo intercontinental transport to distant regions. Model estimates indicate that North American anthropogenic emissions enhance surface ozone in Europe by 5 to 10 ppbv during transatlantic transport events [*Li et al.*, 2002]. As a result, regional air quality and climate issues become hemispheric or, in some cases, global issues. International cooperation is thus critical to understanding and mitigating these environmental issues.

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