Direct radiative forcing from anthropogenic carbonaceous aerosols over India

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A chemical–meteorological box model has been formulated to estimate direct radiative forcing from carbonaceous aerosol constituents. Anthropogenic black carbon and organic matter aerosol emissions from industrial fuels and biomass burning in India were estimated at 0.44 and 2.47 Tg y^{-1}, respectively for 1990. Global black carbon and organic matter estimates were 12.30 and 81.00 Tg y^{-1}, respectively, from the same source inventory for the base year 1984. Meteorological parameters for 1990 from 18 India Meteorological Department (IMD) stations are used to estimate spatial average lifetimes and burdens of carbonaceous aerosols. The transport mechanisms considered are advection by surface winds, dry deposition and wet deposition. Estimated lifetimes of black carbon aerosols are lower in May to October (3.2 to 5.1 day) with effective wet deposition, and higher in November to April (6.2 to 12.9 day), with an annual mean lifetime of 6.5 ± 4.7 day. Monthly mean burdens of black carbon aerosols range from 0.59 to 2.37 mg m^{-2}, and follow the same as lifetimes with an annual mean burden of 1.20 ± 0.87 mg m^{-2}. Monthly mean direct radiative forcing from black carbon aerosols is positive (net warming) and is higher (+ 0.51 to + 0.72 W m^{-2}) in December and January than May to October (+ 0.03 to + 0.16 W m^{-2}), with an annual mean of + 0.27 ± 0.17 W m^{-2}. Organic matter aerosol lifetimes, on the other hand are lower in May to October (2.0 to 3.7 day) and higher in November to April (4.6 to 11.9 day), with an annual average of 5.1 ± 4.3 day. Their monthly mean burdens range from 2.07 to 12.10 mg m^{-2} with an annual mean of 5.20 ± 4.36 mg m^{-2}. Estimated monthly mean direct radiative forcing from organic matter is high (– 1.49 to – 2.11 W m^{-2}) in December and January and is low (– 0.13 to 0.43 W m^{-2}) in May to October, with an annual mean of – 0.69 ± 0.55 W m^{-2}. Organic matter aerosols cause a decrease in radiation flux of comparable magnitude to that previously estimated from sulphate aerosols (– 1.1 ± 1.0 W m^{-2}), while black carbon aerosols cause a one-quarter increase in the radiation flux. Further estimates of these effects from fly ash and mineral aerosols are needed.

EMISSIONS of anthropogenic aerosols to the troposphere have increased in recent decades[1], and the consequent increase in their concentrations may influence the earth's climate. Aerosol effects include scattering and absorption of incident solar radiation (direct effect) and modifying the microphysical and optical properties of clouds (indirect effect). Much of the recent work on the direct radiative forcing of aerosols (i.e. the change in net irradiance at the top of the atmosphere from the presence of aerosols) has focused on sulphate aerosols[2–4]. However, carbonaceous aerosols of anthropogenic origin may exert a significant direct radiative forcing[5–6]. Originating from fossil fuel combustion and biomass burning, carbonaceous aerosols contain black carbon directly emitted during the combustion process (primary aerosols) and organic matter formed in the atmosphere by condensation of organic vapours (secondary aerosols). A chemical–meteorological box model is formulated in this study to estimate monthly mean burdens of black carbon and organic matter, and the resulting optical depths and direct radiative forcing over the Indian subcontinent. These are compared with global estimates for these species and with previously estimated direct radiative forcing from sulphate aerosols[7] over the Indian subcontinent.

Model description

A first estimate of carbonaceous aerosol contribution to direct radiative forcing is made using regional emissions and atmospheric parameters for India. A carbonaceous aerosols emission inventory is compiled from aggregate fossil fuel (industrial coal and petroleum) and biomass use data along with appropriate emission factors. A box model is used to estimate the carbonaceous aerosol burdens considering aerosol transport and removal mechanisms such as advection, wet deposition and dry deposition. Meteorological data originated from 18 stations spread uniformly over India. Monthly mean meteorological, deposition and optical parameters were averaged by the Fourier amplitude sensitivity test (FAST) over their ranges, to obtain spatially averaged aerosol lifetimes and burdens, optical depths and radiative forcing over India.

Emissions

For 1990, a carbonaceous aerosol emission inventory for fossil fuel combustion and biomass burning was compiled

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using appropriate emission factors for black carbon and organic matter. The industrial coal and petroleum use data were from Mitra[8] and biomass burning data from Kaul[9], Kaul and Shah[10], Sinha and Joshi[11] with other black carbon and organic matter emission factors for fossil fuel and biomass burning from reported literature[12–23] (Table 1). While carbonaceous aerosol emissions from biomass burning may vary seasonally, as a first estimate, we assume that emissions from both fossil fuel and biomass burning are uniformly distributed throughout the year. Total black carbon emissions are 0.44 Tg y^{-1} for India for 1990, with 0.24 Tg y^{-1} from fuel combustion and 0.20 Tg y^{-1} from biomass burning. Estimated organic matter emissions are 2.47 Tg y^{-1} with fossil fuel combustion and biomass burning contributing 1.07 and 1.40 Tg y^{-1}, respectively (Table 1). In comparison, estimates of global black carbon emissions from fossil fuel combustion and biomass burning are 12.30 and 13.96 Tg y^{-1}, with Asia contributing 2.22 Tg y^{-1} for the base year 1984 (refs 24, 25). Estimated global organic matter emissions are 81.00 Tg y^{-1} from the above sources also for 1984 (ref. 25). Both these global emissions increased with the increase in energy use between 1984 and 1990.

Box model for aerosol lifetimes and burdens

These emissions are used as the source term in a box model calculation for estimation of burdens and radiative forcing. The formulated box model covers an area ($L \times W$) of 7.3×10^{12} m^2 (8–36°N and 70–92°E), including the landmass of India and surrounding oceans, with an average mixed layer height of 0.5–2 km and with uniformly mixed constituents[2]. This follows from previous work to estimate formation and direct radiative forcing from sulphate aerosols over the Indian subcontinent[7]. With previously mentioned transport and removal mechanisms, the lifetime (L_t) of the aerosols in the box is given as:

$$L_t = \frac{1}{K} = \frac{1}{K_a} + \frac{1}{K_w} + \frac{1}{K_d},$$

where K_a is the advection removal rate (s^{-1}); K_w is the wet deposition rate (s^{-1}), and K_d is the dry deposition rate (s^{-1}). K_a, K_w and K_d are given as:

Table 1. Fuel use data and emission estimates of carbonaceous aerosols for 1990

Fuel type	Fuel use (Mt)	Emission factors (g kg^{-1})		Emissions (Tg y^{-1})	
		Black carbon	Organic matter	Black carbon	Organic matter
Fossil fuel					
Coal	214.2*	1.000[@]	4.333[$]	0.214	0.928
Lignite	11.0	1.000	4.333	0.011	0.048
Coke	3.5	1.000	4.333	0.004	0.015
LPG	2.3	0.060	0.260	5.0×10^{-4}	0.001
Kerosene	8.2	0.300	1.300	0.002	0.011
HS diesel	20.7	0.700	3.033	0.014	0.063
Fuel oils	8.8	0.100	0.433	0.001	0.004
Motor gas	3.5	6.0×10^{-5}	2.6×10^{-4}	2.1×10^{-7}	9.1×10^{-7}
Aviation gas	1.8	6.0×10^{-5}	2.6×10^{-4}	1.1×10^{-7}	4.7×10^{-7}
Sub total				0.247	1.069
Biomass					
Wood	252.1[Δ]	0.280[&]	1.302	0.070	0.328
Cattle dung	106.1	0.325	3.634	0.034	0.386
Agri. residue	99.2	0.446	2.935	0.044	0.291
Forest	26.7	1.530[#]	12.402	0.041	0.331
Grassland	5.2	1.530	12.402	0.008	0.064
Sub total				0.198	1.401
Total				0.445	2.470

*Fossil fuel use data[8].
[Δ]Biomass use data[9–11].
[@]Black carbon emission factors for fossil fuels[12,13].
[$]Organic matter emission factors for fossil fuels: black carbon/organic carbon = 0.3 (refs 14–16); organic matter/organic carbon = 1.3 (ref. 17).
[&]Biomass fuel emission factors: black carbon/particulate mass, and organic carbon/particulate mass[18,19]; particulate mass emission factors[20].
[#]Forest and grassland burning: black carbon/particulate mass, organic carbon/particulate mass and particulate mass emission factors[21–23].

$$K_a = \frac{1}{\left(\frac{V\cos\theta}{L} + \frac{V\sin\theta}{W}\right)},$$

where V is the surface wind velocity (m s^{-1}); L is the length of the box (m); and W is the width of the box (m).

$$K_w = \frac{RS\,\rho_{water}}{H\,\rho_{air}},$$

where R is the monthly mean rainfall (m s^{-1}); S is the scavenging ratio; ρ_{water} is the density of water (g cc^{-1}), and ρ_{air} is the density of air (g cc^{-1}).

$$K_d = V_d / H,$$

where V_d is the dry deposition velocity of aerosols (m s^{-1}) and H is mixing height (m).

Advection of aerosols out of the box is assumed to occur by monthly mean horizontal surface winds with wind speed data obtained from the 18 IMD stations. An average dry deposition velocity of 0.1 cm s^{-1} used for black carbon and organic matter is consistent with earlier studies[6,24–27]. Wet deposition rates and corresponding wet deposition fluxes are determined using IMD rainfall data and appropriate scavenging ratios. The reported scavenging ratios for black carbon and organic matter are 18 to 350 and 35 to 760, respectively[28] (Table 2).

Carbonaceous aerosol burden (B), which is an area concentration integrated from the surface of the earth to the top of the atmosphere, is given by[2,3].

$$B = \frac{QL_t}{A},$$

where Q is the aerosol emission rate (g day^{-1}); L_t is the aerosol atmospheric lifetime (day); and A is area of the box (m^2).

Direct radiative forcing

Direct radiative forcing is estimated assuming an optically thin layer of aerosol where multiple reflections within the aerosol layer are neglected[2,3]. The change in radiative forcing in a column of air integrated from the surface of the earth to the top of the atmosphere, as a result of the change in planetary albedo occurring in the cloud-free area of the atmosphere, is given by[2,3]:

$$\Delta F = -\tfrac{1}{4} F_T (1 - A_c)\Delta R_a,$$

where ΔF is the change in forcing (W m^{-2}); $(1/4)F_T$ is incident top of the atmosphere radiative flux; A_c is the fractional areal cloud cover; and ΔR_a is the change in planetary albedo (Table 3). This assumes that planetary albedo is enhanced only in cloud-free regions. The assumption of an optically thin aerosol allows ΔR_a to be related to the aerosol transmissivity and reflectivity as[22]:

$$\Delta R_a = r + \frac{t^2 R_s}{1 - R_s r} - R_s,$$

where R_s is the albedo of the surface; r is the reflectivity [$r = (1-e^{-\tau})\,\omega\beta$]; and t is the transmissivity [$t = e^{-\tau} + (1-\beta)(1-e^{-\tau})\,\omega$] of the aerosol layer. Here, β is the globally averaged fraction of the upscattered radiation (Table 3), τ is the optical depth and ω is the aerosol single scattering albedo (ratio of the mass scattering coefficient to the mass extinction coefficient).

Optical depth is a product of the aerosol burden, and the mass extinction coefficient relating to the scattering or absorption efficiency of the aerosols[3] is given as:

$$\tau = \alpha B,$$

The mass extinction efficiency of black carbon aerosols is independent of relative humidity, whereas that of organic matter increases with relative humidity because of its hygroscopic nature. The mass scattering efficiency of organic matter aerosols (relative humidity < 50%) is 3 to 5 $m^2\ g^{-1}$, and increases by a factor of 1.7 with water accretion at a higher relative humidity of about 80% (refs 29, 30). In this study, we use the suggested empirical

Table 2. Carbonaceous aerosols deposition properties

Parameter	Range of values
Dry deposition velocity[27], V_d (cm s^{-1})	0.1
Scavenging ratio[28], S	
Black carbon	18–350
Organic matter	35–760

Table 3. Constants used in the direct radiative forcing calculations[2,3]

Symbol	Parameter	Value
F_t	Top of atmosphere radiative flux	1370 W m^{-2}
T_a	Fraction of light transmitted by above aerosol layer	0.76
R_s	Fractional albedo of underlying surface	0.15
β	Backscatter fraction	0.29

enhancement factor of 1.7 for increase in relative humidity from 35% to 85% to calculate the organic matter mass scattering efficiency[25].

Spatial averaging

FAST used for a global sensitivity analysis of non-linear models[31,32] is used to obtain the spatial average direct radiative forcing from ranges of input parameters. FAST associates each uncertain parameter with a specific frequency in the Fourier transform space of the system. The system sensitivities are then determined by solving the system equations for discrete values of the Fourier transform variable and computing the Fourier coefficient associated with each parameter. Computer codes are available for applying FAST on user defined subroutines[32]. The parameters are normally distributed about the mean with an additive search curve for small range or uncertainty and an exponential search curve for a range exceeding an order of magnitude.

Results and discussion

Monthly mean meteorological parameters

Monthly mean meteorological parameters, including surface wind speed, rainfall, relative humidity and areal cloud cover, were obtained for 1990 from IMD, Pune, from 18 meteorological stations (Figure 1). These parameters are measured according to WMO guidelines, and all parameters (except rainfall) were measured twice daily at 03:00 GMT (08:30 IST) and 12:00 GMT (17:30 IST). Wind speed is measured by a manual or an automatic anemometer and wind direction using a standard wind vane. Rainfall is measured continuously using a standard recording rain gauge (SRRG). Relative humidity (RM) is derived from the dry and wet bulb temperatures of a hygrometric chart. Cloud cover is estimated visually, for 8 sections of the sky, as the extent of low, medium and high clouds in each section in 0.1 Oktas, with 1.0 Okta representing one-eighth of the sky. Monthly means and standard deviations of each parameter were spatially averaged over India. These parameters for 1990 (Figure 2) show surface wind speeds between 1.3 and 2.7 m s^{-1} and RH between 50 and 80%. There are large monthly variations in areal cloud cover (23–84%) and rainfall (2–279 mm) with an anomalously high rainfall of 187 mm in May.

Carbonaceous aerosol lifetimes and burdens

The lifetimes of carbonaceous aerosols are dependent on advection, wet and dry deposition. A dry deposition velocity of 0.1 cm s^{-1} is used for black carbon and organic matter, and would result in a removal rate of 5×10^{-7} s^{-1} and a corresponding lifetime of about 23.1 days (mixed layer height of 0.52 to 2 km) in the absence of advection and wet deposition. The wet deposition fluxes are estimated from the IMD rainfall and corresponding scavenging ratios for black carbon and organic matter[28] (Table 2).

The lifetimes of black carbon are low in May to October (3.2 to 5.1 days) with efficient wet deposition, and high in November to April (6.2 to 12.9 days) (Figure 3 *a*). The calculated highest lifetime is 12.9 days in January (due to the lowest rainfall of 2 mm). These lifetimes follow the same trend as sulphate lifetimes calculated in a previous study with wet deposition being the predominant removal mechanism[7]. Estimated annual average lifetime of 6.5 ± 4.8 day is in reasonable agreement with a globally averaged black carbon lifetime of 7.9 days[33]. The integrated column burdens of black carbon (Figure 3 *b*) are also low in May to October (0.59 to 0.93 mg m^{-2}) when high rainfall leads to efficient wet deposition; they are then higher in November to April (1.13 to 2.37 mg m^{-2}) when rainfall is low. The present estimate for annual mean burden of black carbon is 1.20 ± 0.87 mg m^{-2}. Previous GCM estimates of black carbon burdens were 0.50 to 2.00 mg m^{-2} for India and 0.88 mg m^{-2} for the northern hemisphere using 1984 emissions[6].

Calculated lifetimes of the organic matter are low in May to October (2.0 to 3.7 days) and higher in November to April (4.6 to 11.9 days) with an annual average of 5.1 ± 4.3 days (Figure 3 *c*). Lifetimes of the organic matter are lower than black carbon aerosols, because of higher scavenging ratios and consequent higher wet

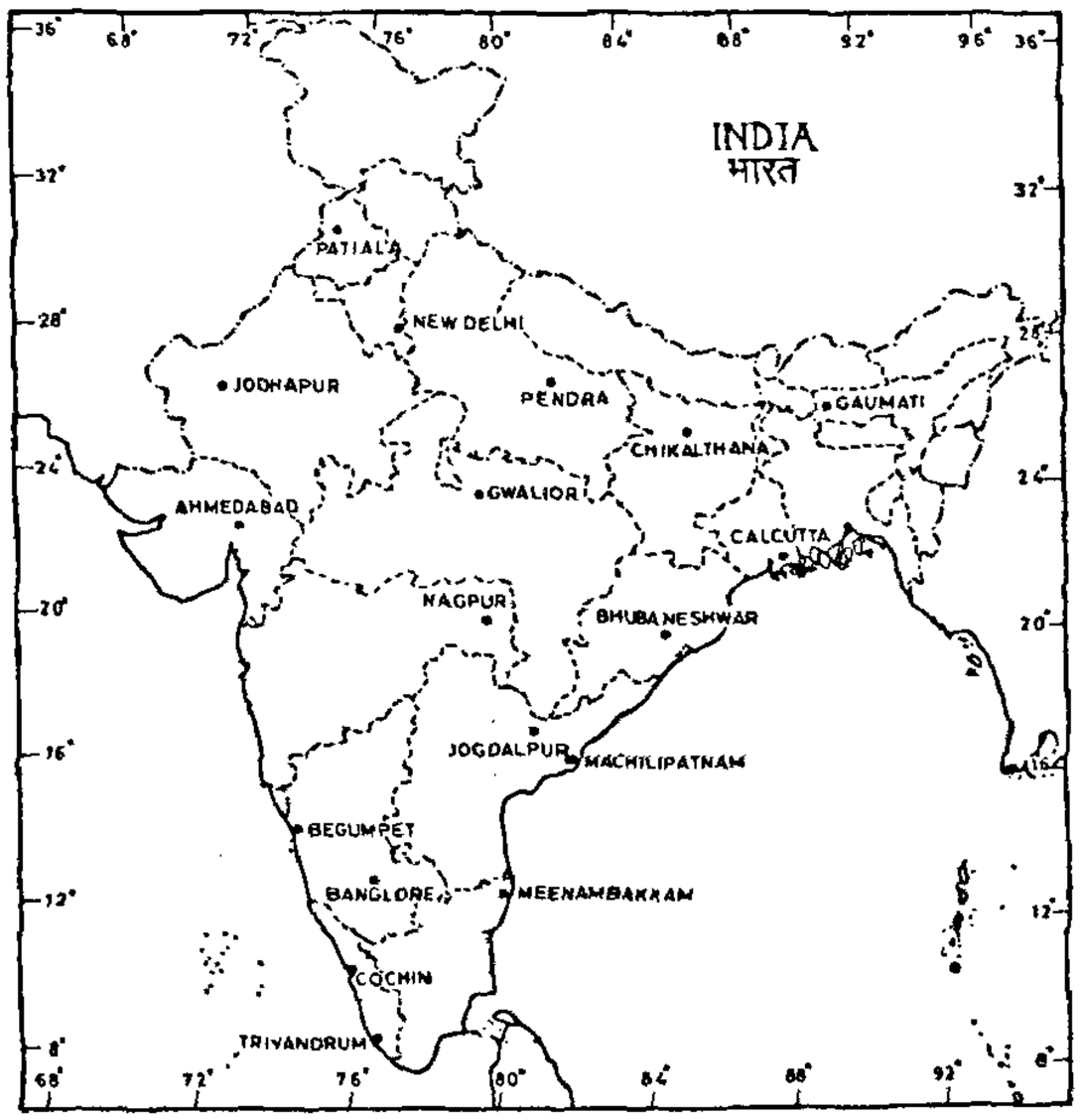

Figure 1. IMD stations chosen for meteorological data compilation for 1990.

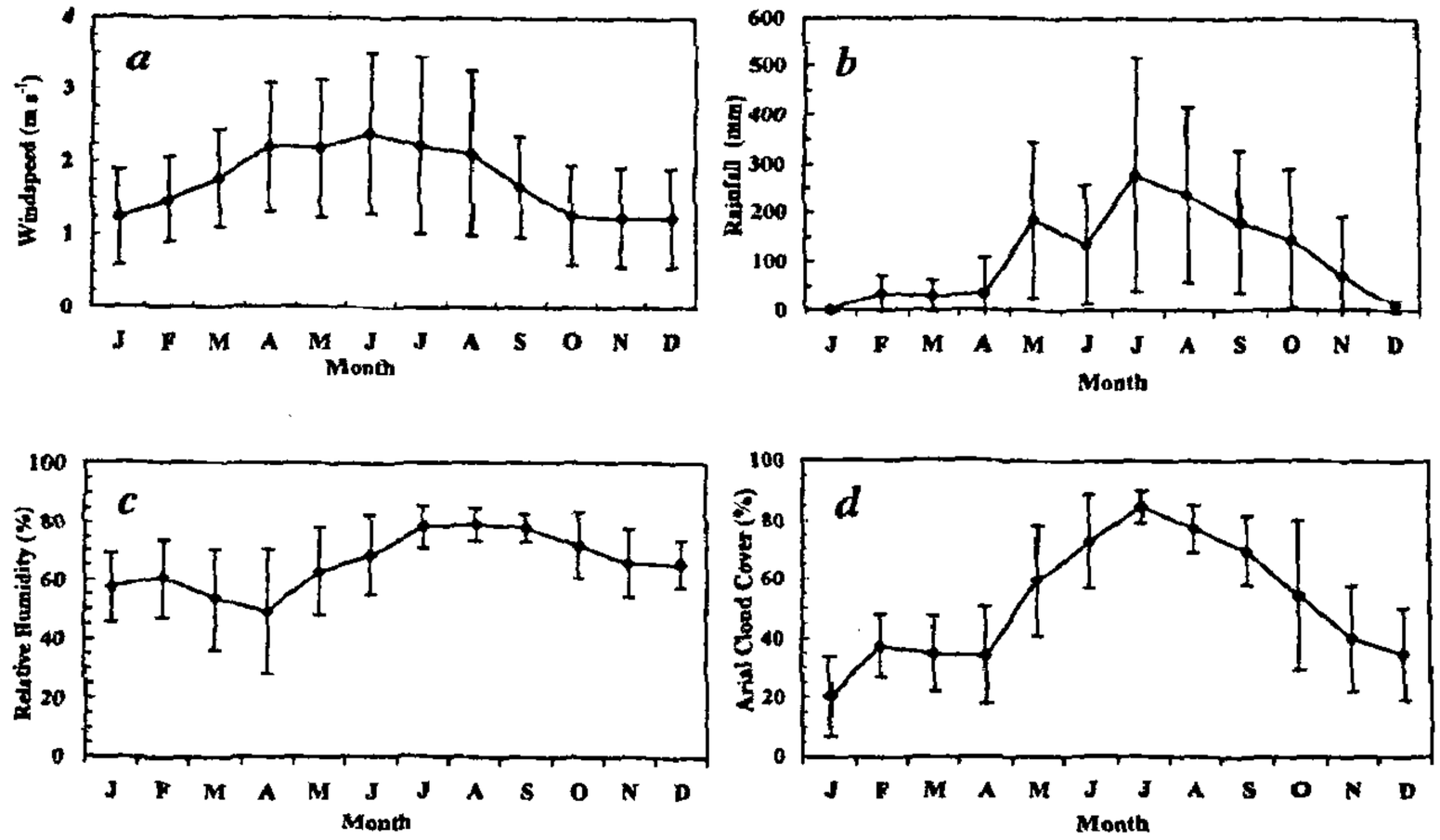

Figure 2. Monthly mean meteorological parameters for 1990: *a*, Surface wind speeds; *b*, Rainfall; *c*, Relative humidity; and *d*, Areal cloud cover for 1990.

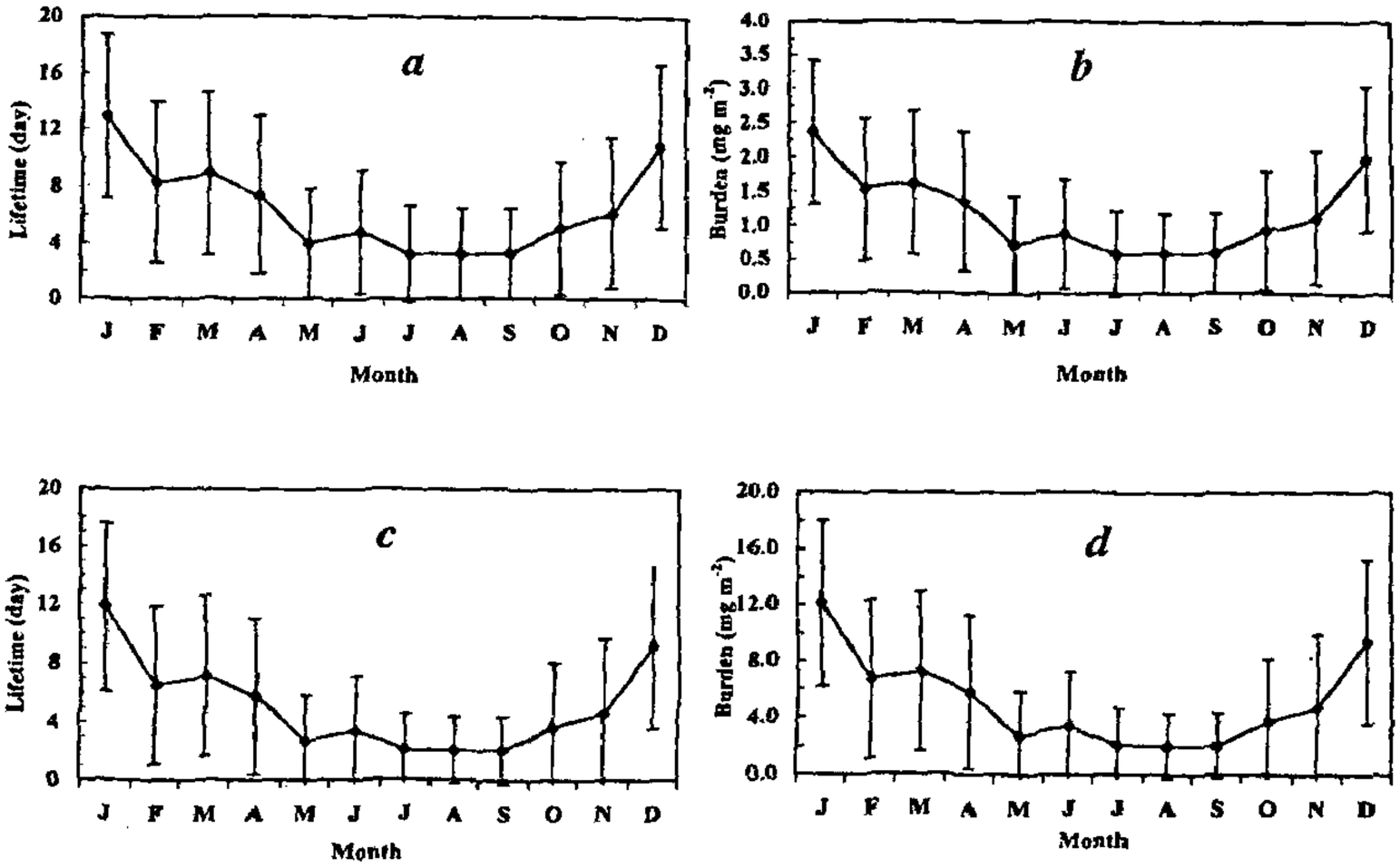

Figure 3. Estimated monthly mean lifetimes and burdens of carbonaceous aerosols over India for 1990. *a*, Black carbon lifetimes; *b*, Black carbon burdens; *c*, Organic matter lifetimes; and *d*, Organic matter burdens.

deposition of organic carbon matter. Monthly mean burdens of organic matter follow the same trend as lifetimes, with highs of 4.74 to 12.10 mg m^{-2} in November to April and lows of 2.07 to 3.75 mg m^{-2} in May to October with an annual average of 5.20 ± 4.36 mg m^{-2} (Figure 3 *d*). No previous estimates of organic matter lifetimes and burdens are available for comparison.

Optical depths and direct radiative forcing

Optical depths are calculated as the product of the burden and the mass extinction efficiency (Table 4).

Table 4. Carbonaceous aerosols optica

Symbol	Parameter
α_s (m^2 g^{-1})	Mass scattering coefficient
	Black carbon[34,35]
	Organic matter[22,36]
α_a (m^2 g^{-1})	Mass absorption coefficient
	Black carbon[35,36]

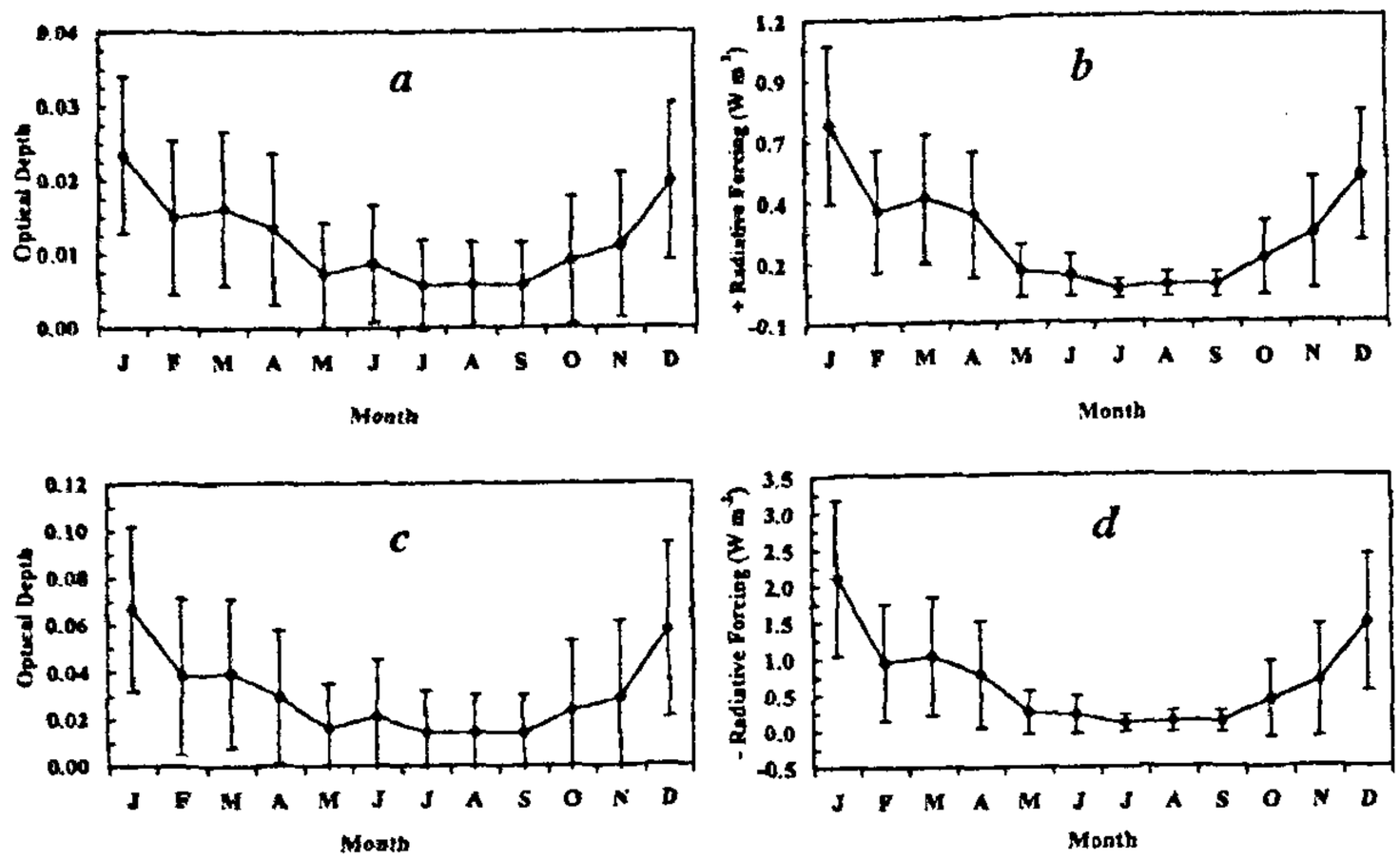

Figure 4. Estimated monthly mean optical depths and direct radiative forcing from carbonaceous aerosols over India for 1990. *a*, Black carbon optical depths; *b*, Direct radiative forcing from black carbon; *c*, Organic matter optical depths; and *d*, Direct radiative forcing from organic matter.

Black carbon optical depths (Figure 4 *a*) tend to follow the lifetimes and burdens with a seasonal variation of lows during May to October (0.006 to 0.009) and highs from November to April (0.011 to 0.023) with an annual mean of 0.112 ± 0.009. The organic matter optical depths (Figure 4 *c*) also follow the lifetimes and burdens with a seasonal variation of lows during May to November (0.014 to 0.024) and highs from November to April (0.029 to 0.067) with an annual average of 0.030 ± 0.027. Larger optical depths result from the higher burdens of organic matter than black carbon. From an earlier estimate, sulphate optical depths over the Indian subcontinent are 0.010 to 0.020 during May to October and 0.030 to 0.060 during November to April with an annual average of 0.030 ± 0.030. In comparison, the annual average of total optical depth from carbonaceous (black carbon and organic matter) aerosols calculated here is 0.042 ± 0.035.

Monthly mean direct radiative forcing from black carbon aerosols varies from $+0.03$ to $+0.72$ W m^{-2} with an annual average of $+0.26 \pm 0.17$ W m^{-2} (Figure 4 *b*). This is a positive radiative forcing because of the absorptive characteristics of black carbon and would result in net warming. Its magnitude is about one-quarter of the previously estimated sulphate negative forcing[7]. The radiative forcing during May to October is lower ($+0.03$ to $+0.11$ W m^{-2}) than in November to April ($+0.26$ to $+0.72$ W m^{-2}). Previous estimates from GCM studies using 1984 emissions range from 0 to $+2.00$ W m^{-2} over India[6]. Monthly mean direct radiative forcing from organic matter is high (-1.49 to -2.11 W m^{-2}) in December and January, is moderate (-0.78 to -1.03 W m^{-2}) during February to April and low (-0.10 to -0.43 W m^{-2}) during May to October (Figure 4 *d*). Here also, radiative forcing follows the seasonal trends of organic matter lifetimes and burdens with an annual average forcing of -0.69 ± 0.55 W m^{-2}. No previously reported studies of radiative forcing from organic matter over India exist for comparison. This radiative forcing from the organic matter is comparable to the previously estimated sulphate forcing (-1.1 ± 1.0 W m^{-2}) (ref. 7) and is also negative, resulting in potential net cooling.

Conclusions

With the chemical–meteorological single box model, direct radiative forcing from black carbon and organic matter aerosols can be compared. Anthropogenic black carbon and organic matter aerosol emissions from India were 0.44 and 2.47 Tg y^{-1}, respectively for 1990. The lifetimes of carbonaceous aerosols are governed by wet deposition with organic matter having larger scavenging ratios and consequently smaller lifetimes (5.1 ± 4.3 days) than black carbon aerosols (6.5 ± 4.7 days). Monthly mean black carbon burdens ranged from 0.59 to 2.37 mg m^{-2}, with a seasonal variation of winter–spring highs and summer lows controlled by wet deposition. Optical depths of black carbon vary from 0.006 to 0.023 following the seasonal trend of burdens. Monthly mean direct radiative forcing from black carbon aerosols vary from $+0.05$ to $+0.72$ W m^{-2} with an annual average of $+0.27 \pm 0.17$ W m^{-2}. This radiative forcing is positive (net warming), and one-quarter magnitude of the previously estimated sulphate negative forcing of -1.1 ± 1.0 W m^{-2}. Organic matter monthly mean burdens ranged from 2.07 to 12.10 mg m^{-2} with an annual mean of 5.20 ± 4.36 mg m^{-2}. The organic matter optical depths are of the order of 0.013 to 0.070, following the same seasonal trend as burdens. Monthly

mean direct radiative forcing is high (– 0.69 to – 2.11 W m^{-2}) in November to April and is low (– 0.13 to – 0.43 W m^{-2}) in May to October, with an annual mean of – 0.69 ± 0.55 W m^{-2}. Forcing from organic matter is comparable with that from sulphate aerosols and is negative resulting in potential net cooling. The present estimates indicate that carbonaceous aerosol constituents could cause radiative forcing over India similar in magnitude to that from sulphate, with black carbon contribution being positive (net warming) and the organic matter contribution being negative (net cooling). It is also of interest to compare the relative radiative forcing from other aerosol types including fly ash and mineral aerosols.

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