

Atmospheric brown clouds: Hemispherical and regional variations in long-range transport, absorption, and radiative forcing

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[1] The study uses satellite observations, global assimilated aerosol data sets, Atmospheric Brown Clouds (ABC) observatories, a Monte Carlo aerosol-cloud-radiation model and a regional chemical transport model (STEM-2K) to characterize the spatial extent of brown clouds, regional and megacity ABC hot spots, chemical composition and the direct radiative forcing. It presents the first annual cycle of aerosol observations and forcing from the ABC observatories in the Indo-Asia-Pacific regions. East Asia, Indo-Gangetic Plains, Indonesian region, southern Africa and the Amazon basin are the regional hot spots defined by the criteria that anthropogenic aerosol optical depths (AODs) should exceed 0.3 and absorbing AOD > 0.03. Over these hot spots, as well as in other polluted oceanic regions, the EC mass exceeds 0.5 μ g m⁻³, the OC mass exceeds 2 μ g m⁻³ and sulfate mass exceeds 10 μ g m⁻³ from the surface to 3 km. The brown clouds also have strong seasonal dependence. In the tropics the seasonal dependence is driven by pollution accumulating during the dry seasons, December to February in Northern Hemisphere tropics and June to August in Southern Hemisphere tropics. In the extratropics the pollution peaks during the summer. The brown cloud problem is not restricted to the tropical regions. Over the eastern half of US and western Europe the AODs exceeds 0.2 and absorption AODs exceed 0.02. Brown clouds also extend well into the western Pacific Ocean, the Indian Ocean reaching as far south as 60° S and the eastern Atlantic Ocean. The largest total SO_2 emission occurs over China and US, while SO_2 emission per unit surface area is maximum over Germany and England. The largest total EC and OC emissions occur over China, but the largest OC emission per unit surface area occur over India. As a result, the maximum negative annual mean TOA direct forcing is over India and Germany. The surface annual-diurnal mean dimming over the regional hot spots is of the order of -10 W m^{-2} and -20 W m^{-2} over megacity hotpots.

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1. Introduction

[2] The Indian Ocean Experiment (INDOEX) (V. Ramanathan et al., Indian Ocean Experiment (INDOEX) White Paper, available at http://www-indoex.ucsd.edu/publications/ white_paper/) led to the discovery of the brown clouds over

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south Asia as a transcontinental and transoceanic problem. Brown clouds generally denote anthropogenic aerosols that not only scatter but also absorb solar radiation due to soot and other absorbing aerosols. The concept of brown clouds itself was established decades before the INDOEX study, but it was largely thought of as an urban phenomenon (e.g., the Denver brown cloud). We have adopted the term "brown cloud" not necessarily to signify highly polluted regions of the planet, but simply to denote absorbing anthropogenic aerosols that absorb solar radiation in addition to scattering it. Anthropogenic aerosols are defined as particulates originating from human activities at a rate not normally observed in nature. In addition, the conventional definition of "brown cloud" applies mainly to surface level pollution related particulates, whereas the term "brown cloud" in this study applies to aerosol over the entire troposphere.

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[3] The main findings of INDOEX were as follows:

[4] 1. The brown clouds over the Arabian Sea and Bay of Bengal [*Rajeev et al.*, 2000; *Ramanathan et al.*, 2001a] were widespread in nature; in addition, gaseous pollutants such as ozone and CO were also widespread [*Mandal et al.*, 1999; *Gupta et al.*, 1999; *de Gouw et al.*, 2001; *Lelieveld et al.*, 2001; *Rhoads et al.*, 1997].

[5] 2. Man-made aerosols in the brown clouds led to a major redistribution of solar radiation by enhancing the solar heating of the atmosphere and causing a large reduction of solar radiation at the surface [*Satheesh and Ramanathan*, 2000; *Ramanathan et al.*, 2001a, hereinafter referred to as R2001a]. The large reduction at the surface was concentrated mainly in the visible parts of the solar spectrum [*Jayaraman et al.*, 1998; *Meywerk and Ramanathan*, 1999].

[6] 3. The surface dimming and atmospheric solar heating were widespread and influenced the radiation budget and forcing of the entire Arabian Sea and the Indian sub continent [R2001a]. For example, averaged over the entire Indian Ocean north of the equator, the seasonal averaged surface dimming was as large as -13 W m⁻² and atmospheric solar heating was enhanced by 25% to 50%.

[7] 4. Chemical characterization of the aerosols [*Reiner et al.*, 2001; *Guazzotti et al.*, 2001] led to the conclusion [R2001a] that roughly 75% of the aerosols were of anthropogenic origin.

[8] 5. The man-made aerosols led to a three fold increase in the number of cloud drops in the Arabian Sea when compared with the pristine south Indian Ocean [*Heymsfield and McFarquhar*, 2001].

[9] 6. At the TOA, the direct forcing was negligible but the seasonal mean negative indirect forcing was as large as -5 W m^{-2} [R2001a].

[10] 7. Because of their widespread nature, the brown clouds exerted a large north-south heating gradient in the aerosol radiative forcing with a large cooling of the Arabian Sea [R2001a] and minimal cooling south of the equator. Additional INDOEX observations on the vertical and spatial variations in aerosol properties over the Indian Ocean are given by Moorthy et al. [2001], Müller et al. [2001], Neusüß et al. [2002] and Quinn et al. [2002]. Together these findings linked air pollution, aerosols and brown clouds as strong forcing agents for regional climate and water budget [Ramanathan et al., 2001b, hereinafter referred to as R2001b]. These regional climate implications and their effects on the monsoon were explored in several modeling studies [Chung et al., 2002; Menon et al., 2002; Krishnan and Ramanathan, 2002; Ramanathan et al., 2005; Chung and Ramanathan, 2006].

[11] Subsequent to the completion of INDOEX in 2000, two major developments established atmospheric brown clouds as a major global problem. First is the retrieval of aerosols over land and oceanic areas by the MODIS instrument on Terra satellite in 2001, which revealed brown clouds (originating from major populated regions of the world) over the Atlantic, the Pacific and the Indian oceans [*Kaufman et al.*, 2002; *Ramanathan and Ramana*, 2003]. The second development was spurred by another field experiment, ACE-Asia [*Huebert et al.*, 2003; *Schauer et al.*, 2003], in the western Pacific Ocean downwind of east Asia. The results were similar to INDOEX in that it revealed

widespread pollution (mixed with dust) in the western Pacific Ocean with large regional radiative forcing [*Conant et al.*, 2003], very similar to the values revealed by INDOEX.

[12] It is in recognition of these major findings that UNEP established the Atmospheric Brown Clouds project, based on the regional paradigm proposed by Ramanathan and Crutzen [2003]. Details of this regional paradigm and the structure of the new project are given in the ABC website (http://www-abc-asia.ucsd.edu). While the brown cloud problem is common to all areas of the world, the first focus of the ABC project is on Asia, home to about 60% of the world's 6+ billion population. As part of ABC-Asia, a regional observatory system is being developed for the Indo-Asia-Pacific Region (observatory locations are shown later). This paper focuses on the ABC observatories in the Arabian Sea, Nepal, Gosan Korea and Trinidad Head in North America and reports the first continuous annual cycle of observations from the Maldives Climate Observatory (MCO). We will first (in section 3) describe new results on the global perspective of the brown cloud problem which provides the context for the regional and the local issues discussed later.

2. Methods

[13] Figure 1 presents a schematic of the approach presented in this paper to integrate observations of processes (Figure 1, left) with the regional-scale parameters obtained from satellites and regional to global models (Figure 1, right), using the Monte Carlo Aerosol Cloud Radiation model (MACR). The goal is to provide an observationally constrained estimate for the direct, the indirect and the semidirect forcing (Figure 1, middle). An earlier version of this procedure was tested successfully with INDOEX observations [R2001a]. It has been further modified and improved to take advantage of new satellite capabilities for aerosol measurements and new aerosol-transport modeling on global and regional scales. The modified scheme is shown in Figure 1 and the various components are described below:

2.1. MACR

[14] The central component (Figure 1, middle) is MACR, which adopts aerosol-cloud-radiation parameterizations from the observationally based process studies and the regional parameters from satellite measurements and estimates the radiative forcing on local to regional scales. MACR was developed by our group [Podgorny et al., 2000; Podgorny and Ramanathan, 2001; Vogelmann et al., 2001] and applied in INDOEX. It is essentially a radiative transfer model that employs parameterizations of aerosol radiative and cloud properties that are consistent with the chemistry of aerosols. The parameterizations are based on process studies (Figure 1, left) using data collected from the ABC observatories (ABCO; see sections 3 and 4.1) and field campaigns (INDOEX, EAREX, APMEX, and MAC described next). These parameterizations are used in conjunction with regional-scale aerosol and cloud parameters (Figure 1, right) to estimate the regional-scale aerosol forcing (Figure 1, middle) on regional scales.



Figure 1. A schematic of the procedure adopted for estimating the direct, indirect and semidirect aerosol forcing. It is an improved and modified version of the scheme shown by *Ramanathan et al.* [2001a]. MACR is Monte Carlo Aerosol-Cloud Radiation Model. ABCO is ABC observatories. INDOEX, EAREX, APMEX and MAC are field campaign conducted by the ABC project in the Arabian sea and east Asia. STEM-2K and GOCART are the regional and global aerosol chemical transport models, respectively. ISCCP, CERES, MODIS and MISR are satellite data for cloud and aerosol properties.

2.2. Process Studies (Figure 1, Left) for Aerosol Forcing Estimates on Local Scales

[15] The key components are the ABCOs and in situ aircraft observations from field campaigns starting with INDOEX [R2001b; R2001a], and the campaigns organized by the ABC project (http://www-abc-asia.ucsd.edu/ fieldcampaigns.htm): EAREX was held in 2005 at the ABC supersite in Gosan. South Korea: APMEX was held in October 2004 at the ABC supersite in Hanimadhoo, Republic of Maldives, and the MAC campaign of March 2006, which deployed stacked unmanned aircraft vehicles (UAVs) with miniaturized aerosol-cloud-radiation instruments (http://www-abc-asia.ucsd.edu/MAC/secure/Index.htm). The field campaigns are critical for developing model parameterizations for the aerosol-cloud interactions. The data collected during APMEX were described by Ramana and Ramanathan [2006] and Corrigan et al. [2006]. As reported in Ramana and Ramanathan [2006], the MACR parameterizations developed with INDOEX data are in good agreement with ABCO data collected at the Maldives and

hence were unchanged from the descriptions given by R2001a. This study presents for the first time the annual cycle from ABCOs. The relevant parameters that are extracted from ABCOs include annual cycle of the size distribution of aerosols (section 4.1), their chemical composition (section 4.2), direct radiative forcing at the surface (section 4.3) and a scheme for the so-called indirect effect of aerosols, i.e., a parameterization that links aerosols to CCN and subsequently the fraction of CCN that are activated into cloud drops (see R2001a for details). The observatory data for aerosol chemical compositions are needed to partition the observed aerosols into natural and anthropogenic fractions which are critical for deriving the anthropogenic forcing. The partitioning of the observed forcing into anthropogenic fraction was undertaken for the INDOEX study [R2001a], but has not been attempted as yet for the ABCO sites. Lastly the observatory time series are the only source of data for determining interannual to decadal-scale variations in aerosol anthropogenic concentrations and surface dimming.

2.3. Regional-Scale Parameters (Figure 1, Right)

[16] To estimate regional-scale forcing for all of Asia and the adjacent Indian and Pacific Oceans, we need satellite data for clouds and aerosols. The key cloud parameters are cloud fraction and cloud optical depths as a function of altitude and these are obtained from the International Satellite Cloud Climatology Project (ISCCP; http://isccp. giss.nasa.gov/) and Clouds and the Earth's Radiation Energy System (CERES, http://asd-www.larc.nasa.gov/ceres/). The ISCCP data is used when longer-term (before 2002) data are needed whereas the more accurate CERES data are used for recent years (from 2002 onward). Regional-scale aerosol optical properties are obtained from the MODIS (Moderate Resolution Imaging Spectroradiometer; http:// modis.gsfc.nasa.gov/) and MISR (the Multiangle Imaging SpectroRadiometer; http://www-misr.jpl.nasa.gov/) instruments on TERRA and AQUA satellites. In addition, regional surface based AERONET (Aerosol Robotic Network, http://aeronet.gsfc.nasa.gov/data frame.html) stations (over 100 around the world) data for single-scattering albedos (SSAs) are used in the model estimates of the forcing.

2.4. Assimilation Technique for Regional and Global Aerosol Properties

[17] The results presented in section 3 on identifying aerosol hot spots are based on a new aerosol-assimilation scheme developed by our group [Chung et al., 2005]. This assimilation technique uses a combination of recent satellite aerosol data, surface aerosol network observations, aerosol chemical-transport model and MACR. For ground-based data for aerosol parameters (aerosol optical depth (AOD), single scattering albedo (SSA) and asymmetry factor) we use the quality assured level 2.0 data from the AERONET stations around the world [e.g., Dubovik et al., 2000, 2002; Holben et al., 2001]. For satellite aerosol data, we use MISR AODs [e.g., Diner et al., 1998; Kahn et al., 2001, 2005] and MODIS AODs [e.g., Kaufman et al., 1997; Tanré et al., 1997; Remer et al., 2005]. Additional details of the assimilation technique are described by Chung et al. [2005], but the technique basically involves using satellite data where available and filling in the gaps with AERONET stations to map out global monthly mean distribution of AODs. For the single scattering albedos it adopts an objective optimal interpolation scheme [see Chung et al., 2005] for integrating AERONET SSAs with the GOCART(see Figure 1, right) model (Goddard Chemistry Aerosol Radiation and Transport) described by Chin et al. [2002].

[18] The anthropogenic fraction of the derived AOD (resulting from human activity) is obtained from the GOCART model. The anthropogenic fraction estimated by the GOCART model was in agreement (within 15%) with the INDOEX observations [R2001a]. The data-model assimilation was done for a 3-a period from 2001 to 2003, since comprehensive satellite and ground-based aerosol data were available only for this period.

2.5. Regional Aerosol Chemical-Transport Model (STEM-2K)

[19] The regional simulations of aerosol chemistry and optical properties for India are shown in section 5 and the

model used for these simulations is described here. Chemical transport models (CTMs) provide a means to estimate four-dimensional aerosol distributions and aerosol forcing based on estimates of emission distributions. Linking emissions to aerosol distributions is essential to attribute aerosol forcing effects to specific aerosol components and to provide policy makers with the information needed for management of atmospheric composition. CTMs are also important tools for the interpretation of observational data. One of the goals of the ABC project is to utilize the observational data to test and ultimately improve the predictive skills of CTMs. Toward these ends, aerosol distributions were estimated using the STEM (Sulfur Transport and Deposition Model) model [Carmichael et al., 2003; Tang et al., 2004]. For this study the MM5 mesoscale meteorological model using NCEP reanalysis fields for initialization was used for driving the aerosol transport [Grell et al., 1995]. The model was run from 1 August 2004 to 31 August 2005 to analyze the seasonal distribution. The modeling domain covered from 40°E to 140°E longitude and from 21°S to 50°N latitude, with a horizontal spatial resolution of 50×50 km, and with 23 vertical layers extending to a model top of 14 km.

[20] Fossil fuel and biofuel emissions of Streets et al. [2003] for the portion of the model domain in the Asia-Pacific region and the Emission Database for Global Atmospheric Research (EDGAR) [Olivier and Berdowski, 2001] for the rest of the domain were used. Biomass burning emissions were taken from the climatological values from Reddy and Boucher [2004]. Sea salt and dust emissions in the STEM model were calculated online on the basis of the parameterizations of Gong [2003] and Tang et al. [2004] respectively. The STEM predictions for EC, OC, sulfate, other primary particulates (such as mineral particles from cement manufacturing and fugitive dust from roadways and construction) in particle diameters less than 2.5 μ m and between 2.5 and 10 μ m (referred to as PM_{2.5} and other PM₁₀, respectively), along with wind blown dust and sea salt in fine and coarse modes, were calculated and compared to the observations. Further details regarding the model are given by Adhikary et al. [2007]. The version of the STEM model used in this study is referred to as STEM-2K, and as implied earlier, it is used as a forward model to simulate aerosol properties and forcing with meteorological fields and emission of aerosol precursors as the model input.

2.6. Derivation of Aerosol Forcing

[21] Once the observatory data are used to develop and validate the MACR model and the STEM-2K model, then aerosol chemical input from STEM-2K are integrated with satellite derived aerosol AODs and cloud properties (cloud fractions, altitudes and optical depths) to derive aerosol direct and indirect forcing. This paper, however, presents only simulations of aerosol chemical and optical properties from STEM-2K. Aerosol forcing simulations from STEM-2K will be taken up later. Next is the so-called semidirect forcing which deals with the impact of aerosol induced solar heating on cloud fraction and the aerosol influence on cloud life time (so called second indirect effect). These are obtained from a cloud resolving model (Figure 1, bottom right) the input for which is provided by the ABC observatory data. This approach was adopted by us [*Ackerman et*]



Figure 2. Calculated global distribution of anthropogenic aerosol optical depths, in the Northern Hemispheric (a) dry season, December–March; (b) transition, April–June; (c) monsoon, July–August; and (d) transition, September–October 2001–2003. Anthropogenic AODs are derived from our previous work [*Chung et al.*, 2005].

al., 2000] using INDOEX observations to derive the semidirect forcing due to soot induced solar heating. This study, however, is restricted to estimates of direct forcing.

[22] In what follows, section 3 presents regional to global-scale aerosol properties from the assimilation technique to identify regional and megacity aerosol hot spots. It then undertakes a comparative study of AODs and aerosol forcing with the emissions of aerosol precursors. This section provides the large-scale perspective to understand the local-scale aerosol chemical and physical properties and forcing over the ABCOs presented in section 4 along with a brief description of the locations and content of ABCOs. Lastly section 5 describes the regional simulations of the aerosol chemistry and optical properties for the Indo-Asia-Pacific region. The section also includes a validation of the simulations with observations from ABCOs and the assimilated regional data.

3. Identifying Regional and Megacity Aerosol Hot Spots

[23] Global distribution of anthropogenic aerosol optical depths are shown (Figure 2) for specific seasons, to highlight major regimes of ABCs around the planet. The *Chung et al.* [2005] study presents global maps of the forcing as

well as hemispherical and global averages of the forcing. In what follows, we show selected results (relevant to this paper) concerning the regional distribution of the brown clouds, since these were not shown in the Chung et al. [2005] study. The anthropogenic fraction of the total AOD is obtained from the GOCART model as described by Chung et al. [2005]. It should be noted that the interest in this study is on anthropogenic aerosol radiative forcing for which the aerosol properties in the entire tropospheric column is of importance. Hence, when we refer to pollution and brown clouds, we are not necessarily discussing surface concentrations of particles and haze (which are the domain of most particle pollution studies). The word "pollution" in this study refers to anthropogenic aerosols from surface to about 12 km, the region which contributes to the satellite derived AOD.

[24] Starting from North America (in Figure 2), a plume of brown clouds extends from the east coast across the Atlantic Ocean toward Europe; likewise, the European plume spreads toward central Asia; from east Asia another plume spreads across the Pacific Ocean; from south Asia and southern Africa the brown clouds are widespread across the Indian Ocean; biomass burning aerosols from Brazil also spread into the western South Atlantic. The brown clouds also have strong seasonal dependence. In the tropics





Figure 3. Photographs of atmospheric brown clouds from regions included in this study.

the seasonal dependence is driven by pollution accumulating during the dry seasons (December to February in Northern Hemisphere tropics and June to August in Southern Hemisphere tropics) and is primarily affected by meteorological influences. Additionally, in the extratropics the pollution peaks during the summer because of the seasonal dependence of emission and the dependence of aerosol chemistry on temperature and solar UV radiation. Starting first with the North Atlantic Ocean, transport of the pollutants happens through out the year with relatively large AODs occurring during spring, summer and fall seasons. The winter minima is due to the fact that AODs over the source regions in North America are low during this season. The north-south extent of the plume is widespread in the Indian Ocean, with the northern ocean influenced by pollution from south Asia, Saudi Arabia, Middle East and North Africa, while the southern Indian Ocean is subject to pollution from biomass burning in southern Africa. The brown clouds extend almost up to 60°S in the Indian Ocean during the austral dry season (see July to August panel). The Pacific Ocean brown clouds are extensive in the east-west direction, particularly during the spring and fall seasons, when anthropogenic AODs exceed 0.1 in the eastern Pacific Ocean (east of the date line).

[25] The main feature that characterizes the brownish color (see photos in Figure 3) is the presence of absorbing aerosols, particularly black and organic carbon (see *Andreae and Gelencser* [2006] for a review of absorption by carbonaceous aerosols). In order to get an overview of the global distribution of this key ingredient of brown clouds, we show the absorption optical depth in Figure 4. The anthropogenic AOD (shown in Figure 2) is the sum of absorption AOD (shown in Figure 4) and scattering optical depth (mostly

from sulfates, nitrates, organics). The absorption AOD (Figure 4) is mostly due to soot (i.e., sum of elemental carbon, and other combustion residues). The overall regional pattern in Figure 4 is similar to that shown in Figure 2. In the Southern Hemisphere, the major soot (absorbing aerosol) hot spots (yellow and red regions in Figure 4) during the dry seasons (see July to October panels) coincide with regions of biomass burning in South America and southern Africa. From Figures 2 and 4, we identify brown cloud hot spots, defined as regions with anthropogenic AOD > 0.3and absorbing AOD > 0.03. These are the yellow and red shaded regions in both Figures 2 and 4. These two criteria have to be satisfied at least for one season for a region to be considered as a hot spot. Typically, the number concentrations of aerosols and soot within the troposphere have to be larger than the hemispherically averaged values by a factor of 3 to 4, for AODs to exceed 0.3 (0.03 for absorbing AODs). The following regions fall under the hot spot category as defined in this study: (1) east Asia (eastern China, Thailand, Vietnam and Cambodia), (2) Indo-Gangetic Plains in south Asia (the northwest to northeast region extending from eastern Pakistan, across India to Bangladesh and Myanmar), (3) Indonesian region, (4) southern Africa extending southward from sub-Saharan Africa into Angola and Zambia and Zimbabwe, and (5) the Amazon basin in South America.

[26] The regional hot spots in Asia (identified from assimilated data sets) also show as highly polluted regions with sulfates, elemental carbon and organic carbon simulated by the regional chemical transport model (see section 5 for results from STEM-2K), providing another justification for the categorization recommended in this study. However, the brown cloud problem is not just a tropical issue, since



Figure 4. Same as Figure 2 except for anthropogenic absorbing aerosol optical depth [*Chung et al.*, 2005].

even in industrialized regions, absorbing aerosols are prevalent. For example, over the eastern half of US and western Europe the total (scattering and absorbing) AOD exceeds 0.2 and absorbing AODs exceed 0.02. In summary, Figures 2 and 4 clearly establish the highly regional nature of the brown cloud problem, with strong east-west and north-south gradients in aerosol concentrations, aerosol properties and aerosol forcing of climate. We next focus on the potential sources of these asymmetries. Figure 5 shows the annual mean emissions of SO₂, EC and OC (organic carbon) as total emissions (Figure 5a) and as emissions per unit area (Figure 5b), annual mean anthropogenic AOD (Figure 5c) and direct radiative forcing at TOA (Figure 5d), the atmosphere and surface (Figure 5e). The anthropogenic AOD is obtained from Chung et al. [2005] using the GOCART model (see section 2) for the anthropogenic fraction. These factors are seasonally dependent, and to illustrate this feature, Figure 6 shows AODs and forcing for the season when AODs attain peak values. Both the AODs and the forcing were obtained from the assimilation technique of Chung et al. [2005]. The important findings are given below.

[27] 1. India presents an interesting contrast to China and US values. Although its emission of SO_2 is a factor 3 lower than that of US and a factor of 4 lower than that of China (Figure 5a), the annual mean anthropogenic AOD over India is a factor of two larger than that of the US and about

the same as over China (Figure 5c). In addition, the largest (among the regions shown in Figures 5 and 6) annual mean TOA aerosol cooling occurs over India (-1.4 W m^{-2}) (Figure 5d) and the largest annual mean surface dimming (-14 W m^{-2}) also occurs over India (Figure 5e). A similar qualitative picture emerges from the seasonal maxima shown in Figure 6. Several factors contribute to the large response over India. One factor is emissions of EC and OC. Although SO₂ emissions from India are smaller, the EC + OC emissions from India are larger than that of US, but this by itself cannot account for the factor of two larger AOD over India compared with US. Another factor is geography. Note first that the emission per unit area (the critical factor for AOD) presents a much different picture (Figure 5b). The SO₂ emission is almost the same compared with US but the emissions (per unit area) of EC and OC are factors of 5 to 10 larger. This can account for some of the difference in AOD and the forcing but still cannot account for the factor of 3 difference in the anthropogenic AOD between US and India. Meteorology is another factor. The long dry season over India (November to April) accompanied by persistent subsidence resulting in trade wind inversions at about 3 km. This 4 to 6 month long synoptic weather inhibits ventilation of the pollution. This should be contrasted with US and China where the mid-extratropical westerlies are an efficient way to export the pollution. In addition, another contributing factor is the near absence of precipitation during the dry



Figure 5. A comparative study of aerosol emissions, anthropogenic AODs and anthropogenic forcing for various regions. (a) Annual mean emissions of SO₂ for 1995–1999 from *Smith et al.* [2004] and the elemental carbon (EC) and organic carbon (OC) data for 1997 from *Bond et al.* [2004]. FF denotes fossil fuel, BF denotes biofuel and BB denotes biomass burning. (b) Same as Figure 5a but for emissions per unit surface area. (c) Annual mean anthropogenic aerosol optical depths at 550 nm for 2001–2003, (d) annual mean anthropogenic aerosol forcing at the top of atmosphere (TOA) for 2001–2003, and (e) annual mean anthropogenic aerosol forcing for the atmosphere (ATM) and at the surface for 2001–2003. The forcing values are for direct forcing only.

season, which extends the life time of aerosols. As supporting evidence, we see from Figure 6 that the dry season anthropogenic AOD (0.35) for India is nearly a factor of 2.5 larger than the annual AOD. The third major factor is the difference in the role of aerosols from long-range transport. The US is surrounded by the relatively cleaner Pacific and Atlantic, where as India is surrounded by other land areas with large sources of pollution.

[28] 2. Even in industrialized and developed nations (US, UK and Germany) the annual average aerosol AOD is as much as 0.1, with summer maxima of about 0.2. However, although their emission of aerosol precursors are smaller (than other regions in Figure 5) the per unit area emissions of pollutants over Germany and UK are the largest of the regions shown in Figure 5. Thus it is not surprising that the anthropogenic AODs are large and the TOA forcing over Germany is almost as large as over India. Given strict pollution controls and reduced biomass burning, we would

anticipate smaller AODs over these regions, although Europe does employ a high concentration of diesel powered transportation.

[29] 3. With respect to the forcing, we clearly see the nonlinear dependence of the forcing on AOD. China has the largest total anthropogenic AOD, but the least TOA forcing, whereas India and Germany are subject to large cooling (negative radiative forcing). In order to understand this trend, we must look at the atmospheric and surface forcing. In all regions shown in Figures 5 and 6, large dimming at the surface is nearly balanced by comparably large atmospheric solar heating. Thus the TOA forcing (sum of surface and atmospheric heating) is a delicate balance of two competing effects. Let us compare India and China to provide some insights into this delicate balance. First, the surface dimming over China is smaller than that in India (in spite of AOD and absorbing AOD being about the same, see Figure 5) because TOA solar irradiance is larger over India



Figure 6. Anthropogenic AODs and forcing of various regions for the season with maximum AODs. (a) Anthropogenic aerosol optical depths, (b) the anthropogenic aerosol forcing at TOA, and (c) the anthropogenic aerosol forcing in the atmosphere and at the surface for the high AOD seasons over the United States (June–July–August), India (January–February–March), China (July–August–September), United Kingdom (May–June–July), Germany (July–August–September), and the former Soviet Union (July–August–September) 2001–2003.

(since it is at lower latitudes than China). Next, since China is at higher latitudes, its solar zenith angle is larger (more slant) and as a result the slant path for absorption is larger; in addition the surface albedo is also larger. Both of these will tend to enhance solar absorption by absorbing aerosols. As a result, the atmospheric positive forcing nearly cancels out the surface dimming over China.

[30] 4. We also note that the dimming (reduction of solar radiation) is substantial even in industrialized regions (-3 to) -5 W m^{-2}) and exceeds -10 W m^{-2} over India and China. [31] 5. The maximum AOD and forcing is not realized during the same season or month for each region, as shown in Figure 6. In tropical regions such as India, the maximum anthropogenic AOD and forcing occurs during the dry season, whereas in extratropical regions US, Europe and China), it occurs during the summer months. The anthropogenic AOD is obtained from the Chung et al. [2005] study which in turn adopts the GOCART model simulation for the anthropogenic fraction of the total AOD. The GOCART model adopts a comprehensive emissions data for sulfates, nitrates, EC, organic carbon from fossil and bio fuels and biomass burning, which in turn have strong seasonal dependence.

[32] Proceeding down in spatial scale by another order of magnitude, we examine the conditions over megacities (population > 10 million) of the world (Figure 7). Figure 7 shows the population (Figure 7a), the annual mean AOD

and the radiative forcing of the atmosphere and the surface (dimming). We note first that, the AOD in Figure 7 is the total AOD (natural + anthropogenic) and the radiative forcing is the radiative effect of the total AOD (natural + anthropogenic). The Chung et al. [2005] analysis was a gridded analysis (for a grid of about 250 km \times 250 km) and the method developed for portioning the total AOD into natural and anthropogenic fraction may not be applicable to the city scales. Many of the cities are coastal cities and about 0.1 of the total AOD is due to natural aerosols for coastal regions. We obtained the 0.1 AOD as our best guess for natural AOD, by examining the AERONET AODs for rural locations and coastal sites when the low-level wind was of marine origin. Figure 7 again reveals the vast heterogeneity of the relationships between population, aerosol loading (AOD) and the forcing. Clearly aerosol concentration cannot be scaled just to population. For example, the AOD over Bangkok is 4 times that over Sao Paolo whose population is twice as much as Bangkok's. In addition to population, technological development, per capita consumption of energy and use of renewable energy (including biomass) which determine emission of aerosol precursors, we have to account for other variables involving meteorology, geography and long-range transport.

[33] Consistent with our earlier suggestion for regional hot spots (AODs > 0.3), we adopt AOD > 0.4 in Figure 7 to identify brown cloud hotpots for megacities. We are allowing



Figure 7. (a) Population, (b) MODIS annual mean aerosol optical depths and (c) the annual mean aerosol forcing in the atmosphere and at the surface for the top 26 megacities (http://en.wikipedia.org/ wiki/Megacity) in the world. The aerosol forcing is estimated by multiplying MODIS AOD with the MACR-modeled forcing efficiency. The AOD and forcing values are for 2001–2003.

for natural AOD of 0.1 and hence denote hot spots as those regions with AOD > 0.4. Cities with such large AODs are: Bangkok, Beijing, Cairo, Dhaka, Karachi, Kolkata (formerly Calcutta) Lagos, Mumbai (Bombay), New Delhi, Seoul, Shanghai, Shenzen and Tehran. Thirteen of the twenty six megacities in Figure 7 fall under the hot spots category. Cities in even highly developed nations, such as Tokyo, New York, Los Angeles, Paris and London have AODs exceeding 0.2, with Tokyo's AOD of 0.3 being the largest among the developed nation megacities. Megacities with lowest AODs are all in South America (Sao Paolo, Rio de Janeiro and Buenos Aires). It is important to remember that these (Figure 7) are only annual mean values. Monthly mean AODs during pollution seasons (summer months for extratropics and winter/spring in tropics) can be at least twice the annual mean values (e.g., compare AODs between Figures 5 and 6) and daily maxima can exceed the maximum seasonal values by another factor of two.

[34] The direct radiative forcing of the aerosols (natural plus anthropogenic) are shown in Figure 7c. We do not present anthropogenic forcing because the *Chung et al.* [2005] study relies upon the GOCART model whose spatial resolution is too coarse to characterize parameters on the scale of a city. As expected (for brown clouds) the large reduction of surface solar radiation is balanced by a com-

parably large increase in solar heating of the atmosphere. The annual mean surface dimming (i.e., reduction of net solar radiation at the surface) in most tropical megacities exceeds 20 W m⁻², equivalent to reducing solar irradiance at the top of the atmosphere by more than 10%. The increase in solar energy absorbed by aerosols over the atmosphere of some of these megacities is comparable to the heat input by energy consumption.

4. Annual Cycle of Aerosol Properties and Forcing From ABC Observatories

4.1. Tropical Indian Ocean

[35] The results shown in Figures 2–7 clearly underline the importance of understanding the aerosol-climate forcing level at regional to local scales before scaling it up to the hemispherical and global scales. This is one of the fundamental motivations behind establishing the ABC observatories. The initial focus of ABC is on the Indo-Asian-Pacific region since it contains the regional brown cloud hot spots as well as several of the megacity hot spots identified earlier in this study. The ABC observatories (including those under development and in planning stages) are shown in Figure 8. The focus of this study is on data from the ABC observatories in Maldives (an ABC supersite), Nepal, Gosan (ABC



Figure 8. A map of ABC observatories: the Maldives Climate Observatory–Hanimaadhoo (MCO-H), the Maldives Climate Observatory–Gan (MCO-G), the Indian Climate Observatory–Pune (ICO-P), the Indian Climate Observatory–Kharagpur (ICO-K), the Nepal Climate Observatory–GVR (NCO-G), the Nepal Climate Observatory–Pyramid (NCO-P), the Thailand Climate Observatory–Phimai (TCO-P), the Japan Climate Observatory–Okinawa (JCO-O), the Korea Climate Observatory–Gosan (KCO-G), the Pacific Climate Observatory–Momote (PCO-MT), the Pacific Climate Observatory–Midway (PCO-MW), the Pacific Climate Observatory–Mauna Loa (PCO-ML), and the National Atmosphere and Climate Observatory–Trinidad Head (NACO-T).

supersite) and Trinidad Head (California in USA), since these are the sites which currently provide the data needed for this study. In Maldives, ABC has two observatories, one in the island of Hanimaadhoo (near the northern end of the Maldives) and another at Gan just south of the equator. During the dry season, when north easterlies are prevalent Hanimaadhoo is underneath the brown clouds from south and SE Asia, while Gan is at the southern extent of the brown clouds with polluted air some days and cleaner Southern Hemisphere marine air on other days. During the southwest monsoon, both regions sample marine air near the surface, with occasional dust plumes from the Arabian peninsula over Hanimaadhoo. The latitude and longitude locations of these sites and their configuration are shown in Figure 9. The list of instruments in these sites as well as the start dates for data collection are shown in Table 1. Of these four sites, the most complete data are available only for Hanimadhoo site and we start with observations over this observatory, referred to as ABC MCOH which stands for ABC Maldives Climate Observatory at Hanimadhoo. ABC MCOG stands for the ABC Maldives observatory at Gan Island. Details of instrumentation at these two sites and first results of two seasons have been discussed by Corrigan et al. [2006] and Ramana and Ramanathan [2006].

[36] The observations started in October 2004 and Figure 10 shows time series of selected observations from then to

end of December 2005. Figure 10a shows the total aerosol concentration (size greater than 30 nanometers). The temporal variations in the concentration reflects the monsoon cycle that carries polluted air from south Asia over the Maldives during the dry season (November to March, see trajectories given by Corrigan et al. [2006] and Ramana and Ramanathan [2006]) and clean air from the Southern Hemisphere during the wet monsoon season (May to September). The white spaces in Figure 10 indicate transition periods when the monsoon winds are not clearly predictable and mixtures of polluted and clean air can occur. The observatories location provides the opportunity to look at both polluted and clean air masses depending upon the season. Gaps in the data are from instrument malfunctions, which have now been significantly reduced with the introduction of a water based instrument.

[37] The effect of the monsoon cycle is clearly apparent in the continuous measurements of the aerosol absorption as seen in Figure 10b. The aerosol absorption was obtained from the aethalometer, a filter based absorption photometer (see *Corrigan et al.* [2006] for details for data reduction). Filter collection at MCOH also provides daily concentrations of elemental carbon (EC), a common tracer for anthropogenic activity. The sampling and analytical methods used for the filter based measurements that were obtained at the ABC observatories are described elsewhere [*Schauer et al.*, 2003; *Stone et al.*, 2007]. Briefly, PM2.5 and PM10



Figure 9. Photos of the region along with the instrument tower for Hanimandhoo, Maldives; Gan, Maldives; Nepal; Gossan, Korea; and Trinidad Head, US.

particulate matter samples were collected on both 47-mm prefired quartz fiber filters and preweighed 47-mm Teflon membrane filters. The Teflon filters were weighed before and after sampling in a temperature and humidity controlled room using a microbalance to obtain the accumulation of particle mass on the filter. Quartz fiber filters were analyzed for organic carbon (OC) and elemental carbon (EC) using the thermal-optical method adopted by the ACE-Asia project [Schauer et al., 2003]. The sample flow through the sampler legs were controlled using critical orifices and were measured before and after each sample event. The measured ECOC and particulate matter mass concentrations were corrected for field blanks specific to each sampling site. The nominal detection limits for the sampling and analytical methods used in the ABC study are: 1.0 μ g m⁻³, 0.5 μ g m⁻³, and 0.05 μ g m⁻³ for particle mass, particle organic carbon and particle elemental carbon, respectively.

[38] An order of magnitude increase in both the absorption coefficient and EC concentration (from filters) is observed as the monsoon season ends. Black carbon falls off again with the return of the monsoon. The correlation between these two different types of measurements serves as a cross validation.

[39] Along with the scattering coefficient, the absorption coefficient is used to calculate the single scatter albedo (SSA), which is basically the fraction of the total radiation that is scattered by the aerosols. The fractional absorption is given by (1-SSA). Figure 10c shows the SSA at MCOH through December 2005. The SSA displays an inverse trend to the total particle concentration and absorption data, increasing from an SSA of about 0.92 during the peak dry season to about 0.98 or larger during the wet season. Aerosol particles coming from Asia during the dry season

contain more black carbon which causes them to absorb more light. Conversely, the wet monsoon brings particles that are relatively free of anthropogenic influence, but still contain small amount of absorbing aerosols (transported down from above 1 km to the surface) which is the source for SSA smaller than 1.

[40] While Figures 10a-10c show surface values, Figure 10d gives an indication of changes happening in the entire column. Temporal variation of monthly mean AOD (aerosol optical depths) over the Maldives is shown in Figure 10d and reveals the large build up in AOD with the onset of the dry season. Focusing first on the surface based AERONET radiometer measurements, we note that the AOD increases from about 0.1 in October 2004 to about 0.3 during the peak of dry season in February, followed by a steady decrease to 0.15 in October 2005. The monthly mean AODs were low ($\sim 0.1-0.15$) during the southwest monsoon and increased in association with wind direction reversal to a value of ~ 0.3 as the northeast monsoon advances. The steady increase in columnar AOD and decrease in SSA were attributed to the arrival of air masses carrying fresh continental pollutants that were characterized by relatively high concentrations of submicron absorbing aerosols [Corrigan et al., 2006]. Thus the abrupt increase in surface level aerosol and EC concentrations (Figures 10a and 10b) and the abrupt decrease in surface SSA during October were also manifested in the aerosol concentrations above the surface levels. On the basis of INDOEX aircraft data [R2001a], most of the contribution to the increase in AOD comes from levels below 3 km (which is the top of the trade wind inversion). This suggests that most of the pollution transport from south Asia occurs below the 3 km level.

Table 1. List of Radiation and Aerosol Instruments Deployed at Various ABC Observatories^a

| Parameter | Instrument Type, Manufacturer | ABC Sites | Start Date |
|---|--|--------------------------------------|--|
| | Radiation Instruments | | |
| Diffuse solar radiation | CM 21 Pyranometer $(0.3-2.8 \ \mu\text{m})$, Kipp & Zonen; CM 22 Pyranometer $(0.2 - 3.6 \ \mu\text{m})$ Kipp & Zonen | MCOH, NACO-T; MCOH, NACO-T | Oct 2004, Apr 2002; Oct 2004, Apr 2002 |
| Global solar radiation | CM 21 Pyranometer (0.3–2.8 μ m), Kinn & Zonen | MCOH, KCOG, NACO-T | Oct 2004, Feb 2003, Apr 2002 |
| Direct solar radiation | CH1 Pyrheliometer $(0.2-4.0 \ \mu m)$, Kipp & Zonen; Pyrheliometer with Quartz window, Eppley; Pyrheliometer | МСОН, КСОG; МСОН, NACO-T | Oct 2004, Feb 2003; Oct 2004, Apr 2002 |
| Net radiation in the far infrared | CG4 Pyrgeometer ($4.5-42.0 \mu m$), Kipp & Zonen; CNR1 Pyrgeometer ($5-50 \mu m$), Kipp & Zonen | MCOH, NACO-T; KCOG | Oct 2004, Apr 2002; Feb 2003 |
| Sun tracking | 2AP-GD Sun tracker with Sun sensor, pointing and shading ball assembly, Kinn & Zonen | MCOH, KCOG, NACO-T | Oct 2004, Feb 2003, Apr 2002 |
| Global PAR | narrowband radiometer GUV-2511 (0.305, 0.313, 0.32, 0.34, 0.38, 0.395, 0.4 – 0.7 µm) Bio Spherical inst | МСОН | Oct 2004 |
| Global spectral irradiance | Grating Spectroradiometer ($0.325-1.075 \ \mu$ m), FieldSpec Handheld, Analytical Spectral Devices | MCOH, MCOG | Oct 2004, Aug 2004 |
| Aerosol spectral optical depth, columnar precipitable water vapor | Microtops II Sun photometer (AOD at 380, 440, 500, 675, 870, 1020 nm; PWV at 940 nm), Solar Light Co.; CIMEL Sun photometer (AOD at 340, 380, 440, 500, 670, 870, 1020 nm; PWV at 940 nm), AERONET | MCOH, MCOG; MCOH, KCOG, NACO-T | Oct 2004, Aug 2004; Dec 2004, Apr 2001, Feb 2005 |
| Columnar ozone | Microtops II Ozonometer (305, 312, 320 nm), Solar Light Co. | МСОН | Oct 2004 |
| Wind speed and direction, temperature, humidity, pressure, rainfall | R.M. Young Company instruments | MCOH, MCOG, NACO-T | Oct 2004, Apr 2006, Apr 2002 |
| | Aerosol Instruments | | |
| Total particle concentration | Condensation Particle Counter, TSI-3022; Condensation Particle Counter, TSI-3010; Condensation Particle Counter, TSI-3022; Condensation Particle Counter, TSI-3010 | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Mar 2006, Mar 2005, Apr 2002 |
| Light scattering | Nephelometer, TSI-3563; Nephelometer, Ecotech-M9003; Nephelometer, TSI-3563; Nephelometer, TSI-3563 | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Mar 2006, Mar 2005, Apr 2002 |
| Particle absorption | Aethalometer, Magee AE-31; Aethalometer, Magee AE-31; Aethalometer, Magee AE-31; PSAP, Radiance Research | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Mar 2006, Mar 2005, Apr 2002 |
| Size distribution | Scanning Particle Mobility Sizer $(0.01-0.5 \ \mu m)$, TSI | MCOH, KCOG | Oct 2004, Mar 2005 |
| Size distribution | Aerodynamic Particle Sizer (0.5–20 μ m), TSI 3321 | МСОН | Oct 2004 |
| CCN | CCN counter, DMT | MCOH | Oct 2004 |
| Organic/black carbon (PM2.5, PM10) | custom URG filter sampler | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Sep 2004, Mar 2005, Aug 2005 |
| Inorganic particulates (PM2.5, PM10) | custom URG filter sampler | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Sep 2004, Mar 2005, Aug 2005 |
| Mass (PM2.5, PM10) | custom URG filter sampler | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Sep 2004, Mar 2005, Aug 2005 |
| Metals and organic tracers (PM2.5, PM10) | custom URG filter sampler | MCOH, MCOG, KCOG, NACO-T | Oct 2004, Sep 2004, Mar 2005, Aug 2005 |
| Dust | custom high-volume sampler (cellulose filters) | MCOH | Oct 2004 |

^aMCOH, Maldives Climate Observatory, Hanimaadhoo; MCOG, Maldives Climate Observatory, Gan; KCOG, Korea Climate Observatory, Gosan; NACO-T, North America Climate Observatory, Trinidad.

[41] One remarkable feature in all of the four panels is that the transition from the wet monsoon aerosol values during October to the dry monsoon values (and flow) during early November occurred abruptly over a period of a few weeks above MCOH and revealed a dramatic contrast between the natural marine aerosols transported from the south Indian Ocean by the southwest monsoon and that of the polluted aerosols transported from the south and SE Asian region by the northeast monsoon [also see *Ramana and Ramanathan*, 2006].

[42] The complementary observatory on the far southern island of Gan (MCOG) is important since it lies 800 km south of MCOH, which puts it just below the equator. MCOG is influenced by the Intertropical Convergence Zone (ITCZ) to a greater extent than MCOH. As a result, the air is typically less polluted and experiences a slightly different monsoon pattern. Figure 11 shows the total particle con-



Figure 10. Annual time series of aerosol measurements over MCO-H. (a) Daily average total particle concentration (instrument is the TSI CPC 3022); (b) daily average absorption and black carbon concentration (instrument for absorption is 7 wavelength aethalometer Magee AE-31, and the black carbon concentration is from filter analysis using a thermal optical analytical method); (c) daily average SSA (SSA derived from absorption measurements taken with aethalometer and with scattering taken from the TSI 3 wavelength Nephelometer model 3563); and (d) temporal variation of monthly mean aerosol optical depths (AOD) measured using Microtops and CIMEL Sun photometers and from MODIS instrument on board NASA's Terra satellite. The frequency of the measurements at MCO-H is about 1 min for total particle concentration and 5 min for black carbon concentration. Sun photometers (CIMEL and Microtops) at the surface measure the AOD at 15-20 min intervals. However, the cloud screening reduces the number of points available for the analysis. MODIS measures AOD nearly once a day in the tropic area.

centration at both MCOH and MCOG for the period when MCOG data were available. It is apparent that MCOG is a less polluted location and provides an excellent contrast to MCOH. During early March (2006) when the north easterly flow (bringing pollution from south Asia) was still prevalent and extended up to Gan latitudes, the concentration at MCOG varied from 250 to 500 /cc while at MCOH it was about 4 times higher (1000 to 1600 #/cc). By middle of April, the northeasterly flow has retreated from Gan and the concentration drops from 250 to 500 to about 25 to 75 #/cc, perhaps providing a baseline for pristine marine air in the southern Indian Ocean under low wind conditions (2 to



Figure 11. Comparison of particle concentrations measured at MCO-H and at MCO-G from April to June 2006.

5 m s⁻¹). The north to south spatial variations of aerosol properties have also been described in detail by *Sheridan et al.* [2002] and *Jayaraman et al.* [2001].

[43] The critical parameter for estimates of radiative forcing and cloud condensation nuclei is the size distribution which is shown in Figure 12b for the 4 seasons (for MCOH). The scanning mobility particle sizer (SMPS)



Figure 12. Annual cycle of aerosol number density. (a) Daily total particle number variation for the diameter Dp < 500 nm. (b) Seasonal cycle of size distribution of aerosol number density.



Figure 13. 24-hour average fine particle concentrations measured at the ABC Observatories from September 2004 through December 2006.

effectively measured size and number of particles between 0.01 and 0.5 μ m with a resolution of 64 channels per decade. Size distributions were obtained at the surface from MCOH every 5 min. None of the four profiles indicate new particle formation. New particle formation would manifest by concentrations increasing with a decrease in size for diameters less than 30 nm. Thus this part of the Arabian Sea mostly witnesses aged particles (outside the monsoon season in summer) or primary sea salt aerosols (summer). Lowest concentrations are found during the SW monsoon season and the size distributions are typical of marine boundary layer aerosols [Roberts et al., 2006] with a primary peak around 200 to 300 nm (diameter). Two peaks are seen during the other seasons also with one primary and one secondary peak. Given the 2 to 4 days of traveltime from south Asia, most of these aerosols must have been processed through clouds (see Roberts et al. [2006] for the effect of cloud processing on aerosol size distribution), which may explain the peak around the larger diameter range between 150 to 300 nm. The daily time series of integrated number density for diameter less than 500 nm (particles that have the most influence as CCN and radiative forcing) is shown in Figure 12a. Aerosol concentrations increase rapidly from 500 #/cc in early October to 3000 #/cc and drop to less than 50 #/cc during the peak SW monsoon period in August. This low value supports our earlier deduction from Gan data (south of equator) that the baseline aerosol concentration for clean marine air is between 25 to 75 #/cc or less.

4.2. Brown Cloud Carbonaceous Content in the Indo-Asia-Pacific Region

[44] We next focus on the dominant ingredient that is responsible for solar absorption in the brown clouds, which is the carbonaceous component. It is basically a mixture of elemental carbon (alternately referred to as black carbon) and organics (basically hundreds of species). In order to comprehend its temporal and spatial variations, we compare the carbonaceous content at the ABC observatories in Maldives, Nepal, Gosan (South Korea) and Trinidad Head



Figure 14. 24-hour average fine particle organic carbon (OC) concentrations measured at the ABC Observatories from September 2004 through December 2006.

(coast of northern California). Unless otherwise mentioned the results in this section pertain to aerosol and carbon mass for particles with diameter less than 2.5 microns, alternately referred to as fine aerosols.

[45] We begin with total (all aerosol types and species) mass of fine (diameter < 2.5 microns) aerosols in Figure 13. As expected, during most of the year, the aerosol mass over MCOH and Nepal are about a factor of 5 to 10 larger than that at MCOG or Trinidad Head. Because of fast long-range transport from south Asia, the aerosol mass in a remote marine location (MCOH) is as high (10 to 100 μ g m⁻³) as it is over a rural/urban site in Nepal. Another interesting feature is that for the most part, the seasonal cycle over the Arabian Sea is not very dissimilar to that over Nepal. There is significant interannual variability in the aerosol mass. For example, the mass concentration at MCOH is about 1 to 5 μ g m⁻³ during September 2004, whereas it ranges from 5 to 20 μ g m⁻³ during September 2005.



Figure 15. 24-hour average fine particle elemental carbon (EC) concentrations measured at the ABC Observatories from September 2004 through December 2006.



Figure 16. Black carbon to total carbon ratios (EC/TC) from filter data providing an indication of changing air masses at the different ABC observatories.

Figure 14 shows elemental carbon (EC) mass while Figure 15 shows organic carbon (OC) mass. Both EC and OC in Arabian Sea (MCOH) is smaller than that over Nepal by a factor of 2 to 5. The seasonal cycle of EC and OC over MCOH is similar to that shown in Figure 10 for aerosol number density. Before the onset of the dry season, the EC (and OC) concentrations reach negligible values over the Arabian Sea (both MCOH and MCOG) during September and October and increase by about an order of magnitude within a month into the dry season and stays at high values $(>0.1 \ \mu g \ m^{-3})$ for another 5 months. It finally attains negligible values from May to August. During the months when Trinidad Head data are available (September 2005 to January 2006), the EC and OC concentrations in MCOH are comparable to those in Trinidad Head, indicating both sites are subject to continental pollution (south Asia in the case of MCOH and North America in the case of Trinidad Head). Again there are significant interannual variations. For example during November and December, the 2005 concentrations of EC are comparable to those in 2006; but for OC the 2005 concentrations are a factor of 2 to 3 larger.

[46] EC to total carbon ratios (EC/TC) from filter data (Figure 16) provide an indication of changing air masses at the different ABC observatories. The EC/TC ratio is sometimes used to characterize combustion aerosol sources, such as fossil fuel vs. biomass, and can serve as an initial guide to pursue more specific source characterization analysis. For example, Novakov et al. [2000] show that EC/TC ratio in cities in Japan are typically about 0.5 whereas in regions dominated by biomass burning the ratio is in the range of 0.1 to 0.2. The plot illustrates the vast regional differences in the aerosol carbon partitioning. As the total carbon concentration (TC) increases, the Nepal EC/TC ratio decreases indicating more biomass fuel combustion. Conversely, the economically developed location of Gosan, Korea shows a slight increase of the EC/TC ratio as pollution levels increase. In summary the filter data (Figures 13-16) and the aerosol physical property data (Figures 10-12) illustrate the large seasonal, regional and interannual variations in aerosol chemistry and physical properties.

4.3. Radiative Forcing at the Surface From ABC_MCOH

[47] In this subsection the forcing is obtained directly from radiation flux observations as opposed to section 3 in which the forcing was obtained from MACR model, which itself employs observationally constrained input.

[48] Next we validate MACR by comparing its forcing with observed forcing at the surface. This part of the study is basically shown in the top two boxes of Figure 1 (left). We will first estimate the clear sky aerosol radiative forcing following the procedure outlined by *Satheesh and Ramanathan*, [2000] and as modified slightly by *Ramana and Ramanathan* [2006]. It basically involves taking daily variations in observed AODs (Figure 17) for each month and correlating with daily variations in ob-



Figure 17. Temporal variation of daily mean aerosol optical depth (AOD) at 500 nm over MCOH from October 2004 to December 2005 measured using Microtops and CIMEL Sun photometers. The solid line is the monthly mean AOD.



Figure 18. Diurnal average aerosol broadband $(0.3-2.8 \ \mu m)$ radiative forcing at the surface as a function of AOD for May 2005 at MCOH. The measured data are shown by points. Overlaid dashed lines are theoretical (MACR) radiative forcing calculations for different values of SSA (1.0, 0.96 and 0.92) with 0.05 AOD intervals. The aerosol forcing efficiency from the measurements is $-59 \pm 4 \text{ W m}^{-2}/\text{AOD}$.

served clear-sky broadband solar fluxes. The major challenge in this approach involves the identification of clear sky fluxes. The technique adopted in our study is explained by Satheesh and Ramanathan [2000], who also quantifies the uncertainty in the estimated forcing as about 10% (2 sigma). Figure 17 shows daily mean and monthly mean AODs from October 2004 to December 2005. Basically, AOD peaks during January and steadily decreases to a minimum during October. The variations in AOD follow variations in surface concentrations of aerosols and thus the seasonal variations in AOD are largely driven by the variations in anthropogenic aerosols from south and SE Asia. The daily variation in AOD is large enough to provide a good dynamic range for correlating AOD with observed solar radiation fluxes. An example of the correlation is shown for May 2005 in Figure 18. From the measured diurnal solar radiation fluxes, we subtract the expected diurnal flux for a no-aerosol atmosphere, and hence the correlation line asymptote to zero value as AOD approaches zero. The no-aerosol estimates use the observed column water vapor amount and column ozone amount [see Ramana and Ramanathan, 2006]. This procedure does not affect the correlation since we are simply subtracting no-aerosol estimates from all data points. The slope of the correlation curve is the forcing efficiency, i.e., the rate of decrease of the global flux (direct plus diffuse) with respect to an increase in AOD. When the efficiency is multiplied with the AOD, it yields the forcing at the surface. Figure 18 also shows the theoretical values for the slope (calculated by



Figure 19. (a) Temporal variation of cloud-free monthly mean aerosol forcing efficiency at MCOH from October 2004 to December 2005. Monthly mean aerosol forcing efficiency is obtained as the line of best fit to the respective diurnal average aerosol forcing and AOD. (b) Temporal variation of cloud-free monthly mean aerosol forcing at the surface from October 2004 to December 2005. Monthly mean aerosol forcing is determined by multiplying the monthly mean aerosol forcing efficiency with monthly mean AOD.



Figure 20. Comparison of monthly mean clear-sky surface aerosol forcing between observation and calculations over MCOH. The mean bias, root mean square (RMS) error, slope and correlation coefficient (correl) are presented. Aerosol forcings are given in W m^{-2} .

MACR model) for various SSAs. The MACR calculations adopt the AOD, the angstrom coefficient and the SSA from the AERONET observations at MCOH. These are column average values and for the vertical distribution of these parameters, we scale these with the vertical distribution of aerosol number density as observed during INDOEX (see R2001a). As the SSA decreases (i.e., as the absorption increases), more solar radiation is attenuated at the surface and hence the slope gets steeper. The best match between the slope from data and the theoretical slope is obtained for an SSA = 0.96, which indicates a moderately high absorbing aerosol. For January however, the best match is obtained for an SSA close to 0.9, which is a highly absorbing aerosol, consistent with INDOEX results and the SSA, OC, and EC data shown in Figures 10, 14, and 15, respectively. It is comforting that the aerosol physical and chemical data are consistent with the measured radiative forcing efficiency. The seasonal variation of the surface forcing efficiency and the forcing are shown in Figure 19. The peak surface forcing occurs during the dry season months of December to February, when the forcing exceeds -20 W m^{-2} and even during the monsoon season, aerosol forcing is as high as -10 W m⁻

[49] The next critical step (Figure 1) is to validate MACR with the observed forcing. This step is critical since we cannot derive the cloudy sky direct forcing from observations and we need to rely on MACR. The MACR input is from the assimilated aerosol data from *Chung et al.* [2005] and thus in validating MACR we are also validating the assimilated data given by *Chung et al.* [2005]. The monthly diurnal mean aerosol radiative forcing values are calculated by using MACR under clear and cloudy sky condition. For

cloudy sky forcing CERES (Cloud and Earth's Radiant Energy System) cloud data (e.g., low, middle and high clouds) over MCOH region are used for the year of relevance to this study, i.e., 2004 to 2005. The surface forcing comparison between observations and MACR calculations under the clear sky conditions shown in Figure 20, reveal a small mean bias of 0.3 W m^{-2} with a small RMS error of only 2 W m⁻². The seasonal variation of the clear sky surface forcing shown in Figure 21 is very similar to the observed forcing (Figure 19). Since the clear sky surface forcing compares favorably with the observed values, we are showing MACR predictions for TOA forcing. The forcing at the surface and the TOA during the dry season (December to February) are almost twice those during the monsoon season (June to August). The January TOA clear sky forcing of -12 W m⁻² is equivalent to enhancing the clear sky albedo (about 0.13) of the region by almost 20% to 0.16. We now use MACR to estimate the forcing under average cloudy skies (Figure 21). In general, as expected, the cloudy sky forcings are smaller than clear sky forcings, i.e., cloudy sky forcing is 60-70% of clear sky forcing at the surface. Clouds reduced the aerosol effect at TOA and the maximum reduction of TOA forcing occurs during the monsoon season (June to August) when the cloudiness is at its maximum value.

5. Simulations With Regional Aerosol Chemical-Transport Model

[50] The simulated regional concentrations of sulfate, organic carbon and elemental carbon near the surface and at about the 3 km level from STEM-2K are shown in Figure 22 for 1 October to 15 November (early dry season). The heavily polluted regions in the Indo-Gangetic Plains and eastern China (the hot spots identified from satellite data in Figures 2 and 4), are simulated well by the model. A highly polluted spot in Indonesia (reflective of the biomass burning) is also seen similar to that shown in Figures 2 and 3. In these hot spots, EC mass exceeds 0.5 μg m⁻³, the OC mass exceeds 2 μ g m⁻³ and sulfate mass exceeds 10 μ g m⁻³ both at the surface and at \sim 3 km. Adjoining the two hot spots in south Asia and eastern China is one large brown cloud over the entire Indo-Asian-western Pacific region. In the Indian Ocean the plume extends down to the equator in the western end, while it reaches as far south as 10°S in the eastern portion. Hence it is not surprising that observed EC and OC concentration over Gan (close to equator) reached as high a value as in Hanimaadhoo during October 2004.

[51] The annual cycle of the simulated surface values of sulfates, EC and OC are compared with observed values over ABC-MCOH in Figure 23. The model is able to capture the overall seasonal variations, with peak values during the dry season months and minima during monsoon and postmonsoon transition months. In addition the magnitude of the range of values observed (i.e., minima and maxima) for each species are accurately predicted. In general the predictions for sulfate, BC and OC show similar performance, but with sulfate being slightly poorer. The correlation coefficients (and regression slopes) are: 0.54 (0.7) for sulfate; 0.64 (1.3) for BC; and 0.6 (0.95) for OC. The major deficiencies in the predictions of these species are: a too early arrival of polluted air masses in the



Figure 21. Monthly mean aerosol radiative forcing (W m^{-2}) (a) at TOA and (b) at surface during October 2004 to December 2005. The solid and open bars represent the clear and cloudy conditions, respectively. For model calculation, aerosol optical properties measured over MCO-H are used.

postmonsoon transition period months of October and November (when the predictions for the transition period October/November are removed the performance of the model increases significantly, with correlation coefficients of 0.77, 0.72, and 0.67, for sulfate, BC and OC, respectively); and an underprediction of the sulfate peaks during the post– dry season transition months (March and April). These problems suggest that the wet removal of aerosols in the postmonsoon transition period is too weak, and that the sulfate production in the post–dry season is too slow. These processes are discussed in further detail by *Adhikary et al.* [2007].

[52] The model readiness for being useful for climate forcing calculations depends on its ability to simulate the column AOD, which is shown in Figure 24. Figure 24 also shows the contribution to the total AOD from each of the aerosol species. The model AOD for MCOH peaks in November at AOD of about 0.5, whereas the observed AOD peaks in January at an AOD of about 0.3. This over estimation of November AOD is consistent with the

overestimation of surface sulfates. EC and OC during the postmonsoon transition months. For January, however, the model AOD is consistent with the observed AOD of 0.3. Another major deficiency is that the model AOD reaches a minimum of about 0.2 in February and increase to a secondary maximum value of 0.4 in July, whereas the observed AODs decrease steadily from its maximum value of 0.3 in January to a minimum of about 0.18 during the monsoon months. The over prediction of AOD during the monsoon season is mostly due to the large contribution from fine dust. Dust AODs in fact peak during the monsoon season over the Arabian Sea (as shown by Li and Ramanathan [2002]) but it does not extend as far south as MCOH. The model is over estimating the southward extent of the dust plumes from the Arabian region, reflecting a too weak wet removal of the fine mode dust. The calculated anthropogenic contribution to the AOD maximizes during the dry season, exceeding 70% from November through March, consistent with the INDOEX findings of R2001a.



Figure 22. Simulated regional concentrations of sulfate, organic carbon and elemental carbon at surface and 3 km. Plotted are mean values during the period of the ABC-APMEX field campaign (1 October to 15 November (i.e., the early dry season)).

[53] Also shown in Figure 24 are predicted annual mean AOD for selected megacities in Asia with brown-cloud hot spots as defined earlier, along with those for MCOH and Katmandu. Also shown are the monthly variations in the total calculated AOD, the annual mean anthropogenic AOD, and the contribution of absorbing aerosols to the anthropogenic AOD (as a fraction). These results show a general agreement (within 20%) with the observed AODs, also shown in Figure 24. For example, it correctly predicts Karachi, Delhi, Dhaka and Beijing as the haziest cities among the cities shown in Figure 24. In general the anthropogenic contribution to the annual mean AOD is $\sim 2/3$ rds, with much higher values in the peak pollution periods, with the remainder of the contribution due to dust. The large role of absorbing aerosol in the Asia megacities is also clearly shown, with their annual contribution to AOD of 10% or higher. Predictions of the contributions of various anthropogenic species to AOD play an important role in evaluating the expected impact of future changes in anthropogenic emissions.

[54] In spite of using observed meteorological fields as well as using high spatial resolution, much improvement is needed to capture the spatial and seasonal variations in aerosol chemistry and AOD (and hence aerosol forcing). Model improvements will require reducing the large uncertainties in anthropogenic as well as natural aerosol (and their precursor) emissions, and better parameterizations of aerosol wet removal and aerosol chemistry, especially for high aerosol loadings and low-latitude wet conditions (i.e., the tropics). The observational data being obtained under ABC provide the means to help identify model deficiencies, as well as to stimulate the development of better parameterizations.

6. Conclusions

[55] The study uses a spectrum of satellite observations, global assimilated aerosol data sets, ABC observatories, a Monte Carlo aerosol-cloud-radiation model (MACR) and a regional chemical transport model (STEM-2K) to characterize physical and chemical properties of brown clouds and their direct radiative forcing. The analysis extends from global, to regional, megacity scale to local scales. It also presents the first annual cycle of observations from south Asian observatories and uses it to understand the role of the monsoon in regulating aerosol forcing over the Indo–south Asian region. The major findings of this study are summarized below.

[56] It identifies five major regional hot spots where anthropogenic aerosol optical depths exceed 0.3 and the absorption AOD exceeds 0.03. These hot spots were identified using observationally constrained AODs and single scattering albedos. These hot spot threshold values are seasonal mean values and thus are symptomatic of persistent pollution on seasonal to annual timescales. The hot spots are as follows: (1) east Asia (eastern China,



Elemental Carbon Concentration at MCOH



Figure 23. Comparison of predicted and observed (a) surface fine mode (PM2.5) sulfate, (b) EC, and (c) OC at MCO-H.

Thailand, Vietnam and Cambodia), (2) Indo-Gangetic Plains in south Asia (the northwest to northeast region extending from eastern Pakistan, across India to Bangladesh and Myanmar), (3) Indonesian region, (4) southern Africa extending southward from sub-Saharan Africa into Angola and Zambia and Zimbabwe, and (5) the Amazon basin in South America. The brown clouds decrease the annual mean value of surface solar radiation by 10 W m⁻² or more, and the atmospheric aerosol solar heating is comparably large.

[57] The chemical transport model reveals that, in the hot spots over the Indo-Gangetic Plains and east Asia, the simulated EC mass exceeds 1 μ g m⁻³, the OC mass exceeds 4 μ g m⁻³ and sulfate mass exceeds 10 μ g m⁻³ both at the surface and at ~3 km. Adjoining the two hot spots in south Asia and eastern China, is one large brown cloud over the entire Indo-Asian-western Pacific region.

[58] The study also identifies thirteen megacity hot spots for brown clouds based on the column aerosol optical depth exceeding 0.4: Bangkok, Beijing, Cairo, Dhaka, Karachi, Lagos, Mumbai, New Delhi, Seoul, Shanghai, Shenzen and Tehran. The reduction of annual mean surface net solar radiation is in the range of -20 to -60 W m⁻² accompanied by a large solar heating of the atmosphere. It is important to investigate to what extent such a redistribution of solar heating stabilizes the lower atmosphere and leads to an increase in the frequency of inversion and major pollution events in megacities.

[59] Even in industrialized regions, anthropogenic AODs are high and absorbing aerosols are prevalent. For example, over the eastern half of US and western Europe the total (scattering and absorbing) AOD exceeds 0.2 and absorbing AODs exceed 10% of total AOD.

[60] The analysis next focuses on regional differences between emissions, aerosol properties and forcing. The emissions inventories of Bond et al. [2004], the aerosol properties and forcing from the regional to global assimilation product of Chung et al. [2005] were used for this analysis. The AODs and forcing over India presents an interesting contrast to those over US, Europe and China. Although its emission of SO_2 is a factor 3 lower than that of US and a factor of 4 lower than that of China, the annual mean anthropogenic AOD over India is a factor of two larger than that of the US and about the same as over China. Furthermore, the largest (among the regions shown in Figures 5 and 6) annual mean TOA aerosol cooling occurs over India (-1.4 W m^{-2}) and the largest annual mean surface dimming (-14 W m^{-2}) also occurs over India. The unusually long (4 to 5 months) dry season with a persistent trade wind inversion and differences in geography and longrange transport are identified as the major factors (in



Figure 24. (a) Seasonal cycle of simulated total AOD, individual aerosol contributions, and anthropogenic AOD at MCOH. (b) Annual averaged AOD at MCOH, Katmandu, and selected megacities in Asia with brown-cloud hot spots (AOD > 0.4). Also shown is the range of monthly mean values (max and min) in the total calculated AOD, the annual mean anthropogenic AOD, and the contribution of absorbing aerosols to the anthropogenic AOD (as a fraction).

addition to emissions of EC and OC) in the anomalously large aerosol loading and forcing over India.

[61] In order to relate the column total and absorption optical depths to aerosol physical and chemical properties, the analysis focuses next on data from ABC observatories in four locations: Arabian Sea observatory in Maldives (two locations, MCOH in Hanimadhoo at 6.78°N, 73.18°E, MCOG in Gan at 0.69°S, 73.15°E), in Nepal (Godavari at 27.08°N, 83.3°E), in South Korea (KCOG in Gosan, 36.68°N, 126.17°E) and in North America (NACO-T in Trinidad Head, 41.05°N, 124.15°W). The Maldives observatories clearly show that during the dry season, because of long-range transport from south Asia, aerosol concentrations exceed 2500 #/cc during the dry season and EC concentrations reach a peak of 1 μg m⁻³, OC exceeds 5 μ g m⁻³, single scattering albedos dip below 0.9 and AODs reach a peak of 0.4 and the dimming at the surface dips below -25 W m⁻². These observations also establish the sort of surface values of aerosols that will be encountered in the brown cloud hot spots. During the wet season on the other hand, at the same location the aerosol number

concentration is less than 500 #/cc, the EC concentration is less than 0.1 μ g m⁻³, SSA greater than 0.98 and AOD less than 0.1. The Southern Hemisphere site at Gan helps establish the background values for a pristine marine atmosphere. During clean season, the surface aerosol concentrations at MCOG and MCOH reach values as low as 50 #/cc. The limited observations at Gosan in South Korea, which is downwind of Korean and east Asian pollution, also reveal high values of EC as in Maldives, but the OC values are lower indicating the more fossil fuel related pollution in east Asia. The highest year-round EC concentrations (0.5 to 2 μ g m⁻³) and OC (5 to 20 μ g m⁻³) were found in the Nepal observatory, raising major issues about the role of EC solar heating on Himalayan glaciers.

[62] The Maldives data were used to obtain directly from observations clear sky direct forcing at the surface for all 12 months of the year. The cloudy sky forcing was obtained from MACR, which in turn employed observed aerosol properties and cloud data from satellites. MACR simulation of the clear sky forcing was validated with direct forcing observations from MCOH. The cloudy sky direct forcing reaches peak values of -23 W m⁻² in January 2005 (peak dry season) and reaches a low value of -7 W m⁻² in October 2004 (postmonsoon).

[63] The assimilated data as well as the observations from the ABC observatories are used to validate a regional chemical transport model for the Indo-Asia-Pacific region (STEM-2K), which is then used to gain insights into the role of pollution in aerosol optical properties and forcing. The model is able to simulate the heavily polluted regimes in the Gangetic plains and in China. In these regions the plume extends from the surface to about 3 km with EC values exceeding 0.5 μ g m⁻³; OC exceeding 2 μ g m⁻³ and sulfate mass exceeding 10 μ g m⁻³. The model tends to overpredict the dust concentrations and AODs presumably because of lack of adequate wet removal of dust during the monsoon season. It is able to simulate the high AOD values over the Asian megacities within 20% of the values derived from the regional to global assimilation technique. It reveals that the absorption AODs over megacities exceed 10% of the total AODs. Comparing the regional hot spots in Gangetic plains and eastern China, the sulfate and EC concentrations are much higher in east China (Figure 22) whereas OC is larger over the Gangetic plains indicating the major contribution from fossil fuel combustion in China and that of biomass burning in the Gangetic plains. The Indonesian hot spots also reveal large concentrations of EC and OC. Furthermore the hot spot plumes from east Asia and Indonesia spread across over the western Pacific warm pool.

[64] In spite of using observed meteorological fields as well as using high spatial resolution, much improvement is needed in STEM-2K to capture the spatial and seasonal variations in aerosol chemistry and AOD (and hence aerosol forcing). Model improvements will require reducing the large uncertainties in anthropogenic as well as natural aerosol (and their precursor) emissions, and better parameterizations of aerosol wet removal and aerosol chemistry, especially for high aerosol loadings and low-latitude wet conditions (i.e., the tropics). The observational data being obtained under ABC provide the means to help identify model deficiencies, as well as to stimulate the development of better parameterizations.

[65] In summary the spectrum of observations and model calculations presented here clearly establish the brown clouds as major regional climate forcing terms. The surface observations from the ABC observatories have played a major role in validating the models and establish the seasonal and interannual variations in the forcing. The next step is to develop the ABC approach (Figure 1) to estimate the complete annual cycle of the aerosol forcing for the entire Indo-Asia-Pacific region and in addition determine the interannual and decadal-scale variations in the forcing as input to regional and global climate models.

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References

- Ackerman, A. S., O. B. Toon, D. E. Stevens, A. J. Heymsfield, V. Ramanathan, and E. J. Welton (2000), Reduction of tropical cloudiness by soot, *Science*, 288, 1042–1047.
- Adhikary, B., G. R. Carmichael, Y. Tang, L. Lueng, J. J. Schauer, B. Stone, and M. V. Ramana (2007), Characterization of the seasonal cycle of south Asian aerosols: A regional-scale modeling analysis, *J. Geophys. Res.*, doi:10.1029/2006JD008143, in press.
- Andreae, M. O., and A. Gelencser (2006), Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6, 3131–3148.
- Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J.-H. Woo, and Z. Klimont (2004), A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, 109, D14203, doi:10.1029/2003JD003697.
- Carmichael, G. R., et al. (2003), Regional-scale chemical transport modeling in support of the analysis of observations obtained during the TRACE-P experiment, *J. Geophys. Res.*, *108*(D21), 8823, doi:10.1029/ 2002JD003117.
- Chin, M., P. Ginoux, S. Kinne, B. N. Holben, B. N. Duncan, R. V. Martin, J. A. Logan, A. Higurashi, and T. Nakajima (2002), Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sun photometer measurements, J. Atmos. Sci., 59, 461–483.
- Chung, C. E., and V. Ramanathan (2006), Weakening of north Indian SST gradients and the monsoon rainfall in India and the Sahel, J. Clim., 19, 2036–2045.
- Chung, C., S. Nigam, and J. Carton (2002), SST-forced surface wind variability in the tropical Atlantic: An empirical model, *J. Geophys. Res.*, *107*(D15), 4244, doi:10.1029/2001JD000324.
- Chung, C. E., V. Ramanathan, D. Kim, and I. A. Podgorny (2005), Global anthropogenic aerosol direct forcing derived from satellite and groundbased observations, *J. Geophys. Res.*, 110, D24207, doi:10.1029/ 2005JD006356.
- Conant, W. C., J. H. Seinfeld, J. Wang, G. R. Carmichael, Y. Tang, I. Uno, P. J. Flatau, K. M. Markowicz, and P. K. Quinn (2003), A model for the radiative forcing during ACE-Asia derived from CIRPAS Twin Otter and R/V Ronald H. Brown data and comparison with observations, *J. Geophys. Res.*, 108(D23), 8661, doi:10.1029/2002JD003260.
- Corrigan, C. E., V. Ramanathan, and J. J. Schauer (2006), Impact of monsoon transitions on the physical and optical properties of aerosols, *J. Geophys. Res.*, 111, D18208, doi:10.1029/2005JD006370.
- de Gouw, J. A., et al. (2001), Overview of the trace gas measurements on board the Citation aircraft during the intensive field phase of INDOEX, J. Geophys. Res., 106(D22), 28,453–28,468.
- Diner, D. J., et al. (1998), Multi-angle Imaging SpectroRadiometer (MISR)–Instrument description and experiment overview, *IEEE Trans. Geosci. Remote Sens.*, 36, 1072–1087.
- Dubovik, O., A. Smirnov, B. N. Holben, M. D. King, Y. J. Kaufman, T. F. Eck, and I. Slutsker (2000), Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements, J. Geophys. Res., 105, 9791–9806.
- Dubovik, O., B. N. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanré, and I. Slutsker (2002), Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, 59, 590–608.
- Gong, S. L. (2003), A parameterization of sea-salt aerosol source function for sub- and super-micron particles, *Global Biogeochem. Cycles*, 17(4), 1097, doi:10.1029/2003GB002079.
- Grell, G., et al. (1995), A description of the fifth generation Penn State/ NCAR mesoscale model (mm5), NCAR Tech. Note NCAR/TN-398+1A, 107 pp., Natl. Cent. for Atmos. Res., Boulder, Colo.

- Guazzotti, S. A., K. R. Coffee, and K. A. Prather (2001), Continuous measurements of size-resolved particle chemistry during INDOEX-Intensive Field Phase 99, J. Geophys. Res., 106(D22), 28,607–28,628.
- Gupta, P. K., R. C. Sharma, S. Koul, D. C. Parashar, T. K. Mandal, and A. P. Mitra (1999), Study of trace gas species including greenhouse gases over the Indian Ocean during INDOEX precampaign cruises of 1996, 1997 and 1998 on Sagar Kanya, Cur. Sci., 76(7), 944–946.
- Heymsfield, A. J., and G. M. McFarquhar (2001), Microphysics of INDOEX clean and polluted trade cumulus clouds, *J. Geophys. Res.*, *106*(D22), 28,653–28,674.
- Holben, B. N., et al. (2001), An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET, J. Geophys. Res., 106, 12,067–12,097.
- Huebert, B. J., T. Bates, P. B. Russell, G. Shi, Y. J. Kim, K. Kawamura, G. Carmichael, and T. Nakajima (2003), An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts, *J. Geophys. Res.*, 108(D23), 8633, doi:10.1029/ 2003JD003550.
- Jayaraman, A., D. Lubin, S. Ramachandran, V. Ramanathan, E. Woodbridge, W. D. Collins, and K. S. Zalpuri (1998), Direct observations of aerosol radiative forcing over the tropical Indian Ocean during the January-February 1996 pre-INDOEX cruise, *J. Geophys. Res.*, 103(D12), 13,827–13,836.
- Jayaraman, A., S. K. Satheesh, A. P. Mitra, and V. Ramanathan (2001), Latitude gradient in aerosol properties across the intertropical convergence zone: Results from the joint Indo-US study onboard Sagar Kanya, Cur. Sci., 80, 128–137.
- Kahn, R., P. Banerjee, and D. McDonald (2001), Sensitivity of multiangle imaging to natural mixtures of aerosols over ocean, J. Geophys. Res., 106, 18,219–18,238.
- Kahn, R., et al. (2005), MISR calibration and implications for low-lightlevel aerosol retrieval over dark water, J. Atmos. Sci., 62, 1032–1052.
- Kaufman, Y. J., A. E. Wald, L. A. Remer, B. C. Gao, R. R. Li, and L. Flynn (1997), The MODIS 2.1-mu m channel–Correlation with visible reflectance for use in remote sensing of aerosol, *IEEE Trans. Geosci. Remote Sens.*, 35, 1286–1298.
- Kaufman, Y. J., D. Tanré, and O. Boucher (2002), A satellite view of aerosols in the climate system, *Nature*, 419, 215–223.
- Krishnan, R., and V. Ramanathan (2002), Evidence of surface cooling from absorbing aerosols, *Geophys. Res. Lett.*, 29(9), 1340, doi:10.1029/ 2002GL014687.
- Lelieveld, J., et al. (2001), The Indian Ocean Experiment: Widespread air pollution from South and Southeast Asia, *Science*, 291, 1031–1036.
- Li, F., and V. Ramanathan (2002), Winter to summer monsoon variation of aerosol optical depth over the tropical Indian Ocean, *J. Geophys. Res.*, 107(D16), 4284, doi:10.1029/2001JD000949.
- Mandal, T. K., D. Kley, H. G. J. Smit, S. K. Srivastava, S. K. Peshin, and A. P. Mitra (1999), Vertical distribution of ozone over the Indian Ocean (15°N–20°S) during First Field Phase INDOEX-1998, *Cur. Sci.*, 76(7), 938–943.
- Menon, S., J. Hansen, L. Nazarenko, and Y. Luo (2002), Climate effects of black carbon aerosols in China and India, *Science*, 297, 2250–2253.
- Meywerk, J., and V. Ramanathan (1999), Observations of the spectral clearsky aerosol forcing over the tropical Indian Ocean, J. Geophys. Res., 104(D20), 24,359–24,370.
- Moorthy, K. K., A. Saha, B. S. N. Prasad, K. Niranjan, D. Jhurry, and P. S. Pillai (2001), Aerosol optical depths over peninsular India and adjoining oceans during the INDOEX campaigns: Spatial, temporal, and spectral characteristics, *J. Geophys. Res.*, 106(D22), 28,539–28,554.
 Müller, D., K. Franke, F. Wagner, D. Althausen, A. Ansmann, and
- Müller, D., K. Franke, F. Wagner, D. Althausen, A. Ansmann, and J. Heintzenberg (2001), Vertical profiling of optical and physical particle properties over the tropical Indian Ocean with six-wavelength lidar: 1. Seasonal cycle, J. Geophys. Res., 106(D22), 28,567–28,576.
- Neusüß, C., T. Gnauk, A. Plewka, H. Herrmann, and P. K. Quinn (2002), Carbonaceous aerosol over the Indian Ocean: OC/EC fractions and selected specifications from size-segregated onboard samples, *J. Geophys. Res.*, 107(D19), 8031, doi:10.1029/2001JD000327.
- Novakov, T., M. O. Andreae, R. Gabriel, T. W. Kirchstetter, O. L. Mayol-Bracero, and V. Ramanathan (2000), Origin of carbonaceous aerosols over the tropical Indian Ocean: Biomass burning or fossil fuels?, *Geophys. Res. Lett.*, 27, 4061–4064.
- Olivier, J. G. J. and J. J. M. Berdowski (2001), Global emissions sources and sinks, in *The Climate System*, edited by J. Berdowski et al., pp. 33– 78, Taylor and Francis, Philadelphia, Pa.
- Podgorny, I. A., and V. Ramanathan (2001), A modeling study of the direct effect of aerosols over the tropical Indian Ocean, J. Geophys. Res., 106(D20), 24,097–24,106.
- Podgorny, I. A., W. Conant, V. Ramanathan, and S. K. Satheesh (2000), Aerosol modulation of atmospheric and surface solar heating over the tropical Indian Ocean, *Tellus, Ser. B*, 52, 947–958.

- Quinn, P. K., D. J. Coffman, T. S. Bates, T. L. Miller, J. E. Johnson, E. J. Welton, C. Neusüss, M. Miller, and P. J. Sheridan (2002), Aerosol optical properties during INDOEX 1999: Means, variability, and controlling factors, J. Geophys. Res., 107(D19), 8020, doi:10.1029/2000JD000037.
- Rajeev, K., V. Ramanathan, and J. Meywerk (2000), Regional aerosol distribution and its long-range transport over the Indian Ocean, J. Geophys. Res., 105, 2029–2043.
- Ramana, M. V., and V. Ramanathan (2006), Abrupt transition from natural to anthropogenic aerosol radiative forcing: Observations at the ABC-Maldives Climate Observatory, J. Geophys. Res., 111, D20207, doi:10.1029/2006JD007063.
- Ramanathan, V., and P. J. Crutzen (2003), New directions: Atmospheric brown "Clouds", Atmos. Environ., 37, 4033–4035.
- Ramanathan, V., and M. V. Ramana (2003), Atmospheric brown clouds: Long range transport, *Environ. Manage.*, 12, 28–33.
- Ramanathan, V., et al. (2001a), Indian Ocean Experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, J. Geophys. Res., 106, 28,371–28,398.
- Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld (2001b), Atmosphere– Aerosols, climate, and the hydrological cycle, *Science*, 294, 2119–2124.
- Ramanathan, V., C. Chung, D. Kim, T. Bettge, L. Buja, J. T. Kiehl, W. M. Washington, Q. Fu, D. R. Sikka, and M. Wild (2005), Atmospheric brown clouds: Impacts on South Asian climate and hydrological cycle, *Proc. Natl. Acad. Sci. U. S. A.*, 102, 5326–5333.
- Reddy, M. S., and O. Boucher (2004), A study of the global cycle of carbonaceous aerosols in the LMDZT general circulation model, J. Geophys. Res., 109, D14202, doi:10.1029/2003JD004048.
- Reiner, T., D. Sprung, C. Jost, R. Gabriel, O. L. Mayol-Bracero, M. O. Andreae, T. L. Campos, and R. E. Shetter (2001), Chemical characterization of pollution layers over the tropical Indian Ocean: Signatures of emissions from biomass and fossil fuel burning, J. Geophys. Res., 106(D22), 28,497–28,510.
- Remer, L. A., et al. (2005), The MODIS aerosol algorithm, products, and validation, J. Atmos. Sci., 62, 947–973.
- Rhoads, K. P., P. Kelley, R. R. Dickerson, T. P. Carsey, M. Farmer, D. L. Savoie, and J. M. Prospero (1997), Composition of the troposphere over the Indian Ocean during the monsoonal transition, *J. Geophys. Res.*, 102(D15), 18,981–18,996.
- Roberts, G., G. Mauger, O. Hadley, and V. Ramanathan (2006), North American and Asian aerosols over the eastern Pacific Ocean and their role in regulating cloud condensation nuclei, *J. Geophys. Res.*, 111, D13205, doi:10.1029/2005JD006661.
- Satheesh, S. K., and V. Ramanathan (2000), Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface, *Nature*, 405, 60-63.
- Schauer, J. J., et al. (2003), ACE-Asia intercomparison of a thermal-optical method for the determination of particle-phase organic and elemental carbon, *Environ. Sci. Technol.*, *37*, 993–1001.
- Sheridan, P. J., A. Jefferson, and J. A. Ogren (2002), Spatial variability of submicrometer aerosol radiative properties over the Indian Ocean during INDOEX, J. Geophys. Res., 107(D19), 8011, doi:10.1029/ 2000JD000166.
- Smith, S. J., R. Andres, E. Conception, and J. Lurz (2004), Historical sulfur dioxide emissions 1850–2000: Methods and results, *Res. Rep. PNNL-*14537, U. S. Dep. of Energy, Washington, D. C. (Available at http:// www.pnl.gov/main/publications/external/technical_reports/PNNL-14537.pdf).
- Stone, E. A., G. C. Lough, J. Schauer, P. Siva, C. Corrigan, and V. Ramanathan (2007), Understanding the origin of black carbon in the atmospheric brown cloud over the Indian Ocean, J. Geophys. Res., doi:10.1029/2006JD008118, in press.
- Streets, D. G., et al. (2003), An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108(D21), 8809, doi:10.1029/2002JD003093.
- Tang, Y., et al. (2004), Impacts of dust on regional tropospheric chemistry during the ACE-Asia experiment: A model study with observations, J. Geophys. Res., 109, D19S21, doi:10.1029/2003JD003806.
- Tanré, D., Y. J. Kaufman, M. Herman, and S. Mattoo (1997), Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances, J. Geophys. Res., 102, 16,971–16,988.
- Vogelmann, A. M., V. Ramanathan, and I. A. Podgorny (2001), Scale dependence of solar heating rates in convective cloud systems with implications to general circulation models, *J. Clim.*, 14, 1738–1752.

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