GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L12107, doi:10.1029/2004GL019702, 2004

Aerosol radiative forcing over a tropical urban site in India

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Received 11 February 2004; revised 25 March 2004; accepted 20 May 2004; published 22 June 2004.

[1] Using collocated measurements of aerosol radiative properties and radiative fluxes, aerosol radiative forcing is estimated at a tropical urban site in India, located between the sub-continent and the Indian Ocean Experiment [INDOEX] sites. Observed sun/sky radiance data are used to derive aerosol spectral optical depth, single scattering albedo [SSA], asymmetry parameter, precipitable water and total column ozone. These serve as inputs to a radiative transfer model, to estimate aerosol forcing at the surface, the top-of-the atmosphere [TOA] and the atmosphere. During the dry season of 2001 and 2002 [November-April], these were found to be -33, 0 and 33 Wm⁻², respectively. Using measured radiative fluxes during different aerosol loading conditions yield a surface forcing of -31 Wm⁻². The surface forcing efficiency as computed from the two independent methods is found to be -88 and -84 Wm⁻² respectively, while mean SSA at 500 nm is found to be 0.81. INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 1610 Global Change: Atmosphere (0315, 0325); 1704 History of Geophysics: Atmospheric sciences. Citation: Pandithurai, G., R. T. Pinker, T. Takamura, and P. C. S. Devara (2004), Aerosol radiative forcing over a tropical urban site in India, Geophys. Res. Lett., 31, L12107, doi:10.1029/2004GL019702.

1. Introduction

[2] One of the major uncertainties in estimating the magnitude of climate change is related to the radiative forcing effects of aerosols [IPCC, 1996]. It is believed that the radiative forcing due to aerosols is comparable to that of greenhouse gases, but opposite in sign [IPCC, 2001]. On global scale, the direct radiative forcing of anthropogenic aerosols from sulfates, soot and organic aerosols is estimated to be in the range of -0.3 to -1.0 Wm⁻². Yet, these estimates are highly uncertain because of uncertainties in the optical properties and the spatial distribution of aerosols. At local scales, aerosol forcing can exceed the global values by an order of magnitude. Hence, there is a need to obtain information on radiative forcing in different regions of climatic significance. Recently, major field experiments such as the Tropospheric Aerosol Radiative Forcing Observational Experiment [TARFOX] [Russell et al., 1999], the Indian Ocean Experiment [INDOEX] [Ramanathan et al., 2001], and the Asia Pacific Regional Aerosol Characterization Experiment [ACE-Asia] [Huebert et al., 2003] have provided important information. However,

experiments of this type are scarce over the Indian subcontinent, especially over urban/industrial regions, a primary source for anthropogenic aerosols.

[3] The study reported on in this paper has been conducted over land, about 200 km from the Arabian Sea, and as such, can complement findings obtained during INDOEX. The main objective has been to estimate aerosol radiative forcing over an urban continental site, by simultaneously observing the major radiative parameters of aerosols, column water content and ozone, to be supplemented with modeling results. This study is believed to be the first to make use of collocated aerosol and radiation measurements at an Indian urban station.

2. Experimental Setup

[4] The instruments used in this study are installed on the roof of the Indian Institute of Tropical Meteorology [IITM], Pune [18°32'N, 73°51'E, 559 m AMSL] and are listed in Table 1. The Prede sky radiometer [POM-01 L, Prede Inc., Japan] that is widely used in the SKYNET aerosol-radiation network in Asia [http://atmos.cr.chiba-u.ac.jp/aerosol/ skynet] is operated to make sun/sky observations that can be used to retrieve radiative characteristics of aerosols. The instrument provides highly accurate angular and spectral scans with scan accuracy of $\pm 0.1^{\circ}$ with a maximum scan rate of 30 degrees/sec. Calibration methodology and data reduction procedures for this instrument are presented in Nakajima et al. [1996] and Boi et al. [1999]. The instrument can make measurements of both direct and diffuse sky radiances at pre-defined scattering angles at regular intervals. A Kipp and Zonen ventilated Pyranometer [Model CM21] is used to measure down-welling short-wave radiative fluxes. Down-welling long-wave radiative fluxes are acquired with a pyrgeometer [Eppley Model PIR] and used mainly to aid in selection of clear sky cases. Both sensors are ventilated and inspected and cleaned daily. A Microtops-II Sunphotometer/Ozonometer is a hand-held instrument consisting of 5 channels, used to measure total column ozone (TCO) and precipitable water content (PWC) using pre-calibrated constants [Morys et al., 2001]. CM21 pyranometer has an absolute accuracy of 5% but the error due to directional response can be ± 10 Wm⁻². Once a month, sky radiometer is operated to collect disc scans to estimate solid view angles at different wavelengths as part of the calibration and used in the data processing. Also, on very clear sky days, absolute calibration constant $V_0(\lambda)$ is estimated using the modified Langley plot technique and found to be consistent with the manufacturer calibration.

3. Methodology

[5] The sky radiometer was operated from December 2000 to April 2002 on clear-sky days at 15-minute

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Table 1. Instruments Used and Parameters Derived

Instruments Used	Parameters Derived
Sky radiometer	Aerosol optical depths, volume size distribution,
PREDE [POM-01L]	single scattering albedo, phase function, refractive
400, 500, 675, 870, 1020 nm	index, asymmetry parameter
Pyranometer	Down-welling Global Shortwave flux
Kipp & Zonen Model [CM21]	[0.305–2.8 µm]
Precision Infrared Radiometer	Down-welling Infrared radiative flux
Eppley Laboratory Inc.	[3.5–50 µm]
Microtops-II Sunphotometer/Ozonometer	Total column ozone and precipitable water content
305, 312, 320, 940 and 1020 nm	* *

intervals, to acquire direct solar and diffuse sky radiances at predefined scattering angles. The sun/sky radiance data have been analyzed by using the radiative transfer model SkyRadPack version 3.0 [*Nakajima et al.*, 1996] for retrieval of aerosol optical depth [AOD], SSA, volume size distribution, phase function and asymmetry parameter. Microtops sunphotometer/ozonometer was operated manually at 15-minute intervals to get TCO and PWC. One-minute averaged short and long-wave radiative fluxes were recorded continuously.

[6] Daytime-mean AOD, SSA and asymmetry parameter at 400, 500, 675, 870 and 1020 nm wavelengths as well as TCO and PWC were computed. These were used as inputs in the radiative transfer model Santa Barbara Discreteordinate Atmospheric Radiative Transfer (SBDART) [*Ricchiazzi et al.*, 1998] to simulate SW fluxes at the surface for aerosol laden and aerosol-free atmospheres.

4. Aerosol Radiative Forcing

[7] Direct aerosol radiative forcing (ARF) is defined as the difference in net fluxes at the surface or at the TOA, with and without aerosols. ARF has been estimated using two methods. First, we used sun/skyradiometer derived aerosol properties and Microtops sunphotometer-derived TCO and PWC, as inputs into the SBDART, to compute ARF. Next, we used surface radiation measurements only, for different aerosol conditions similar to *Conant* [2000]. A clear day (with AOD of 0.1) was selected as a reference day and the 24-hour average net flux has been computed by assuming a broad band surface albedo of 0.15. The difference between the average net flux on other clear sky days with respect to the reference day is correlated with the change in AOD, to estimate ARF.

[8] Using the 1-minute averaged short- and long-wave radiation data, cloud screening has been performed to select about 33 clear sky days to be used in this study. In addition, derived AOD and Angstrom exponent values that differ from the daily mean by 3σ (three standard deviation) were eliminated as cloudy data. Daily mean aerosol parameters, TCO and PWC were used to simulate clear sky radiative fluxes using the SBDART model for all the 33 experimental days. This model accounts for multiple scattering in a vertically inhomogeneous non-isothermal plane-parallel atmosphere. Other input parameters used by the program are the spectral solar radiation, the solar geometry, the surface reflective properties and a model atmosphere. Tropical model atmosphere [McClatchey et al., 1972] and a fixed surface albedo of 0.15 have been assumed and the discrete ordinate model was run with 8 streams. The output parameters that have been used in this study were the surface downward and TOA SW radiation per wavelength interval between 0.305 and $2.80 \,\mu m$. Estimated short-wave radiative fluxes integrated in the above spectral region at half-hourly intervals are compared with pyranometer observed fluxes and have been found to be within a mean bias of -3.8 Wm^{-2} (Figure 1). Fluxes at the surface and TOA were estimated under aerosol free atmospheres. Aerosol forcing was computed at half-hourly intervals and integrated over 24 hours to obtain daily values. Direct ARF at the surface and at the TOA estimated for the clear sky days is shown in Figure 2. ARF values ranged from



Figure 1. Scatter plot between pyranometer-observed and SBDART modeled SW fluxes.



Figure 2. Day-to-day variation in aerosol forcing estimated at the surface and at the TOA at Pune.



Figure 3. Scatter plot between aerosol forcing efficiency at the surface and at the TOA as a function of AOD at 500 nm.

-17 to -59 Wm⁻² at the surface and -7 to +9 Wm⁻² at the TOA for AODs range of 0.2 to 0.6. These surface forcing values were negatively correlated [-0.89] with daytime mean AOD at 500 nm. Slope of the regression line gives the aerosol forcing efficiency (F_{eff}) values of -73 and -6 Wm^{-2} , at the surface and at the TOA respectively. F_{eff} at surface or TOA is defined as the change in net flux [Δ F] at the surface or TOA per unit AOD at 500 nm. Feff obtained from the above regression method may not provide a representative value of surface forcing because of the day-to-day variation in the SSA and the asymmetry parameter [Conant, 2000]. Hence, the ratio between aerosol forcing and AOD has been used to compute forcing efficiency for individual experimental days. Day-to-day variation in Feff at the surface by both methods and at the TOA is shown in Figure 3. Mean F_{eff} values at the surface is found to be -88 and -84 Wm⁻², from method 1 and method 2, respectively. Mean F_{eff} at the TOA is found to be -1 Wm^{-2} . The sign of the aerosol forcing at the TOA depends on surface albedo, SSA and asymmetry parameter. Here the surface albedo was assumed constant. The relationship between aerosol forcing at TOA, SSA and asymmetry parameter were examined and high correlation (0.8) has been found between aerosol forcing and SSA as compared with the asymmetry parameter (Figure 4). The sign of the forcing changes from negative to positive for values of SSA below 0.80. To assess the effect of surface albedo, aerosol forcing values were derived by using (i) constant surface albedo of 0.15 (spectral independent) and (ii) spectrally dependent vegetation model [Reeves et al., 1975]. Feff has been found to be -83, 12 Wm^{-2} , at the surface and at the TOA, respectively, when spectrally dependent surface albedo was used. While not much change was found in Feff at the surface, at the TOA there was considerable change.

[9] The estimated surface F_{eff} in this study is higher than similar estimates made over the Indian Ocean during INDOEX, reported to be about -71 to -82 Wm⁻² [*Podgorny et al.*, 2000; *Satheesh and Ramanathan*, 2000; *Conant*, 2000; *Bush and Valero*, 2002]. *Babu et al.* [2002] used black carbon measurements and estimated the values



Figure 4. (top) Correlation between aerosol forcing at the TOA and SSA. (bottom) Same as (top) but with asymmetry parameter.

of SSA using the Optical Properties of Aerosols and Clouds [OPAC] code [*Hess et al.*, 1998]. Radiative forcing efficiency of -97 Wm^{-2} was estimated over Bangalore, India [13°N, 77°E] [*Babu et al.*, 2002], using model-values of SSA [0.73] and asymmetry parameter. Comparison of F_{eff} between the present study, INDOEX and Bangalore is shown in Figure 5. F_{eff} at the surface and at the TOA increases with decrease in values of SSA. The positive and negative signs of aerosol forcing at the TOA may be due to differences in SSA [*Horvath et al.*, 2002; *Babu et al.*, 2002].



Figure 5. Aerosol radiative forcing efficiency at the surface, TOA and atmosphere and their comparison with previous investigations over India.



Figure 6. Monthly mean aerosol forcing at the surface observed during dry seasons of years 2001 and 2002.

Uncertainty in optical thickness retrieval is 0.02 with a consequent error in forcing of about 4%. SSA retrievals have an estimated error of 0.02 to 0.06 [maximum error of 0.06 is for low optical depths (<0.2) and low zenith angles (<15 deg) retrievals] and the associated uncertainty in forcing is about 10% to 25%. The average SSA estimated for the days considered in the present study is 0.81 at 500 nm. This low value is mainly due to the high BC content from the large number of diesel vehicles [Dickerson et al., 2002], from fossil fuel combustion, industrial emissions and biomass burning practices. Relative humidity at Pune during winter season [December-February] is about 30% and during pre-monsoon [March-May] it is about 50-70%. As F_{eff} is a strong function of SSA, forcing has been estimated for each month by using monthly mean aerosol radiative parameters in a radiative transfer model. Monthly mean radiative forcing at the surface and the year-to-year variability is shown in Figure 6. Seasonal mean surface forcing values are -23 and -26 Wm⁻² for years 2000-01 and 2001-02, respectively.

5. Conclusions

[10] 1. Simultaneous measurements of aerosol radiative properties and radiative fluxes were analyzed to study the effect of aerosols on shortwave radiation. The aerosol radiative forcing efficiency in the broadband shortwave is -88 Wm^{-2} , at the surface, which is slightly higher than obtained during INDOEX. At the TOA the forcing efficiency is -1.0 Wm^{-2} , which is low as compared to INDOEX.

[11] 2. Mean aerosol radiative forcing values at the surface, TOA, and atmosphere during dry seasons of years 2001 and 2002 are -33, 0, 33 Wm⁻², respectively. The small effect on TOA reflected radiation in urban stations is mainly due to the low single scattering albedo and associated enhanced atmospheric absorption.

[12] 3. Differences of 3 Wm^{-2} in surface radiative forcing were found during two dry seasons [November to April] of 2000–01 and 2001–02.

Agency of Japan [NASDA] to the University of Maryland. The authors would like to thank Dr. G. B. Pant, Director, IITM for his encouragement of this joint research work. Thanks are due to Prof. T. Nakajima, CCSR, Tokyo University for providing access to SkyRadPack code. Thanks are also due to Dr. R. S. Maheskumar, Mr. K. K. Dani, Mr. S. Saha, Mr. S. Sonbawne for their help in skyradiometer operation.

References

- Babu, S. S., S. K. Satheesh, and K. K. Moorthy (2002), Aerosol radiative forcing due to enhanced black carbon at an urban site in India, *Geophys. Res. Lett.*, 29(18), 1880, doi:10.1029/2002GL015826.
- Boi, P., G. Tonna, G. Dalu et al. (1999), Calibration and data elaboration procedure for sky irradiance measurements, *Appl. Opt.*, 38(6), 896–907.
- Bush, B. C., and F. P. J. Valero (2002), Spectral aerosol radiative forcing at the surface during the Indian Ocean Experiment (INDOEX), *J. Geophys. Res.*, 107(D19), 8003, doi:10.1029/2000JD000020.
- Conant, W. C. (2000), An observational approach for determining aerosol surface radiative forcing: Results from the first field phase of INDOEX, *J. Geophys. Res.*, 105(D12), 15,347–15,360.
- Dickerson, R. R., M. O. Andreae, T. Campos et al. (2002), Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry, J. Geophys. Res., 107(D19), 8017, doi:10.1029/2001JD000501.
- Hess, M., P. Koepke, and I. Schult (1998), Optical properties of aerosols and clouds: The software package OPAC, *Bull. Am. Meteorol. Soc.*, 79, 831–844.
- Horvath, H., L. Alados Arboledas, F. J. Olmo et al. (2002), Optical characteristics of the aerosol in Spain and Austria and its effect on radiative forcing, *J. Geophys. Res.*, 107(D19), 4386, doi:10.1029/2001JD001472.
 Huebert, B. J., T. Bates, P. B. Russell et al. (2003), An overview of ACE-
- Huebert, B. J., T. Bates, P. B. Russell et al. (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.
- Intergovernmental Panel on Climate Change (IPCC) (1996), *Climate Change 1995: The Science of Climate Change*, edited by J. T. Houghton et al., 572 pp., Cambridge Univ. Press, New York.
- Intergovernmental Panel on Climate Change (IPCC) (2001), Climate Change 2001: The Scientific Basis: Contribution of Working Group 1 to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by J. T. Houghton et al., 881 pp., Cambridge Univ. Press, New York.
- McClatchey, R. A., R. W. Fenn, J. E. A. Selby et al. (1972), Optical properties of the atmosphere, 3rd ed., *Environ. Res. Pap. 411*, 108 pp., Air Force Cambridge Res. Lab., Hanscom AFB, Mass.
- Morys, M., F. M. Mims III, S. Hagerup et al. (2001), Design, calibration, and performance of MICROTOPS II handheld ozone monitor and Sun photometer, J. Geophys. Res., 106, 14,573–14,582.
- Nakajima, T., G. Tonna, R. Rao et al. (1996), Use of sky brightness measurements from ground for remote sensing of particulate polydispersions, *Appl. Opt.*, 35, 2672–2686.
 Podgorny, I. A., W. C. Conant, V. Ramanathan, and S. K. Satheesh (2000),
- Podgorny, I. A., W. C. 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.
- Ramanathan, V., et al. (2001), Indian Ocean Experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, *J. Geophys. Res.*, 106(D22), 28,371–28,398.
- Reeves, R. G., A. Anson, and D. Landen (Eds.) (1975), *Manual of Remote Sensing*, 1st ed., 2144 pp., Am. Soc. of Photogramm., Falls Church, Va. Ricchiazzi, P., S. Yang, C. Gautier, and D. Sowle (1998), SBDART: A
- Ricchiazzi, P., S. Yang, C. Gautier, and D. Sowle (1998), SBDART: A research and teaching software tool for plane-parallel radiative transfer in the Earth's atmosphere, *Bull. Am. Meteorol. Soc.*, 79, 2101–2114.
- Russell, P. B., J. M. Livingston, P. Hignett et al. (1999), Aerosol-induced radiative flux changes off the United States mid-Atlantic coast: Comparison of values calculated from Sun photometer and in situ data with those measured by airborne pyranometer, J. Geophys. Res., 104(D2), 2289– 2307.
- 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.

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^[13] Acknowledgments. This work was supported by DST-NSF Grant No. DST/INT/US [NSF-RP053] to the IITM and to the University of Maryland, and grant RDC101GC1 from the National Space Development

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