Measurements of carbonaceous aerosols at urban and remote marine sites

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Concentrations of total carbon (TC) and black carbon (BC) in ambient air at Delhi (urban site) and over Indian Ocean (remote marine) were determined as a part of INDOEX programme. Over Indian Ocean, the TC and BC concentrations varied from 1.81 to 10.05 μg/m^3 and 0.13 to 1.36 μg/m^3 respectively during FFP-98. During the same season at Delhi, the TC and BC ranged from 7.50 to 40.27 μg/m^3 and 0.49 to 2.84 μg/m^3 respectively. In addition, at Delhi, the TC and BC concentrations were noticed very low during the monsoon season. However, the percentage BC during monsoon season was very high compared to winter season. High concentrations of TC were observed due to high organic carbon (OC) which might be due to biomass burning of various kinds. Similar to Delhi, near Indian coast, the concentration of OC was very high while towards ITCZ and across ITCZ, OC content was relatively lower.

RECENT studies on atmospheric aerosol composition show that at urban sites, about 50% mass of fine particle is due to carbonaceous aerosols[1]. The presence of carbonaceous aerosols has also been reported in very remote regions away from any source[2,3] suggesting that they can be transported to a long distance even to remote oceans. Carbonaceous aerosols are produced during combustion processes like vehicular exhaust, power plant emissions, domestic heating, biomass burning, etc. The dark component of combustion-derived aerosols often termed as carbon soot, has remarkable physical and chemical properties. These particles scatter and absorb light contributing to the reduction of visibility[4,5]. They affect heterogeneous reactions in the atmosphere or within cloud and rain drops[6]. These particles can also serve as scavenger for other atmospheric pollutants, especially highly carcinogenic hydrocarbons[7].

Carbonaceous aerosols consist of highly polymerized organic matter with low hydrogen and oxygen content. They can be classified into two categories, viz. black carbon (BC) and organic carbon (OC). BC has refractive behaviour to thermal and chemical attacks[8]. The main source of BC is combustion and thus it can be used as a tracer for industrial pollution. OC particles have a refractive index closer to that of sulphate particles, which means that they mainly scatter solar radiation and take part in global cooling effect. Observations show that both at remote and source sites the OC concentrations are higher than BC[9].

Considering the importance of carbonaceous aerosols both in urban and remote regions, the present study was carried out as a part of INDOEX where samples of aerosols were collected at Delhi (highly source influenced site) and over Indian Ocean (remote marine) during INDOEX-FFP 98 (February–March, 1998). During this period, the NE monsoon airmasses carry pollutants from Indian subcontinent to the ocean. In this study, the concentrations of TC and BC have been determined in ambient air and they have been compared with the data available worldwide. The present study will be useful to estimate the budget of carbonaceous aerosols in Indian subcontinent and their transport to the ocean that may further be useful to understand the role of Indian emissions of carbonaceous particles in global cooling.

Experimental details

Sampling

The samples of carbonaceous aerosols were collected at Delhi, which is a highly polluted city and over Indian Ocean during INDOEX-FFP 98 (February–March 1998) At Delhi, the samples were collected on the terrace of National Physical Laboratory (NPL) at a height of around 13 m from the ground level. These samples were collected on Whatman EPM 2000 glass fibre filters using a high volume sampler (HVS) at an average flow rate of 1 m^3 min^{-1} on 8–24 h basis. In addition, at Delhi, the samples of carbon soot were also collected in September 98. Over the ocean, the samples were collected on *Sagar Kanya* (SK cruise #133). These samples were collected on Whatman GF/F glass fibre filters with low volume pump at a flow rate of 50 LPM on 12–48 h basis around 3 m above the deck surface of the ship and around 15 m above the sea surface. Collected samples were stored in cool and dry place.

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Analysis

The samples were analysed at Laboratories des Sciences du Climate et de I" Environment, Giff (France). Before analysis, the filters were treated with HCl fumes for decarbonation[10]. It removes all the carbonate present on filter and remaining carbon is composed of organic carbon and soot carbon[11]. After decarbonation, the samples were analysed by two-step thermal method. During the first step, so-called pre-combustion step, the samples were heated at around 300°C to estimate organic carbon. The second step was combustion of the filters in a furnace of a carbon analyser at 1100°C. At this point, CO_2 is evolved which was detected coulometrically. The further details of the method are given in Cachier *et al.*[11].

Results and discussion

Carbonaceous aerosols at Delhi

Table 1 gives the concentrations of total carbon (TC) and black carbon (BC) during winter (January, February) and monsoon (September) seasons at Delhi. TC includes both BC and OC (organic carbon). During winter season, northeasterly monsoon prevails while during September, southwesterly monsoon comes to the end in this region. During winter season at Delhi, TC concentrations are 23.76 ± 3.17 μg/m^3 while BC concentrations 1.71 ± 9.95 μg/m^3. The total percentage contribution of BC to TC is 10% while during September, although TC concentrations are 4.93 ± 1.18 μg/m^3 but the BC percentage contribution is 50% which is a significant contribution to TC. During winter, the very high TC may be due to increased concentrations of OC which is contributed by increased heating sources such as fuel wood burning, charcoal burning and other biomass burning. In addition, meteorological conditions (lower inversion, calm conditions and lower temperature) also stagnate pollutants contributing to high TC.

As shown in Table 2, the use of fossil fuels (lignite, peat) and burning of charcoal, fuel wood, dung, etc. contribute a large amount of OC compared to BC. The very high concentrations of OC during winter months (northeasterly monsoon) are also corroborated by the INDOEX FFP-98 observations over Indian Ocean, which have been discussed in the next section. During September, no extra heating is required for mankind and the main sources of OC and BC are diesel, petrol, coal, etc., giving rise the lower concentrations of TC in monsoon.

Table 1. Total carbon and black carbon concentrations (μg/m^3) at Delhi

	TC	BC	%BC
January–February (N = 8)	23.76 ± 13.17	1.71 ± 0.95	9.57 ± 7.54
September (N = 7)	4.93 ± 1.18	2.34 ± 0.52	49.58 ± 15.07

Table 2.

Fuel source	BC emission factor in g C (aerosol)/ kg C (fuel)	OC emission factor in g C (aerosol)/ kg C (fuel)
Fossil fuels		
Natural gas	0.0003	0.0012
Heavy fuel	0.02	0.08
Motor gasoline	0.1	0.4
Kerosene	0.3	1.2
Jet fuel	1	1
Diesel	5	5
LPG	0.06	0.25
Coal	3	3
Lignite	4	15
Peat	1	6
Coke	3	3
Biomass burning		
Charcoal production	3.5	45
Charcoal consumption	2	7
Dung consumption	2	25
Fuelwood	2.5	11
Tree burning	1.5	13
Grass burning	0.8	6

Source: Cachier, 1998.

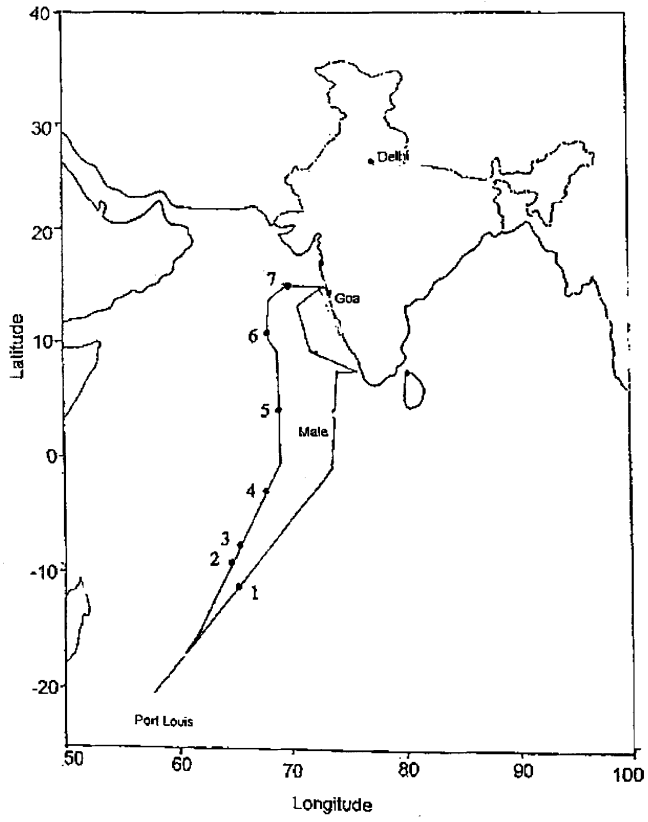

Figure 1. Map showing the location of Delhi and cruise track of INDOEX (FFP-98) (cruise #133) along with sample number and location of sampling point.

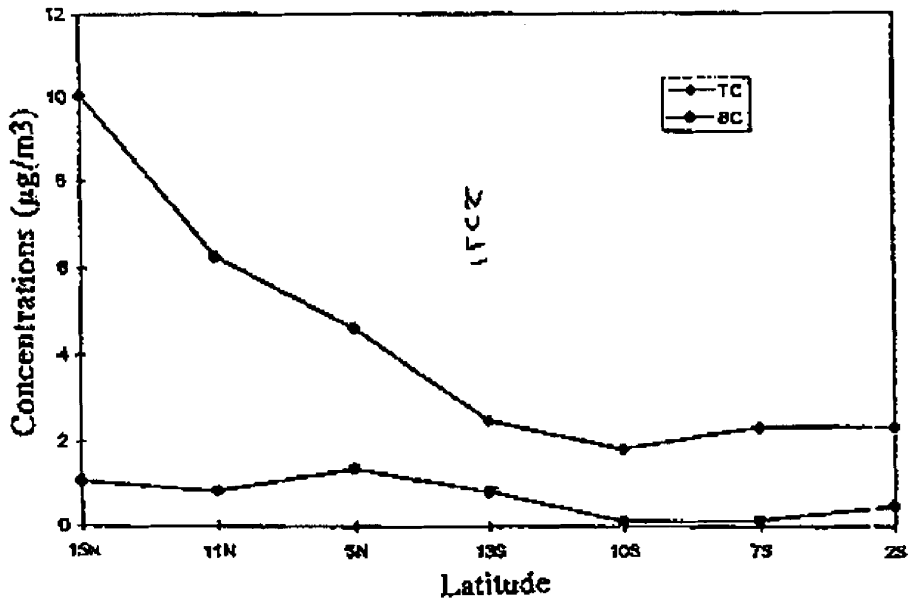

Figure 2. Latitudinal variation in total carbon (TC) and black carbon (BC) during INDOEX (FFP-98) (cruise #133).

Carbonaceous aerosols over Indian Ocean

Figure 1 shows the cruise track of INDOEX FFP-98 along with the location of Delhi. In March, a total of seven samples were collected on the locations as shown in Figure 1. Sample 1 represents the Inter Tropical Convergence Zone (ITCZ) concentration of carbon aerosols (ITCZ was located 10–13°S during onward leg). Samples 2–4 were collected in the southern hemisphere across the ITCZ during return leg (ITCZ was located near 2°S). Samples 5–7 were collected near Indian peninsula.

Figure 2 shows the variation of TC and BC over Indian Ocean during March 98 in order of latitude with the location of ITCZ. It shows that before ITCZ, both TC and BC are higher than in the ITCZ samples and across ITCZ region. Figure 2 also indicates that near Indian peninsula, the ratios of TC and BC are very high, suggesting a very significant contribution of OC coming from the Indian subcontinent. Across ITCZ, there is no much variation in TC and BC ratios, which suggests that in the southern hemisphere the OC and BC are contributed by similar sources. At Amsterdam Island (77°E, 38°S) in Indian Ocean, biomass burning influence has been observed by Cachier *et al.*, during dry season (June to September 1996).

Correlation coefficient between TC and BC over Indian Ocean was 0.64 while at Delhi, it was observed to be 0.05 in January–February and 0.33 in September respectively. The weak correlation at Delhi may be due to contribution from different sources and varying methods of combustion.

Table 3. Comparison of concentrations of carbonaceous aerosols at different sites of the world

	TC ($\mu g/m^3$)	BC ($\mu g/m^3$)	% BC/ TC	References
Urban				
Gif winter	19.0	5.4	28	9
Gif spring	20.2	4.6	23	9
Gif summer	13.3	2.5	19	9
Lisbon	–	15.9	–	2
Augsburg	–	2.37	–	2
Delhi	14.34	2.03	14	Present study
Rural				
Landes forest	1.8	0.3	16	9
Amazon basin	4.38	0.62	14	2
Coastal				
Corsica	1.9	0.3	16	9
Remote continental				
Southwestern US	–	0.22	–	2
Ocean near continent				
North sea	1.55	0.68	44	2
Peru	0.58	0.099	17	2
Remote ocean				
Atlantic	0.16	0.04	25	2
Pacific	0.14	0.014	10	2
Indian ocean	2.16	0.2	9	Present study

TC, Total carbon; BC, black carbon.

Global scenario

Table 3 represents TC and BC concentrations at different sites all around the world. TC and BC at Delhi are compa-

rable with global data reported for other urban sites. Winter values of Delhi are comparable with spring values of Giff. TC and BC observed in this study over Indian Ocean are one order of magnitude higher than Atlantic and Pacific oceans. But the percentage contribution of BC to TC is comparable. The higher TC might be due to the influence of biomass burning in African region and Indian subcontinent, which contributes significant OC aerosols.

Conclusion

To understand the characteristics of air pollution at an urban site (Delhi) and the extent of its transport to remote marine region (Indian Ocean), field studies were carried out. During winter season at Delhi, TC concentrations are 23.76 ± 13.17 μg/m^3 while BC concentrations 1.71 ± 9.95 μg/m^3. The total percentage contribution of BC to TC is 10% while during September, although TC concentrations are 4.93 ± 1.18 μg/m^3 but the BC percentage contribution is 50% which is a significant contribution to the TC. During winter, the very high TC may be due to increased concentrations of OC, which is contributed by increased heating sources such as fuel wood burning, charcoal burning and other biomass burning. In addition, meteorological conditions also stagnate pollutants contributing to high TC. Similar to Delhi, the concentration of OC was very high near Indian coast while towards ITCZ and across ITCZ, OC content was observed relatively lower.

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