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Origin of carbonaceous aerosols over the tropical Indian Ocean: Biomass burning or fossil fuels?

T.Novakov,[1] M.O. Andreae,[2] R. Gabriel,[2] T.W. Kirchstetter,[1] O.L. Mayol-Bracero,[2] and V. Ramanathan[3]

Abstract. We present an analysis of the carbon, potassium and sulfate content of the extensive aerosol haze layer observed over the tropical Indian Ocean during the Indian Ocean Experiment (INDOEX). The black carbon (BC) content of the haze is as high as 17% of the total fine particle mass (the sum of carbonaceous and soluble ionic aerosol components) which results in significant solar absorption. The ratio of black carbon to organic carbon (OC) (over the Arabian Sea and equatorial Indian Ocean) was a factor of 5 to 10 times larger than expected for biomass burning. This ratio was closer to values measured downwind of industrialized regions in Japan and Western Europe. These results indicate that fossil fuel combustion is the major source of carbonaceous aerosols, including black carbon during the events considered. If the data set analyzed here is representative of the entire INDOEX study then fossil fuel emissions from South Asia must have similarly contributed to aerosols over the whole study region. The INDOEX ratios are substantially different from those reported for some source regions of South Asia, thus raising the possibility that changes in composition of carbonaceous aerosol may occur during transport.

1. Introduction

The tropical Indian Ocean is a unique region of the world. Here, northern polluted air connects directly with pristine air from the Southern Hemisphere by a cross-equatorial monsoon flow into the Intertropical Convergence Zone (ITCZ). This northeast-to-southwest monsoon occurs every year between December and April, advecting polluted air masses from the Indian subcontinent [*Ramanathan et al,* 1996; *Krishnamurty et al.,* 1998] over the Bay of Bengal and the Arabian Sea. This flow, as observed during the Indian Ocean Experiment (INDOEX) [*Ramanathan et al.,* 1999; *Jayaraman et al.,* 1998; *Rajeev et al.*, 2000], results in the buildup of an extensive aerosol layer over the northern Indian Ocean. A unique characteristic of this layer is its light-absorbing nature, which reduces the diurnal average surface solar heating of the ocean by as much as 10-40 W m^{-2} (*Ramanathan et al.,* 1999; *Jayaraman et al.,* 1998; *Meywerk and Ramanathan*, 1999]. During wintertime, the aerosol layer is characterized by single scattering albedo around 0.9 or lower [*Satheesh et al.,* 1999] and optical depths in the range of 0.2 to 0.7 [*Ramanathan et al.,* 1999; *Krishnamurty et al.,* 1998]. Inferences from modeling studies show that the aerosol absorption is due to high concentrations of combustion-generated black carbon (BC) [*Satheesh et al.,* 1999], and that about 70% of the total aerosol burden could be derived from anthropogenic sources [*Krishnamurty et al.,* 1998; *Satheesh et al.,* 1999]. A largely unanswered, but important issue concerns the relative influence of the different anthropogenic sources on the BC and organic carbon (OC) content of the haze over the Indian Ocean. Specifically, what are the relative contributions of biomass and biofuel burning which, together with fossil fuel combustion, are the main aerosol sources in the Indian subcontinent [*Streets and Waldhoff*, 1998]? In this paper, we describe the results of an empirical approach to assess the contribution of these sources to the carbonaceous fraction of submicron aerosols measured during INDOEX.

We use the following reasoning in our approach. Both BC and OC are products of incomplete combustion of carbon-containing fuels. Since the emitted mass concentrations of OC and BC depend on the fuel types and combustion efficiencies, the BC/TC (TC = BC + OC) and BC/OC ratios for biomass and fossil fuel are different. Furthermore, elevated fine potassium (a tracer for biomass) to BC ratios (K^+/BC) are known indicators of biomass burning. Because fossil fuel combustion is the principal source of anthropogenic sulfate, the aerosol SO_4^{2-}/BC and SO_4^{2-}/TC ratios for fossil fuel burning should be higher than for biomass burning. Our analysis is based on comparisons of INDOEX data with published data from: (a) biomass smoke observations, (b) ambient measurements at a few locations possibly characteristic of INDOEX sources, and (c) measurements from other oceanic and continental regions influenced by fossil fuel burning. Because of the uniqueness of the sources, emissions from South Asia should be discernibly different than emissions from industrialized countries (i.e. Japan) where both the fuels and combustion technologies are different. These differences can be inferred from the following considerations.

Carbonaceous aerosols in India are derived from widespread use of biofuels, coal and petroleum products. In 1996, coal and oil burning contributed 70% and 23%, respectively to fossil-fuel-derived CO_2 emissions in India [*Marland et al.,* 1999]. A large fraction of coal is burned in households and poorly controlled industrial and commercial sources. Unlike modern power plants, these sources are important contributors to OC and BC. In 1985-86, about 370 Tg of biomass (excluding agricultural clearing) was burned in India; about 60% were biofuels used by the households [*Joshi*, 1991]. For comparison, about 220 Tg of coal were used in the same period [*EIA*, 1999]. A recent analysis by *Streets and Waldhoff* [1999] also suggests that fossil fuel and biofuel burning in India releases comparable amounts of carbon-containing trace gases to the atmosphere.

The extensive biofuel burning prevalent in India and surrounding countries is a major source of OC. (Estimated global organic aerosol emissions from biofuels is 9.4 Tg per year, about 1/2 of the emissions from burning of forests and savannas [*Liousse et al.*, 1996].) It is important to note that consumption of distillate petroleum (i.e., diesel fuel) products in 1997 in India was about six times that of gasoline, because diesel engines produce more BC than gaso-

[1]Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, California
[2]Biogeochemistry Department, Max Planck Institute for Chemistry, Mainz, Germany
[3]Scripps Institution of Oceanography, University of California, San Diego, La Jolla, California

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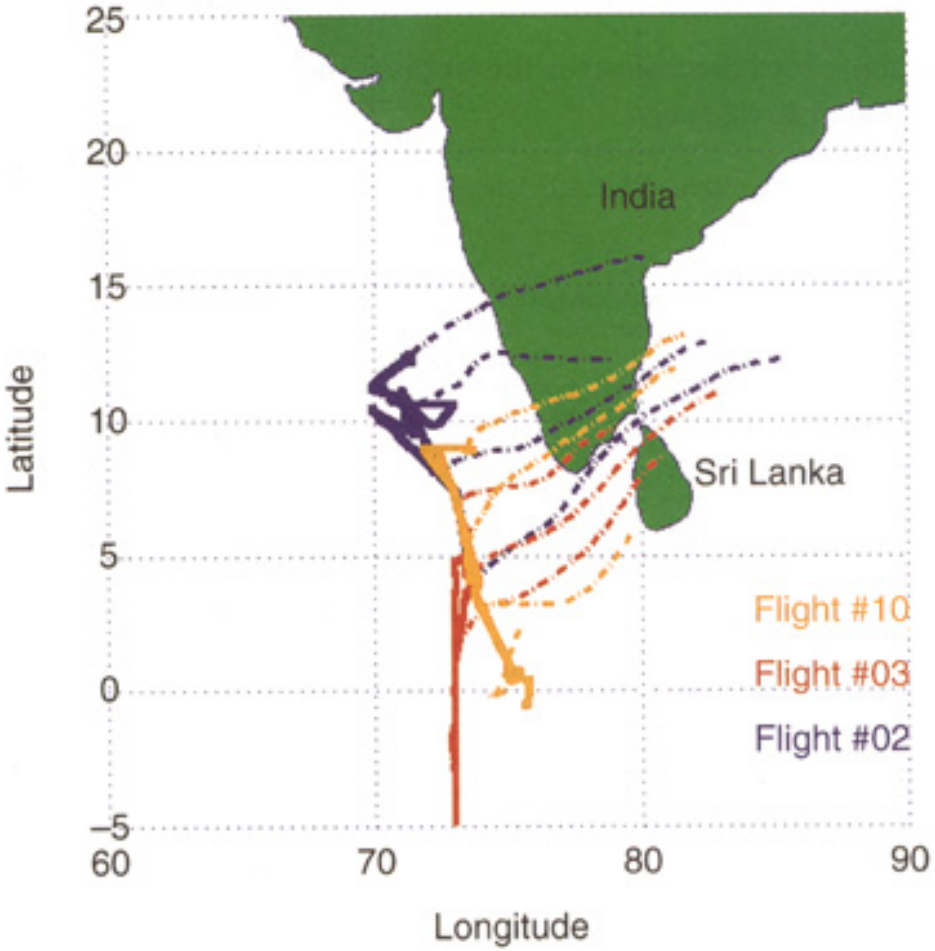

Fig. 1. Flight paths (solid lines) and isobaric backtrajectories calculated for 1500 m altitude (dashed lines) corresponding to the three INDOEX flights.

line [*Kleeman et al.*, 2000]. A similarly large fraction of distillate fuel is used in Bangladesh, Pakistan and Sri Lanka. [*EIA,* 1999].

On the other hand, 30% coal and 56% of oil burning in Japan contributed to fossil fuel CO_2 emissions in 1996 [*Marland et al.,* 1999]. In Japan and Korea, the distillate fuel consumption was only 1.4 and 2.4 times higher than gasoline consumption [*EIA,* 1999]. Furthermore, biofuel use in these countries is minimal [*Streets and Waldhoff*, 1999].

2. Results and discussion

Data were obtained by analyzing filter samples of aerosol particles (with diameters smaller than 1.3 μm) collected from the National Center for Atmospheric Research (NCAR) C130 aircraft during Research Flights 2 (February, 18), 3 (February 20), and 10 (March 9, 1999) [Details of sampling and analytical methods are described in Mayol-Bracero et al.; Gabriel et al., *J. Geophys. Res.* special INDOEX issue, in preparation].

Aerosol samples were taken at approximately constant altitudes in the boundary layer (lower than 2 km) during these three long trans-equatorial flights north of the ITCZ from about 8°N to about 8°S. The location of the ITCZ inferred from satellite observations was between 5°S to 2°S during these flight days. Back trajectories (Fig.1) demonstrate that most of the trajectories originate in the Indian sub-continent and Sri Lanka. Although the flight paths varied over the study region, the data (Table 1) corresponding to these flights show a remarkable consistency among the various mass concentration ratios. Near constancy of BC/TC and BC/OC ratios and the fact that TC and BC are highly correlated ($r^2 = 0.81$) are strong indications of the predominantly primary nature of INDOEX aerosol carbon. (Prior to the experiment, it was assumed that secondary OC produced by photochemical reactions of biogenic gaseous compounds would be a major contributor to the carbonaceous aerosol mass [*Ramanathan et al.*, 1996].)

Table 1 shows how the BC/TC and BC/OC ratios determined during these flights compare with those measured in studies of biomass burning in Brazil [*Ferek et al.,* 1998] and Ivory Coast [*Cachier et al.,* 1991]. INDOEX BC/TC and BC/OC ratios are about 5 and 8 times higher, respectively, than the ratios for Brazil and Ivory Coast. The potassium to BC ratio (K^+/BC) for biomass smoke is about 3.5 to 7 times higher than found in the INDOEX aerosol. Aerosol sulfate to TC and BC ratios (SO_4^{2-}/TC and SO_4^{2-}/BC) were up to an order of magnitude higher than in biomass smoke. These data suggest that the INDOEX carbonaceous aerosols are significantly influenced by fossil fuel combustion sources whose effluents are characterized by higher BC/TC and BC/OC and higher SO_4^{2-}/TC and SO_4^{2-}/BC ratios (compared with biomass burning). For biomass burning emissions, we expect a BC/TC ratio of about 0.11. This value is quite representative of a large set of emission measurements from diverse types of biomass burning, including biofuel use [M.O. Andreae and P. Merlet, Emissions from biomass burning, manu-

Table 1. Comparison of INDOEX data with data from biomass burning studies and ambient measurements in Pakistan

Flight Location	BC/TC	BC/OC	K^+/BC	SO_4^{2-}/TC	SO_4^{2-}/BC	Size cut (μm)	References
			INDOEX				
Flight 2	0.51±0.03	1.0±0.1	0.17±0.07	1.3±0.4	2.5±0.8	<1.3	*This work*
Flight 3	0.49±0.08	0.93±0.27	0.16±0.04	1.2±0.4	2.4±0.6	<1.3	*This work*
Flight 10	0.43±0.11	0.83±0.33	0.11±0.01	0.9±0.3	2.1±0.7	<1.3	*This work*
			Biomass burning				
Brazil	0.10±0.03	0.12±0.03	0.52±0.11	0.03±0.01	0.28±0.13	<4.0	*Ferek et al.*, 1998
Ivory Coast	0.12±0.07	0.14±0.08	NA	NA	NA	<5.0	*Cachier*, 1991
			Urban, industrial and rural Pakistan				
Lahore	0.19±0.03	0.24±0.04	0.23±0.10	0.21±0.04	1.1±0.1	<10	*Smith et al.*, 1996

± Standard devitions of reported mean values; NA not avalilable

script in preparation, 2000]. The INDOEX BC/TC ratio is four times higher and fairly consistent throughout the period. For the three flights discussed here, the average BC/TC is 0.46±0.07, whereas this ratio for the entire (C-130) INDOEX data set from the Indian plume averages 0.42±0.09. If we assume that fossil fuel sources (diesel, industry, household coal) have a characteristic BC/TC ratio of about 0.5±0.05, then biomass burning would contribute have a 21±8% to carbonaceous aerosols. Conversely, fossil fuel combustion would account for about 80% of the average INDOEX carbonaceous aerosol mass.

An alternate approach for discerning source contributions is to compare INDOEX data with those obtained in source regions in India and other locations in Asia where fossil fuel combustion sources dominate (i.e. Japan and South Korea). Unfortunately, to our knowledge, there are no reported measurements of carbonaceous aerosols in India. The only measurement of some relevance to this analysis was conducted in Lahore, Pakistan [*Smith et al.*, 1996] using non size-segregated sampling at the city center, an industrial site and a rural site. The annual mean ratios (1992-93) for the three sites are given in Table 1. These data show that the average BC/TC ratio for the three INDOEX flights is 2.6 times and BC/OC is about 4 times higher than the Lahore ratio. The SO_4^{2-} content of INDOEX samples relative to TC and BC is about 5 and 2 times higher than in the ambient Lahore aerosol. The average INDOEX K^+/BC ratio is slightly lower than for Lahore. Most of these differences may be attributable to the fact that the Lahore samples contained coarse urban aerosols which are typically high in OC, but contain little or no BC [*Kleeman and Cass*, 1998]. The enhanced SO_4^{2-}/BC ratio in the INDOEX plume as compared to the Lahore aerosol would be consistent with a longer time (about 2 to 4 days) having been available to oxidize SO_2 to SO_4^{2-} during long-range transport.

In Table 2, we summarize data obtained in urban sites in Japan, Korea and a few marine locations in the region. Surprisingly, the BC/TC and BC/OC ratios for INDOEX are remarkably similar to those measured at these fossil fuels dominated sites. We also note that the INDOEX SO_4^{2-}/TC and SO_4^{2-}/BC ratios are similar to those measured at these polluted marine sites, even though the South Asian sources are radically different from those in Japan. For comparison, Table 2 also includes the pertinent ratios measured over the eastern Atlantic in 1997 [*Novakov et al.*, 2000] during the ACE-2 program. The ACE-2 BC/TC and BC/OC ratios are similar to INDOEX values. (We note that the ACE-2 SO_4^{2-}/TC ratio is higher because of the influence of high sulfur (coal) burning sources.) The results summarized in Table 2 lead to the deduction that the fine aerosol BC, OC, and SO_4^{2-} in the Indian plume during INDOEX was dominated by fossil fuel sources, particularly by BC-rich emissions from diesel engines and open coal burning. To further explore these questions, size-resolved compositional data on the aerosol composition

Table2. Comparison of INDOEX ratios with those measured elsewhere in Asia and eastern Atlantic

Location	BC/TC	BC/OC	SO_4^{2-}/TC	Size cut (μm)	References
INDOEX (flights 2,3,10)	0.46±0.07	0.90±0.24	1.0±0.4	<1.3	*This work*
		Urban Southeast Asia			
Sapporo, annual 1982	0.54±0.14	1.19±0.37	0.47±0.08	<8.0	*Ohta and Okita*, 1990
Nagoya, annual 1984-86	0.44±0.034	0.80±0.11	NA	<10	*Kadowaki*, 1990
Sendai, annual 1986-87	0.37±0.031	0.60±0.04	0.37±0.03	<1.0	*Hayasaka et al.*, 1992
Seoul, summer 1994	0.43±0.05	0.76±0.1	NA	<2.5	*Kim et al.*, 1999
		Marine Southeast Asia			
Hachijo-Jima, winter 1981	0.41±0.07	0.70±0.31	0.91±0.15	<10	*Ohta and Okita*, 1984
Chichi-Jima, winter 1981	0.54±0.21	1.2±1.4	1.1±1.6	<10	*Ohta and Okita*, 1984
South coast of Japan, winter 1982	0.36±0.04	0.56±0.13	1.1±0.3	<0.6	*Ohta and Okita*, 1984
		Eastern Atlantic			
ACE-2 summer 1997	0.38±0.12	0.58±0.22	5.5±2.8	<1.0	*Novakov et al.*, 2000

± Standard devitions of reported mean values; NA not available

in the suspected source regions in India should be known. This information, unfortunately, does not exist at present.

Finally, our analysis inevitably relies on comparing INDOEX data with published data that may be biased by sampling artifacts. For example, organic vapor adsorption on the filter material, if unaccounted for, may result in an overestimation of aerosol OC concentrations. While the INDOEX and ACE-2 data were corrected for this artifact, other data we use were not. The magnitude of the overestimation for urban samples is generally in the range of 10 - 30% [*Chow et al.*, 1994] and decreases for more loaded samples [*Rau,* 1989]. The bias in the OC data available in the literature is probably less than 30% because these were most likely obtained with heavier sample loadings, and therefore should not influence our overall conclusions.

3. Conclusions

Given the large carbon monoxide concentrations and prevalent biomass and biofuel use in the Indian subcontinent, it is tempting to suggest that burning of these fuels is the main source of the highly absorbing (low single scatter albedo) haze observed during INDOEX [*Ramanathan et al.,* 1999]. The results of this analysis show that both biomass and fossil fuel combustion contribute to the carbonaceous particles, including the light absorbing black carbon. However, the contribution of fossil fuel burning (by transportation, industrial, and domestic sectors) is considerably larger and can be as much as 80% of the total. A more rigorous attribution of carbonaceous aerosols over the Indian Ocean to specific sources must await the results of further studies.

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T. Novakov and T.W.Kirchstetter, Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, MS-73, 1 Cyclotron Road, Berkeley, CA 94720, USA.
(tnovakov@lbl.gov)

M.O. Andreae, R. Gabriel, O.L. Mayol-Bracero, Biogeochemistry Department, Max Planck Institute for Chemistry, P.O. Box 3060, D-55020 Mainz, Germany

V. Ramanathan, Scripps Institution of Oceanography, University of California, San Diego, La Jolla, California 92093, USA.

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