Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry

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[1] Air from South Asia carries heavy loadings of organic and light-absorbing aerosol but low concentrations of ozone. We investigate ambient pollutant concentrations measured during the Indian Ocean Experiment (INDOEX), and we estimate emissions to determine the origin, magnitude, and impacts of air pollution from South Asia and to understand better the uncertainty in emission inventories. In India, the preponderance of motorcycles with small, two-stroke engines and the practice of adulterating gasoline with kerosene lead to high CO emission factors; for 1999, we estimate release of 15 Tg yr^{-1} from fossil fuel use and 40 Tg yr^{-1} from biomass burning. With the addition of isoprene oxidation, the total CO emissions were 67 Tg yr^{-1} from India and 87 Tg(CO) yr^{-1} from all of South Asia. These values indicate a somewhat larger contribution from fossil fuels but slightly lower overall emissions when compared to prior emission inventories. Two-stroke engines also exhibit high emission factors for volatile organic compounds (VOC) and particulate organic matter but produce only modest amounts of NO_x. Near sources in India, VOC to NO_x ratios appear too high for efficient O_3 formation, although other factors probably contribute to observed low O_3 mixing ratios. An inventory based on source characteristics and known emission factors for black carbon (BC) from South Asia yields 0.7 Tg yr^{-1} (upper limit of about 1.0 Tg yr^{-1}) with biomass burning as the dominant source. We can test this inventory with measurements of ambient CO and BC—ship, island, and aircraft observations of air from South Asia all show a positive correlation between CO and BC ($r^2 = 0.71–0.81$). Such strong correlations have also been observed over North America and Europe, but with a lower BC/CO slope. Ambient concentrations indicate high BC emission from South Asia: 2–3 Tg(BC) yr^{-1}. This disagreement with emission inventories demonstrates the need for direct measurements from sources in India. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* Soot, black carbon, CO, emissions, India

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1. Introduction

[2] Carbonaceous aerosols play an important role in the Earth's radiative balance and in climate [*Intergovernmental Panel on Climate Change* (*IPCC*), 1996; *National Research Council* (*NRC*), 1996; *Jacobson*, 2001], but little is known of their concentrations, distributions, and source strength, especially for the developing world. Aerosol chemistry over the Indian Ocean was largely unknown until recently when investigators with Indian Ocean Experiment (INDOEX) organized several field campaigns to measure these species [*Rhoads et al.*, 1997; *Mitra*, 1999; *Novakov et al.*, 2000; *Mayol-Bracero et al.*, 2002; *Quinn et al.*, 2002; *Sarkar et al.*, 2001; *Neususess et al.*, 2002] (W. P. Ball et al., Bulk and size-segregated aerosol composition: Continental impacts during INDOEX 1999, submitted to *Journal of Geophysical Research*, 2001). Measurements in the marine boundary layer (MBL) indicate high concentrations of particulate organic carbon, OC, and black carbon, BC, also-called elemental carbon or soot. Even higher concentrations were found in the lower free troposphere over the ocean, referred to as the residual continental boundary layer, rCBL (A. Clarke et al., An overview of the C-130 flight missions and measurements during INDOEX, submitted to *Journal of Geophysical Research*, 2001). These aerosols exert a profound influence on the amount of radiation reaching the surface of the Indian

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Ocean [*Jayaraman et al.*, 1998; *Satheesh et al.*, 1999; *Ramanathan et al.*, 2001].

[3] Ozone likewise plays an important role in radiative balance, and is central to tropospheric photochemistry. INDOEX investigators organized several field campaigns to make the first extensive measurements of ozone and associated trace gases over the region [*Rhoads et al.*, 1997; *Lal et al.*, 1998; *de Laat et al.*, 1999; *Lal and Lawrence*, 2001; *Lelieveld et al.*, 2001; *Lobert and Harris*, 2002; *Muhle et al.*, 2002; *Stehr et al.*, 2002]. Results indicate that the atmosphere over the Northern Indian Ocean is heavily polluted with aerosols and volatile organic compounds (VOC), but concentrations of ozone are low. Published measurements of air quality over India are inadequate to characterize typical pollutant levels, but ozone concentrations appear to be modest, rarely reaching 100 ppb [*Pandey and Agrawal*, 1992; *Shende et al.*, 1992; *Varshney and Aggarwal*, 1992; *Khemani et al.*, 1995; *Naja and Lal*, 1996; *Singh et al.*, 1997; *Lal et al.*, 2000].

[4] In energy use and combustion methods, South Asia deviates considerably from North America and Europe. In the MBL over the Northern Indian Ocean, the ratio of BC and OC to sulfate is much higher and the concentration of ozone much lower than over the North Atlantic Ocean. [e.g., *Wolff et al.*, 1986; *Galloway et al.*, 1993; *Raes et al.*, 2000]. For example, on the west coast of Ireland when the winds are from Europe BC concentrations rarely exceed 0.3 $\mu g\ m^{-3}$ [*Jennings et al.*, 1996; *Cooke et al.*, 1997], but the average BC concentration found from the ship on the Indian Ocean downwind of India was 1.7 $\mu g\ m^{-3}$ [*Neususess et al.*, 2002]. At the Kaashidhoo Climate Observatory (KCO), a surface site on the island of Kaashidhoo in the Maldives (4.97°N, 73.47°E), an average of 1.1 to1.9 $\mu g\ m^{-3}$ total BC in particle sizes smaller than 1.8 μm aerodynamic diameter was observed [*Chowdhury et al.*, 2001]. Aircraft measurements showed an average of 2.3 $\mu g\ m^{-3}$ in the MBL and 3.2 $\mu g\ m^{-3}$ in a pollution layer aloft [*Mayol-Bracero et al.*, 2002]. *Novakov et al.* [2000] examined OC/BC ratios and concluded that fossil fuel combustion is the dominant source of absorbing aerosol. *Mayol-Bracero et al.* [2002] examined these and relative concentrations of CO, K^+, CH_3CN, and sulfate to reach a similar conclusion, but noted emission inventories and ambient measurements yield conflicting indications of whether fossil- or biofuel use dominates pollution production. They suggest that the nature of emissions from India must differ substantially from that of North America and Europe. Guazzotti et al. (S. A. Guazzotti et al., Characterization of pollution outflow from India and Arabia: Biomass burning and fossil fuel combustion, submitted to *Journal of Geophysical Research*, 2001), using single-particle analysis concluded that biomass burning was the main source of black carbon.

[5] Prior estimates of air pollutant emissions from South Asia, defined as Bangladesh, Bhutan, India, Nepal, Pakistan, and Sri Lanka, indicate CO production approaching or even exceeding production in North America [*Olivier et al.*, 1994; *Galanter et al.*, 2000], but substantially less NO_x, HC, and SO_2. *Olivier et al.* [1994] suggest that only 4% of the CO emitted from South Asia arises from fossil fuel combustion.

[6] Published emissions estimates suggest modest BC from India, but point out the great uncertainty associated with bottom-up (based on individual sources) emission inventories, especially for the developing world [*Penner et al.*, 1993; *Cooke and Wilson*, 1996; *Liousse et al.*, 1996; *Cooke et al.*, 1999; *Reddy and Venkataraman*, 2000]. In this paper we use two, independent approaches to make new emissions estimates of CO and BC from South Asia and compare them to in situ measurements as a first step toward quantifying the origin, magnitude, impacts, and uncertainty of those emissions.

2. Emissions From India

[7] The sources of air pollution in India are uncertain and unlike those of North America or Europe. We apply several approaches to determine the magnitude of these emissions. First we examine fuel consumption in broad terms to identify the relevant types of combustion and to estimate CO release based on the mass of fuel consumed—a top-down approach. These results are compared to an independent method where emission inventories for CO and BC are calculated as the product of numbers of vehicles, average km driven, and average emission factors—a bottom-up approach. Ambient measurements of both CO and BC are then examined to provide an independent appraisal of BC emissions.

2.1. Carbon Monoxide and Hydrocarbons: Estimates From Fuel Consumption

[8] In India, approximately 2/3 of the motor vehicles are "two-wheelers" or motorcycles; roughly 10% are "three-wheelers" or "autorickshaws," peculiarly Indian motorized carts. The remainder are primarily automobiles, jeeps, and trucks powered by diesel engines [*Ministry of Surface Transport* (*MST*), 1993; *United Nations Environmental Program* (*UNEP*), 1999] (M. S. Reddy and C. Venkataraman, A 0.25 × 0.25 inventory of aerosol and sulfur dioxide emissions from India, I, Fossil fuel combustion, submitted to *Atmospheric Environment*, 2001a). A variety of factors economic and sociological contribute to high emissions per vehicle and per km driven [*World Health Organization, United Nations Environmental Program* (*WHO*/*UNEP*), 1992; *Central Pollution Control Board* (*CPCB*) *New Delhi*, 1989; *Halarnkar and Menon*, 1996].

[9] South Asian motorcycles are powered primarily by small, spark-ignited, two-stroke engines; three-wheelers are powered by similar, but somewhat larger engines [*WHO*/*UNEP*, 1992; *MST*, 1993; *UNEP*, 1999]. In four-stroke engines typical of North America and Europe, fuel is combusted and power obtained on alternate downward movements of the piston. Intake and exhaust are controlled by tight-fitting valves at the top of the cylinder. In two-stroke engines power is obtained on every downward thrust of the piston. These engines need no valves of the sort used in four-stroke engines, rather intake and exhaust occur simultaneously through vents near the bottom of the cylinder. Lubrication is achieved with oil mixed into the fuel. The simplicity of construction, low cost, and great power to weight ratio make two-stroke engines highly attractive to developing countries.

[10] Data on emissions from in-use, small, internal combustion engines are sparse. Only recently have direct measurements proved that motorcycles and other two-stroke engines discharge large amounts of pollution per unit of fuel consumed [*Patschull and Roth*, 1995; *Chan and Weaver*,

Table 1. Emissions from Small Internal Combustion Engines Without Pollution Controls

	HC[a]	CO	NO_x	PM
Two-Stroke Engines				
Chan and Weaver [1994][b]	570 (190)	1100 (530)	0.9	26
Chan et al. [1995][c]	204 (37)	405 (103)	0.38 (0.07)	NA
Patschull and Roth [1995][d]	NA	NA	NA	90
Weaver and Chan [1996][e]	330 (150)	280 (190)	0.66 (0.5)	24(7)
Bishop et al. [1999][f]	331 (6)	460 (2)	NA	NA
Priest et al. [2000][g]	350 (180)	800 (200)	1.6 (1.2)	NA
Tsai et al. [2000][h]	380 (200)	750 (470)	0.60 (0.12)	NA
Das et al. [2001][i]	270	790 (390)	NA	NA
Estimate for India	400	800	0.6	90
Four-Stroke Engines				
Chan and Weaver [1994]	240	1540	5.2	NA
Chan et al. [1995]	32 (6)	200 (42)	10 (3)	NA
Priest et al. [2000]	80 (40)	850 (350)	8.5	NA
Tsai et al. [2000]	130 (20)	1440 (540)	1.25 (0.7)	NA

[a] All units are in grams of pollutant per kilogram of fuel; standard deviations are in parentheses.
[b] New motorcycles produced in Thailand, except NO_x data are from European motorcycles.
[c] New 50-cc two-stroke motorcycles and 125-cc four-stroke motorcycles.
[d] New, German, 80-cc engine under load.
[e] Data for in-use motorcycles in Bangkok in the 80th percentile for exhaust opacity; the value for PM is a lower limit.
[f] In-use, two-stroke, snowmobiles.
[g] In-use, two-stroke lawn mowers.
[h] Assuming 25 km/kg for in-use, two-stroke and 40 km/kg for in-use, four-stroke engines.
[i] From 66,000 in-use two-wheelers in Delhi.

1994; *Chan et al.*, 1995; *Weaver and Chan*, 1996; *Nuti*, 1998; *Tsai et al.*, 2000; *Das et al.*, 2001]. Because intake and exhaust occur simultaneously in two-stroke engines, some of the fresh charge mixes into the exhaust and is released to the atmosphere unburned. The carbon number distribution of VOC in the exhaust mirrors that of the fuel and is higher than in exhaust from four-stroke engines [*Gabele and Pyle*, 2000]. Lubricating oil is discharged as pyrolized organic aerosols; the ratio of BC to OC is apparently unknown. Because two-stroke engines generally run rich (with an air-fuel ratio below that required for stoichiometric combustion) high power but poor fuel economy are experienced; HC and CO emissions are high and NO_x emissions low relative to four-stroke engines (Table 1).

[11] Gasoline is taxed heavily and sells in India for the world market price—similar to that paid in Europe or Japan. Kerosene (petroleum distillates composed of relatively long-chain, high molecular weight hydrocarbons) is subsidized, and sells for about 1/5 the price. This leads to the practice of fuel adulteration—mixing kerosene into diesel fuel, and diesel or kerosene into gasoline [*Halarnkar and Menon*, 1996; *UNEP*, 1999; *Patra and Mishra*, 2000]. The fraction of kerosene in fuel can reach 50% [*Patra and Mishra*, 2000], but the extent of this practice and its full impact on emissions are unknown. In this section we assume that direct emissions of CO by in-use, two-stroke, spark ignited engines lie toward the high end of observed rates; we use 800 g(CO)/kg(fuel), the second highest values from Table 1. This value lies between the numbers obtained by *Bradley et al.* [1999] for direct measurements of CO emissions from vehicles in Bangkok, Thailand, 655 g(CO)/kg(fuel), and Katmandu, Nepal, 960 g(CO)/kg(fuel). *Das et al.* [2001] measured CO emissions form 66,000 two-wheelers in Delhi, and found an average of 3% by volume CO in the exhaust at idle. For a stoichiometric air/fuel mixture and a mean fuel formula of CH_2, this corresponds to 790 g CO per kg fuel, essentially identical to our estimate.

[12] Nonmethane hydrocarbons, especially long-chain hydrocarbons characteristic of unburned fuel, oxidize quickly in the atmosphere. We assume that 30% of the carbon emitted as HC is rapidly converted to CO [*Hanst et al.*, 1980] yielding an indirect production of 240 g(CO)/kg(fuel) for a total effective emission rate of 1040 g(CO)/kg(fuel). From these revised emission factors and the rate of consumption of petroleum products in India we can estimate the total amount of CO produced from spark ignited engines (Table 2). This table summarizes prior work on emissions from India.

[13] The total petroleum consumed in India in 1999 was about 96 Tg with gasoline accounting for about 6 Tg, diesel about 38 Tg, and the remainder used primarily as kerosene and fuel oil (Table 2). For this estimate we assume all spark-ignited engines operate on the two-stroke principle and burn 2/3 gasoline and 1/3 kerosene [see *Patra and Mishra*, 2000] for a total of 9 Tg(fuel) yr^{-1}. At 1040 g(CO)/kg(fuel) this yields 9.36 Tg(CO) yr^{-1}. Applying the same rate of fuel consumption, direct emissions of HC's from two-stroke engines are estimated to be 3.6 Tg yr^{-1}. We assume consumption of the remaining petroleum (9 from 96 or 87 Tg yr^{-1}) produces CO at an average rate of about 50 g(CO)/kg(fuel), appropriate for small-scale industrial and domestic combustion in the developing world [*Zhang et al.*, 1999; *Smith et al.*, 2000] (and allowing for evaporative emissions), to produce an additional 4.35 Tg. The total from petroleum use is 13.7 Tg(CO) yr^{-1}. Coal combustion contributes 1.8 Tg(CO) yr^{-1} (see section 2.2); we assume that natural gas use produces insignificant amounts of CO. The derived total production of CO from fossil fuel combustion in 1999 is then 15.5 Tg(CO) yr^{-1}.

2.2. Carbon Monoxide: Estimates From Emission Factors

[14] Detailed bottom-up emission inventories are being prepared for the year 2000 and can be used as an additional tool to interpret INDOEX observations. These inventories

Table 2. Literature Review on Emissions Factors and Fuel Consumption for India, 1999

Category	Fuel use, Tg yr^{-1}	Emission Factor, g(CO)/kg fuel	Emission Factor, g(BC)/kg(fuel)	Emission, Tg(CO) yr^{-1}	Emission, Tg(BC) yr^{-1}
Biofuel, dung	51[a], 106[b]	30–61[a], 80[c]		9.1[d], 2.9[a], 4.5[e]	
Biofuel brush/crop residue	62[a], 99[b]	36–170[a]	0.75[f],	10.6[d], 4.1[a], 10.0[e]	
Biofuel wood	203[a], 252[b]	42–170[a], 130[g], 81[c]	1.4[f],	22.4[d], 14.5[e]	
Total biofuel	316[a], 457[b]	78[h]	0.59[h], 1.0[e]	42[d], 16.7[a], 29[e], 42[i]	
Ag. savanna, forest burning	32[b]			52[i], 11.2[e,h]	
Total Biomass Burning		100[j]	2.2[j]	**85**[i], **31.5**[e,h], **72**[k]	**0.4**[l], **0.2**[b]
Petroleum, gasoline	6[m], 9[n]	800 (1040)[n,o]	(0.04–0.59)[e,p]	9.4[n]	
Petroleum distillate/diesel	38[m]	50[n]	(1.3–2.4)[p], (3.5–12)[e]	1.9[n]	
Other petroleum	52[m]	50[c,n]		2.5[n]	
Petroleum total	94.8[q], 97[m]			13.7[n]	
Coal	129[q]		(0.31–2.21)[e]	1.8[r]	
Total fossil fuel				**6**[i], **15.5**[n]	**0.1**[l], **0.25**[b]
Isoprene oxidation				11[n]	
Total				**85**[i], **64**[s], **66.7**[e,n]	**0.45**[b], **0.46**[l], **0.6**[t], **(0.5–0.8)**[e], **(2–3)**[n]

[a] *Zhang et al.* [1999], for 1990.
[b] *Reddy and Venkataraman* [2000], for 1990.
[c] *Smith* [1988].
[d] *Streets and Waldhoff* [1999], for 1990.
[e] This work, emissions factors.
[f] *Turn et al.* [1997].
[g] *EPA* [1996], for fireplaces.
[h] [*Andreae and Merlet*, 2002].
[i] *Olivier et al.* [1994], for 1990 all of South Asia.
[j] *Crutzen and Andreae* [1990].
[k] *Galanter et al.* [2000].
[l] *Mayol-Bracero et al.* [2002].
[m] Department of Energy International Energy Annual, 2000, available at http://www.eia.doe.gov/emeu/iea. 1998 data × 1.06.
[n] This work, section 2.1.
[o] For motorcycles, see Table 1.
[p] *EPA* [2001] (indicates maximum from vehicles selected for high emissions).
[q] BP AMOCO (http://www.bpamoco.com/centres/energy2002/index.asp, 2001) for 1999.
[r] Table 3.
[s] *IPCC* [1996], anthropogenic only.
[t] *Cooke et al.* [1999], "rest of Asia" × 0.75.

are assembled from disaggregated energy use and related activity data, coupled with appropriate emission factors for each activity type [*Streets and Tsai*, 2001, available at http://www.cgrer.uiowa.edu/people/carmichael/ACESS/Emission-data_main.html]. Because activity data for the year 2000 are not yet available, the approach uses a combination of model forecasts and trend extrapolations to assemble the activity data set. As a foundation for this analysis, the inventories use year-2000 energy forecasts (off a 1995 base) from the RAINS-Asia (regional air pollution simulation) model [*Foell et al.*, 1995; *Downing et al.*, 1997].

[15] The CO inventory for South Asia consists of estimates of emissions from industry and power (including fuel combustion and iron and steel manufacturing), residential combustion of biofuels and fossil fuels, mobile sources (transport), and the combustion of crop residues in the field after harvest. It does not include the burning of vegetation for land clearing or wildfires, best inferred from satellite observations or land-use surveys.

[16] The database for fuel use in India, especially residential combustion of biofuels, is considered to be reliable, as it builds upon the detailed study of renewable energy potential in India, recently completed by a consortium of organizations led by the *Tata Energy Research Institute* (*TERI*) [1999] (available at http://www.dow.wau.nl/msa/renewables/). Residential combustion of biofuels is the single largest contributor to CO emissions in South Asia. For the year 2000, 5300 PJ of woody biofuels (fuelwood and agricultural residues) and 1450 PJ of dried animal waste (dung) were burned [*TERI*, 1999]. Extending this work to the whole of South Asia, yields estimates of 6680 PJ of woody biofuels and 1720 PJ of dung [*Streets and Waldhoff*, 1998]. These biofuel estimates are made at the state level for India and therefore reflect regional differences in quantity and fuel type. Emission factors for residential fuel combustion are taken from a recent measurement program on Indian household stoves [*Zhang et al.*, 1999]: CO emission factors of 77.5 g kg^{-1} for woody biofuels and 43 g kg^{-1} for dung. This yields an estimate of 29.0 Tg of CO released in India, and 37.7 Tg for the entire South Asia region, from the residential combustion of biofuels (see Table 3). This can be compared to the estimate developed by *Zhang et al.* [1999] of 16.7 Tg for CO emissions from biofuel combustion in India for the year 1990/91. We calculate considerably lower emissions from the residential combustion of coal (at an emission rate of 74 g kg^{-1}) and kerosene (at 24 g kg^{-1}) in 2000, consistent with the assessment of *Zhang et al.* [1999]. In combination, these two source types generated only 0.3 Tg CO in South Asia in 2000.

[17] Vehicles represent the second major CO emitting category in India. In this approach, ten vehicle types are examined, including passenger cars, light-duty trucks, heavy-duty trucks, buses, motorcycles, and tractors. Each of the first four categories is further split into gasoline and

Table 3. CO Emissions (Gg) for South Asia, 2000

Region	Industry	Domestic		Transport			Field Combustion	Total
		Biofuels	Fossil Fuels	Cars	Trucks, Buses, Tractors	Motorcycles		
Bangladesh	91	2672	27	14	46	35	261	3146
Bhutan	0	90	0	1	1	1	1	95
India	1850	28989	154	2112	2092	6192	2512	43902
Nepal	1	1236	22	12	15	39	69	1393
Pakistan	72	4099	93	171	243	448	427	5553
Sri Lanka	7	610	10	30	45	135	20	856
Ships								27
South Asia	2021	37696	306	2340	2442	6849	3289	54970

diesel fuels. The approach taken is to multiply the number of vehicles in each category, the average number of km driven in a year, and the average emission factor in g km^{-1}. Vehicle fleet data for the year 2000 are obtained by extrapolation from 1990 to 1996 trends [*International Road Federation* (*IRF*), 2000]. At the time of this analysis, 1996 was the latest year for which vehicle population data are available for India. Mileage data are taken from the same source. The most difficult parameter to characterize appropriately is the average emission factor for each vehicle type in India. After examination of vehicle emission restrictions under Indian regulations, the accessible literature, and simulations performed with EPA's MOBILE 5 model (which incorporate vehicle vintaging and deterioration rates), we settled on the following emission factors for the major vehicle types in South Asia, which are meant to be averages over the entire vehicle fleet appropriate for the year 1999 or 2000: gasoline cars, 19.8 g km^{-1}; diesel cars, 17.4 g km^{-1}; heavy-duty gasoline trucks, 61.2 g km^{-1}; heavy-duty diesel trucks, 17.4 g km^{-1}; and motorcycles, 21.0 g km^{-1}. These values are significantly higher than new vehicles and higher than average vehicle fleets in more-developed countries; they are intended to reflect higher proportions of older vehicles, greater deterioration due to poor engine maintenance, and—though not explicitly—a certain fraction of fuel adulteration. The calculation yields emission estimates for India in 2000 of 6.2 Tg CO from motorcycles, 2.1 from passenger cars, and 2.1 from trucks, buses, and tractors, for a total of 10.4 Tg CO yr^{-1}, in reasonable agreement with that derived for internal combustion engines in the previous section.

[18] Field combustion is also a significant source of CO. Our emission estimates for South Asia are derived from previous work [*Crutzen and Andreae*, 1990; *Streets and Waldhoff*, 2000]. Corn, wheat, and rice residues are considered, using FAO crop production data [*Food and Agriculture Organization* (*FAO*), 2000, available at http://www.fao.org] and US EPA emission factors [*Environmental Protection Agency* (*EPA*), 1996]. We assume crop-to-residue ratios of 1.2 for corn, 1.3 for wheat, and 1.0 for rice [*Lu*, 1993] and that 23% of residues are burned in the field [*Crutzen and Andreae*, 1990]. We estimate that field combustion contributed 2.5 Tg to India's CO emissions, and 3.3 Tg in all of South Asia. Despite the extensive literature on biomass burning, the magnitude of CO emissions from biomass combustion in South Asia remains poorly understood [*Galanter et al.*, 2000].

[19] Table 3 summarizes the anthropogenic CO emission estimates for South Asia by country and source type. We estimate by this method that India emitted 43.9 Tg CO in 2000. Combustion of biofuels accounted for 29.0 Tg (66%) and mobile sources accounted for 10.4 Tg CO (24%), within the uncertainty of the 13.7 Tg derived in the previous section (see "total petroleum," Table 2). The emissions of CO from international shipping in the Indian Ocean are estimated at 27 Gg, based on the activity level from *Streets et al.* [2000] and the emission factor for commercial heavy fuel-oil engines of 0.6 kg m^{-3} [*EPA*, 1996]. The estimate of direct CO emissions for all of South Asia is 55.0 Tg; indirect emissions are discussed below.

2.3. Particulate Organic Carbon Emissions

[20] *Chan and Weaver* [1994] and *Weaver and Chan* [1996] estimated aerosol emissions from two-stroke engines using an indirect exhaust opacity method, or from samples in plastic bags (Table 1). *Patschull and Roth* [1995] measured particles directly with a drift mobility analyzer. If Patschull and Roth's measurement of particles from two-stroke engines is representative of spark ignited engines in South Asia, then consumption of 9 Tg of fuel would, produce about 0.8 Tg yr^{-1} of particulate organic matter (POM), equivalent 0.7 Tg OC yr^{-1} with a mean fuel formula of CH_2. These measurements were made on a new German engine running under optimal conditions; in-use engines operating in developing countries may eject more OC. Emission of 0.7 Tg yr^{-1} represents a substantial fraction of the total OC thought to arise from Indian emissions [*Liousse et al.*, 1996; *Reddy and Venkataraman*, 2000]. If a substantive component of the aerosol from two-stroke engines is soot, then they could be a major source of BC; the ratio of BC/OC in particles from two-stroke engines needs to be measured.

2.4. Particulate Black Carbon: Estimates From Emission Factors

[21] The characterization of Asian particulate sources is challenging, and all estimates of BC in this part of the world need to be viewed with some skepticism. The variety of combustors and fuels is extensive, and not much is known about the distribution and performance of each type. The following analysis for South Asia is based on a detailed assessment of BC emissions in China [*Streets et al.*, 2001]. In the China analysis, 37 different source categories were examined, including all aspects of the combustion of bio-

Table 4. BC Emissions (Gg) for South Asia, 2000

Region	Industry	Domestic	Transport	Power Generation	Field Combustion	Total
Reference Case						
Bangladesh	0.1	37.9	0.8	0.0	2.1	40.9
Bhutan	0.0	1.2	0.0	0.0	0.0	1.3
India	18.3	425.1	35.0	1.0	28.4	507.7
Nepal	0.1	16.5	0.3	0.0	0.8	17.6
Pakistan	1.7	64.1	6.0	1.7	5.6	79.1
Sri Lanka	0.1	6.5	0.7	0.0	0.1	7.4
Ships						15.5
South Asia	**20.2**	**551.2**	**42.8**	**2.7**	**37.1**	**669.4**
High Case						
Bangladesh	0.5	37.9	2.7	0.0	2.1	43.2
Bhutan	0.0	1.2	0.1	0.0	0.0	1.3
India	247.9	425.1	113.6	1.0	28.4	816.0
Nepal	0.5	16.5	1.0	0.0	0.8	18.7
Pakistan	13.6	64.1	20.0	1.7	5.6	105.0
Sri Lanka	0.1	6.5	2.3	0.0	0.1	9.0
Ships						15.5
South Asia	**262.6**	**551.2**	**139.6**	**2.7**	**37.1**	**1008.6**

fuels, coal, oil, and agricultural waste. The application and performance of particulate control devices was taken into account. The emission factors for each source category were selected to represent only that fraction of total particulate releases less than 1 μm in diameter and composed of elemental carbon. Because of the high uncertainty in BC emission estimates, we have performed a ''reference case'' analysis for South Asia—using best estimates of emission factors—and a ''high case,'' in which we estimate an upper bound on BC emissions by assuming ''worst-case'' values for two of the key parameters.

[22] In the reference case, we assume that the array of BC source types is similar to that in China, the only differences being the levels of activity in each source category. Again, we use the RAINS-Asia model to derive year-2000 energy use. The dominant source of BC in India, according to this inventory, is the combustion of biofuels in residential stoves. Our selection of emission factor for this source category is 1.0 g kg^{-1}. This is predicated on the measurements of *Muhlbaier* [1982], but follows a review of 14 experimental data sets on residential fuel combustion [see *Streets et al.*, 2001]. It is 3–4 times higher than the values chosen by *Reddy and Venkataraman* [2000], but similar to that used by Reddy and Venkataraman (M. S. Reddy and C. Venkataraman, A 0.25 × 0.25 inventory of aerosol and sulfur dioxide emissions from India, II, Biomass combustion, submitted to *Atmospheric Environment*, 2001b). Our estimate for BC emissions in 2000 from biofuel combustion in India is 420 Gg, and 540 Gg for the whole of South Asia; the balance in Table 4 is due to the residential combustion of coal and kerosene. Though residential coal combustion is important in China, it is only a very minor contributor to BC emissions in India.

[23] The combustion of coal in industry is a source of considerable uncertainty in India. We have found very little information on the types of combustors used and the extent of particulate controls. In the reference case, we assume that the distribution of source types in India is the same as in China, i.e., 97% (by weight of coal, not number) of industrial coal combustors employ some form of particulate control, but mostly rather inefficient cyclones and wet particle scrubbers; we also assume that 85% of the combustors are stoker-fired. This picture could be inaccurate, though, and we discuss an alternative possibility below. Our emission factor for uncontrolled industrial coal combustion is 0.32 g kg^{-1} with much smaller values for controlled sources. Using these assumptions, BC emissions from industry are quite low (see Table 4). Emissions from power generation are low, due to high-temperature combustion and the widespread employment of electrostatic precipitators for particle control. In this respect, we deviate from the assumptions of *Reddy and Venkataraman* [2000]. We assume an emission rate for power plants of 0.0001 g kg^{-1} (pulverized coal plants with ESP) and 0.36 g kg^{-1} (uncontrolled fuel oil plants) [*Streets et al.*, 2001]. *Reddy and Venkataraman* [2000] assumed 1.0 g kg^{-1} for coal-fired power plants, but for these sources ash dominates the particulate emissions and the carbon fraction is small.

[24] Assessing BC emissions from vehicles is difficult, as discussed above. In the reference case, we again assume that Indian vehicles, on average, have similar performance to Chinese vehicles; both sets of vehicles are inefficient, have high average particulate emissions, and have a significant proportion of ''superemitters.'' To allow for poorer engine maintenance and fuel adulteration in India, we have chosen the high end of our range of fleet-averaged emission factors: 0.21 g kg^{-1} for gasoline vehicles and 3.52 g kg^{-1} for diesel vehicles [*Gillies and Gertler*, 2000; *Streets et al.*, 2001]. Using these emission factors, we obtain an estimate of 35 Gg for BC emissions in India from vehicles and 43 Gg for all of South Asia, under the reference case.

[25] Overall, our estimate for BC emissions in India is 0.51 Tg for 2000 in the reference case, of which by far the dominant share (84%) originates in the residential sector. This is close to the estimate of *Reddy and Venkataraman* [2000], 0.45 Tg ± 0.26 Tg, despite differences at the individual source-category level. For the whole of South Asia, we estimate 0.67 Tg; shipping in the Indian Ocean contributes only 15 Gg. These values are consistent with our estimate for China of 0.91 Tg [*Streets et al.*, 2001], con-

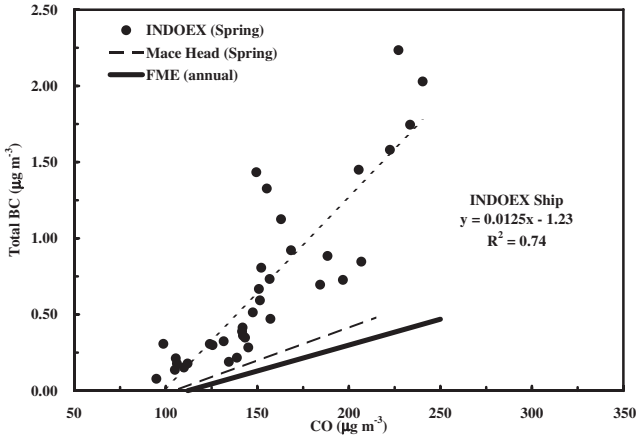

Figure 1. Scatterplot of CO and BC measured from the R/V Ronald Brown over the Indian Ocean during INDOEX, February–April 1999. The short dashed line represents a least squares best fit to the data. The slope for INDOEX data (0.0125) is about three times higher than the annual average slope observed at a North American site (solid line), Fort Meade (FME), Maryland [*Chen et al.*, 2001], and (long dashed line) a European site, Mace Head, Ireland [*Jennings et al.*, 1996].

sidering the greater usage of coal in small combustors in China. Our BC estimates tend to be different from the previous literature [see *Penner et al.*, 1993; *Cooke and Wilson*, 1996; *Cooke et al.*, 1999], because the emission factors they use may include some contribution from larger (>1 μm) particles and ash, they may omit biofuels, and their inventories are for earlier time periods. For example, *Cooke et al.* [1999] assumed a BC emission factor for particles smaller than 1 μm in diameter, from coal-fired power plants in developed countries of 0.149 g kg^{-1} which we consider too high. We used 0.1 mg kg^{-1} [*Streets et al.*, 2001], reflecting the preponderance of ash in the PM from power plants.

[26] In the high case, we assume that more industrial coal is burned without any kind of particulate control (10%) and that the emission factor for uncontrolled coal combustion is at the high end of our range, 2.21 g kg^{-1}. This raises BC emissions from the industrial sector from 18 Gg to 250 Gg (Table 4). Similarly, we assume that vehicles are even more polluting than assumed in the reference case. We take the highest estimates from *Streets et al.* [2001] that take into account a high proportion of "superemitters" as well as a high average emission rate, thus: gasoline vehicles 0.23 g kg^{-1}, diesel vehicles 12 g kg^{-1}, and motorcycles 0.59 g kg^{-1}. These are probably upper limits for fleet averages and yield vehicle emissions in India of 35 Gg to 114 Gg.

[27] Our overall estimate for BC emissions in the high case is 0.82 Tg in India and 1.01 Tg in all of South Asia. We conclude that a bottom-up approach to estimating BC emissions in the region yields estimates in the range of 0.5–0.8 Tg for India and 0.7–1.0 Tg for South Asia.

2.5. Particulate Black Carbon: Estimates From Ambient Measurements

[28] *Chen et al.* [2001] measured CO and BC at a nonurban site in Fort Meade, MD (39°N, 77°W, between Washington, DC and Baltimore, MD) and found these two pollutants highly correlated with an annual average slope of 3.4×10^{-3} g BC/g CO. They used the ratio of ambient concentrations of BC and CO and known emissions of CO to estimate BC emissions, and found a value in reasonable agreement with recent compilations for North America [*Cooke et al.*, 1999]. During the cruise of the R/V Ronald H. Brown in INDOEX 1999, carbon monoxide and aerosols were monitored simultaneously for nearly one month [*Neusuess et al.*, 2002; *Stehr et al.*, 2002]. A strong correlation of BC and CO ($r^2 = 0.74$) was observed in air from South Asia (Figure 1). The intercept at positive CO results from the Northern Hemisphere, low latitude background CO mixing ratio in spring of 90 to 135 ppb (100 to 160 μg m^{-3}) [e.g., *Rhoads et al.*, 1997]. The positive correlation may be due to common sources, but even if it is due only to mixing of emissions from proximate sources, CO can be used as an internal standard to estimate the total production of BC from India. Considering all the data collected in the Northern Indian Ocean, Bay of Bengal, and Arabian Sea, we find a slope of 12.5×10^{-3} g BC/g CO.

[29] The C-130 aircraft operated by the National Center for Atmospheric Research, conducted 18 flights mostly over the Arabian Sea and Northern Indian Ocean in February and March 1999 [*Mayol-Bracero et al.*, 2002]. These measurements also show a strong positive correlation between CO and BC ($r^2 = 0.71$), but with higher concentrations and a higher slope, 27×10^{-3} g BC/g CO (Figure 2), than was observed from the ship.

[30] Data collected in February 1999 at KCO (Figure 3) also show a positive correlation between CO and BC [*Chowdhury et al.*, 2001; *Lobert and Harris*, 2002]. Aerosol particles were collected over 2-d intervals with a filter sampler and with an impactor, both of which select for particles with an aerodynamic diameter <1.8 μm. The best fit slope is 15.0×10^{-3} g BC/g CO for fine aerosol and

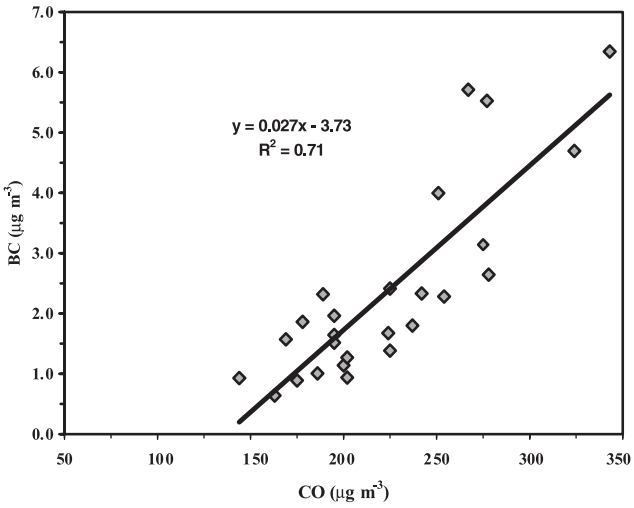

Figure 2. Scatterplot of CO and BC measured during February and March 1999 from the C-130 aircraft over the Indian Ocean. The average concentration and slope are more than twice as high as observed on the ship and may be a result of faster transport and slower removal of aerosol particles in the free troposphere than in the MBL.

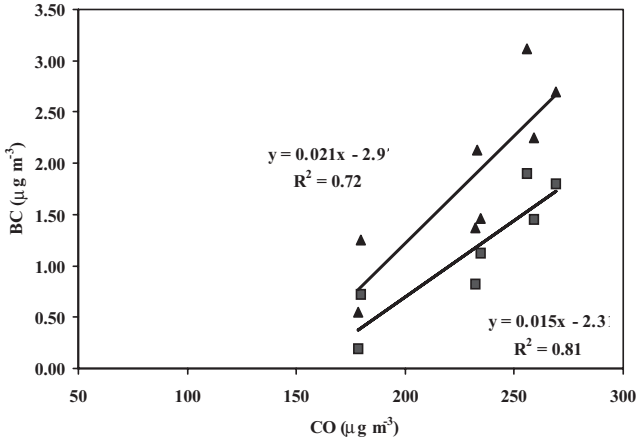

Figure 3. Scatterplot of CO and BC measured from 11 to 26 February 1999 at KCO, the surface site on the island of Kaashidhoo in the Maldives (4.97°N, 73.47°E). Triangles represent aerosol samples were collected over 2 d with a filter that selects for fine particles (aerodynamic diameter <1.8 μm). Squares represent the sum of stages of a MOUDI impactor, also collected over 2 d. The slope and mean concentration of these data collected at KCO are somewhat higher than for the ship data; this may be the result of the island being closer to the sources than the average position of the ship.

20.9×10^{-3} g BC/g CO for the sum of the impactor stages <1.8 μm diameter. The slope and mean concentration of the data collected at KCO are somewhat higher than for the ship data. The island, located about 700 km SW of the Indian coast, is closer to the sources than the average position of the ship, which cruised a broad expanse of the Indian Ocean.

[31] To estimate BC emissions we scale these BC/CO ratios by the estimated total CO emissions for India. The total emission must include CO produced by biogenic hydrocarbon oxidation; assuming a 30% yield and the isoprene emissions from *Guenther et al.* [1995] gives an additional 11 Tg CO. The grand total for India is 66.7 Tg(CO) yr^{-1} (Table 2). For India, we estimate 0.83 Tg(BC) yr^{-1} from the shipboard observations, 1.0 and 1.4 Tg(BC) yr^{-1} from the island data, and 1.8 Tg(BC) yr^{-1} from the aircraft observations. Carbon monoxide is sparingly soluble, and has an atmospheric lifetime of about 1 mo. BC can be attached to soluble particles such as sulfate, and has a shorter lifetime, especially in the MBL where it can be removed by wet or dry deposition to the Earth's surface. Back trajectories indicate that the air sampled from the ship had been over the ocean from 2 to 10 d (R. R. Dickerson, et al., Overview: Cruise of the research vessel Ronald H. Brown during the Indian Ocean Experiment (INDOEX) 1999, submitted to *Journal of Geophysical Research*, 2001), while the air sampled from the aircraft had been in more recent contact with the continent, because wind speeds tend to be higher at higher altitudes. In addition, the free troposphere is decoupled from the MBL where BC can be more rapidly lost to deposition. The difference between the BC/CO ratio measured on the ship and that measured on aircraft implies a loss of about 60% of the BC, a reasonable value for air that has been over the ocean for some days.

[32] Assuming for ship observations, 50 to 70% loss of BC and no loss of CO we estimate fine particulate emissions of 1.7 to 2.8 Tg(BC) yr^{-1} for India. Assuming for aircraft observations 0 to 33% loss of BC (a reasonable upper limit for losses in the short transport time) for we estimate 1.8 to 2.7 Tg(BC) yr^{-1} for India. Using population and fuel use (see also Table 4) we estimate that total combustion emissions from South Asia are about 33% greater than from India alone. Scaling our estimate for India by this factor and adding in oxidation of biogenic hydrocarbons we find a total of about 89 Tg(CO) yr^{-1} for 1999 in South Asia. Using results from all three platforms, our estimate for the whole of South Asia is 1.0 as a lower limit and 2 to 3 Tg(BC) yr^{-1} as the probable range.

3. Discussion

[33] Our estimate of CO emissions is lower than prior values, due to a reduced estimate of agricultural, grassland, and forest burning (11.2 Tg, see Table 2) [*Andreae and Merlet*, 2002]. *Galanter et. al.* [2000] estimated 72 Tg(CO) yr^{-1} compared to our value of 40.2 for all biomass burning in India (Table 2). The EDGAR analysis projected a total of 110 Tg(CO) yr^{-1} (105 from biomass burning) for the whole of South Asia in 1990. The value for total CO production derived in this work, 89 Tg(CO) yr^{-1}, is below these estimates, but probably within the combined uncertainty.

[34] Emissions from two-stroke engines and biomass burning [e.g., *Crutzen and Andreae*, 1990] contain VOC to NO_x ratios higher than emissions from four-stroke or diesel vehicles. For example the VOC/NO_x ratio (Table 1) from two-stroke engines is about 220 ppm C/ppm NO_x. This puts air in Indian cities in the range of severely NO_x-limited ozone production [e.g., *Finlayson-Pitts and Pitts*, 2000]. Existing measurements of ambient air pollution in India are inadequate to characterize fully the composition, but published observations indicate high VOC to NO_x ratios. *Padhy and Varshney* [2000] measured an average of about 15 ppm nonmethane volatile organic compounds in Delhi in spring; *Lal et al.* [2000] measured CO to NO_x molar ratios in the range of 40:1, indicating much greater VOC concentrations and VOC to NO_x ratios than are observed over North America [e.g., *Stehr et al.*, 2002]. The low concentrations of ozone observed over and downwind from India may then be due to the high VOC/NO_x ratio of the initial emissions. As South Asia develops technologically and converts from two-stroke motorcycles to four-stroke automobiles the ratio of VOC to NO_x will decrease and may result in higher ozone levels unless emissions controls are enforced.

[35] *Cooke and Wilson* [1996] derived 0.49 Tg(BC) yr^{-1} for North America and 0.85 Tg(BC) yr^{-1} for the "rest of Asia", all of Asia outside China. Scaling by population, this gives about 0.6 Tg(BC) yr^{-1} for South Asia. With such emissions, their model calculated about 0.1 μg(BC) m^{-3}, about a tenth of what was observed during INDOEX. *Cooke and Wilson* [1996] considered only fossil fuel combustion in 1985, but model emissions still appear to be underestimated.

[36] The sum of uncertainties in each factor of the bottom-up estimate of CO emissions is large, but chemical transport models offer an alternative test of emission inventories. *de Laat* [2000] and *de Laat et al.* [2000] simulated CO over the Indian Ocean during INDOEX and found

reasonable agreement using the EDGAR values for 1990. When there were differences between modeled and measured concentrations, the model was usually high; increasing the CO model emissions in South Asia by 30% led to unreasonably high concentrations. Simulations with the NASA/GSFC model [*Park et al.*, 2001] gave similar results (R. J. Park, personal communication, 2000). These simulations suggest that the CO emission inventories are probably within 30–50% of the actual values.

[37] Measurements of BC during INDOEX are uncertain by as much as 30% [*Neususess et al.*, 2002]. The BC data from Fort Meade, MD were analyzed with a protocol different from that used during INDOEX. The differences in BC concentrations for the two techniques depends on the origin of the samples, but the protocol followed for North American samples tends to produce higher BC values [*Chow et al.*, 2001]. Although the combined uncertainties in the estimated emissions based on ambient measurements are sizable, we feel that there is greater than 95% confidence that this estimate exceeds that from the bottom-up method. Because uncertainties go both up and down, the contribution of emissions from South Asia to the global BC budget could be higher (by roughly a factor of two) than the 2–3 Tg BC yr^{-1} derived here.

[38] The slope of the BC/CO plot for South Asia (Figures 1–3) is three to eight times that observed over North America [*Chen et al.*, 2001] or Europe [*Jennings et al.*, 1996; *Cooke et al.*, 1997], but these three regions each release about the same amount of CO. The *EPA* [2000] estimates about 100 Tg(CO) yr^{-1} from North America (available at http://www.epa.gov/ttn/chief/trends/index.html); *Olivier et al.* [1994] estimate 85 Tg(CO) yr^{-1} for Europe and 110 Tg(CO) yr^{-1} for South Asia; in this work we estimate 89 Tg(CO) yr^{-1} for South Asia. *Chen et al.* [2001] found that BC/CO ratios increased with ambient temperature, and the hot climate of South Asia may contribute to the high concentrations of BC found there. If estimates from the ambient measurements method are correct then South Asia is a major source of atmospheric BC.

4. Conclusions

[39] Air pollution observed downwind of South Asia differs fundamentally from that observed downwind of North America and Europe. Organic carbon, black carbon, and light absorption are all greater; the ozone concentration is lower. The sources differ - over India two-stroke engines, biomass burning, and small-scale coal combustion are major contributors. For example, two-stroke engines emit an order of magnitude more CO per unit fuel than do four-stroke engines with pollution control; the emissions derived here reflect our best estimate of the pollution production characteristic of South Asia. Using emission rates determined for North America or Europe would have led to serious errors.

[40] Our estimate of CO emissions shows a higher contribution from fossil fuel combustion, due to the high HC and CO emission factors for two-stroke engines. The total CO released is lower than previous calculations because of reduced estimates of agricultural and other biomass burning. South Asia, however, remains an important and growing contributor to the global CO budget, with an increasing fraction from fossil fuel consumption. Motorcycles alone emit 9 Tg CO, 3.6 Tg HC, and 0.7 Tg OC per year in India.

[41] Biomass burning and two-stroke engines emit high levels of VOC but low levels of NO_x in comparison to four-stroke engines and large-scale coal-fired power plants. This high VOC to NO_x ratio appears to inhibit formation of photochemical smog (ozone) in Indian cities. The limited in situ measurements available support this contention. As vehicles with four-stroke engines become a larger segment of the fleet, substantial reductions in CO, VOC and OC could be realized, even without emissions controls, but unless NO_x controls are instituted, photochemical smog could worsen.

[42] The current rate of emission of BC can be estimated from the rate of CO emission in India and the ratio of BC to CO observed downwind. This analysis yields 2 to 3 Tg(BC) yr^{-1}, suggesting that South Asia may already be a major contributor to global BC production. Bottom-up estimates of BC emissions from India yield much smaller values than do in situ observations. This suggests an unaccounted-for analytical error, an unaccounted-for source, or unusually high emission factors. If the rate of BC emission implied by ambient measurements is correct, than it is essential to identify the source or process leading to these high concentrations in order to develop an abatement strategy. Direct measurements of emissions sources in South Asia are urgently needed to resolve this issue.

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