Large latitudinal gradients and temporal heterogeneity in aerosol black carbon and its mass mixing ratio over southern and northern oceans observed during a trans-continental cruise experiment

K. Krishna Moorthy,[1] S. K. Satheesh,[2] S. Suresh Babu,[1] and Auromeet Saha[1]

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[1] Extensive, and collocated measurements of the mass concentrations (M_B) of aerosol black carbon (BC) and (M_T) of composite aerosols were made over the Arabian Sea, tropical Indian Ocean and the Southern Ocean during a trans-continental cruise experiment. Our investigations show that M_B remains extremely low (<50 ng m^{-3}) and remarkably steady (in space and time) in the Southern Ocean (20°S to 56°S). In contrast, large latitudinal gradients exist north of ~20°S; M_B increasing exponentially to reach as high as 2000 ng m^{-3} in the Arabian Sea (~8°N). Interestingly, the share of BC showed a distinctly different latitudinal variation, with a peak close to the equator and decreasing on either side. Large fluctuations were seen in M_T over Southern Ocean associated with enhanced production of sea-salt aerosols in response to sea-surface wind speed. These spatio-temporal changes in M_B and its mixing ratio have important implications to regional and global climate. **Citation:** Moorthy, K. K., S. K. Satheesh, S. S. Babu, and A. Saha (2005), Large latitudinal gradients and temporal heterogeneity in aerosol black carbon and its mass mixing ratio over southern and northern oceans observed during a trans-continental cruise experiment, *Geophys. Res. Lett.*, *32*, L14818, doi:10.1029/2005GL023267.

1. Introduction

[2] Black carbon is generally the anthropogenic aerosol component contributing most to the aerosol absorption of solar radiation [*Penner et al.*, 1993] and is considered as a tracer for anthropogenic impact to climate forcing. The absorption by BC could offset the "white house effect" of aerosols such as sulphate, the light-scattering properties of which counteract the green-house warming [e.g., *Haywood and Shine*, 1997; *Jacobson*, 2001; *Babu et al.*, 2002, 2004]. BC also affects the cloud albedo by altering the hygroscopic properties of cloud condensation nuclei and the solar heating caused by BC can reduce cloudiness [*Ackerman et al.*, 2000]. Not only the absolute concentration, but BC share to the composite aerosols also has strong influence on aerosol forcing [*Babu et al.*, 2004]. Being generally in the submicron size range and chemically inert, BC has longer residence time in the atmosphere and is amenable for long distance transport, even to remote oceanic environments; as far down as the Antarctica [*Hansen et al.*, 1988; *Bodhaine*, 1995]. Thus, investigations of BC over the oceans assume importance. Despite, detailed measurements of BC are sparse or almost non-existent over the vast Oceanic regions down Asian continent (which is one of the most densely populated region) and in the pristine Southern Indian Ocean (Southern Ocean). In this paper, we present the results of campaign-mode, collocated measurements of composite aerosols and BC, made over the Arabian Sea (AS), tropical Indian Ocean (IO) and the Southern Ocean (SO) as far as 56°S, in the longitude region between 45°E and 72°E. This is probably for the first time in history, aerosol and BC measurements were made across the hemispheres with a latitudinal coverage of ~70°. The results are examined for spatio-temporal characteristics and latitudinal gradients.

2. Experimental Details and Measurements

[3] The measurements were made during the period 2 January to 1 April of 2004, onboard the oceanographic research vessel (ORV) Sagar Kanya during its first pilot expedition to the SO. The cruise had three phases; Initial phase (SK199; from 2 Jan to 20 Jan 2004); Main phase (SK 200; from 23 Jan to 4 Mar 2004), and Final phase (SK 201; 10 Mar to 2 Apr 2004). The main phase (SK 200) was confined to the SO (20°S to 56°S) while the initial and final phases had north-south transects between 20°S and ~12°N, but separated by 40 to 60 days. The cruise tracks are shown in Figure 1. Except for two brief port-calls at Mauritius (MRU, 20°S, 60°E) in between the phases, the ship was mostly in motion.

[4] Near-real time measurements of the mass concentration (M_B) of BC were made continuously onboard using an Aethalometer (model AE-21 of Magee Scientific, USA). The ambient air was aspirated from the front of the ship (sampling into the incoming air) following the protocols described by *Babu et al.* [2004]. The flow rate was kept at 5 lpm (litres per minute) during the first phase (when BC concentrations were high) and increased to the maximum value permissible for the instrument (6.8 lpm) in the SO. In the return leg (phase 3) the flow rate was kept ~6 lpm. The time base also was increased from 5 min in the north, to up to 60 min for the SO in view of the extremely low concentration encountered there. Following the error budget described in several earlier papers [*Babu et al.*, 2002, 2004] the maximum uncertainly in the measured BC was ~15% in the SO, and was ~10% to 8% in the tropical IO and AS, respectively.

[5] Mass concentrations of the composite aerosols were estimated using a single stage high volume sampler (HVS, model GHV2000P1 of Thermo-Andersen). The sampler was operated from the front deck at a flow rate of 0.6 m^3

[1]Space Physics Laboratory, Vikram Sarabhai Space Centre, Trivandrum, India.
[2]Centre for Atmospheric and Oceanic Sciences, Indian Institute of Science, Bangalore, India.

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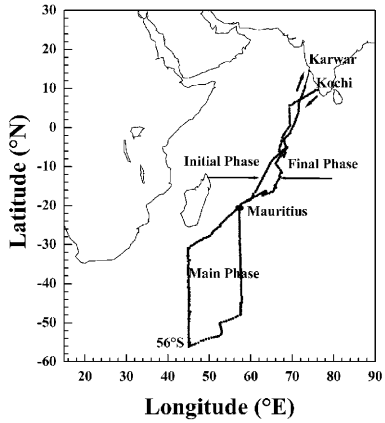

Figure 1. Cruise track of the first pilot expedition to the Southern Ocean of the ORV Sagar Kanya.

min^{-1} for durations ranging from 3 hr to 10 hr. Aerosols were collected on pre-desiccated, tare-weighed, numbered, and sealed quartz fiber substrates. After sampling, the filters were sealed in their respective envelopes and taken to the laboratory where they were desiccated and weighed using the same microbalance. From the difference in the initial and final weights, the mass concentration (M_T) was estimated knowing the flow rate and sampling time. Flow rate of the HVS was calibrated before the campaign and its stability was ensured by repeating the calibration after the campaign. In all 30 samples were collected during the three phases of the expedition of which 14 were in SO.

3. Results and Discussion

[6] The measurements showed M_B to vary over a wide range across the oceanic region surveyed; from as high as $\sim$2000 ng m^{-3} in the AS ($\sim$8 to 12°N, 75 to 77°E) to as low as 20 to 50 ng m^{-3} in the SO. In some locations over the SO, M_B was below the detectable limit ($\sim$10 ng m^{-3}) of the instrument, even after averaging for $\sim$1 hr. The latitudinal variation of M_B is shown in Figure 2, where the points are temporal averages and the vertical bars through them the standard deviations of the mean. In the initial and final phases, when the measurements were made in the tropical oceans, the averaging was done over a 12 hr period, because there were enough samples (short timebase) and the ship covered three to four degrees (latitude) in a day, whereas in the main phase only daily mean values were estimated in view of the longer timebase, low BC values, and the very small spatial variation. The filled circles correspond to measurements made in the first phase (Kochi (10°N, 76.5°E) to MRU) and the cross marks to those made in final phase from MRU to Karwar (12°N, 72°E), almost along the same track (Figure 1) but about 40 to 60 days later, thus indicating the temporal changes as northern winter changes to spring. The star marks correspond to measurements over the SO. Compared to the northern and tropical oceanic regions, BC in SO is very low and shows no perceptible spatio-temporal variations despite a vaster oceanic region being covered in the main phase. This would thus represent the background levels of BC, in remote locations. In contrast, over the tropical oceans, BC shows large variation both spatially and temporally. Examining these in the light of the prevailing mean synoptic winds at 850 hpa for the cruise period, (NCEP reanalysis monthly mean data), it appears that the high M_B values in AS and tropical IO are predominantly due to advection from continents (S and SW Asia). The decreasing source impact leads to the steady decrease in M_B as the ship moved off the west coast of India. This is quite similar to the spatial gradients in AOD/mass concentration reported over this region during the INDOEX of 1998 and 1999 [*Moorthy et al.*, 1999, 2001; *Parameswaran et al.*, 1999]. The temporal changes (from SK199 to SK201) would be associated with the change in the season (over the landmass).

[7] The points in Figure 2 during the initial and final phases indicate an exponential enhancement in M_B as we approach the northern landmass from the south. This latitudinal variation has been parameterised using an equation of the form

$$Y = A \exp(\Lambda/\Lambda_D) \quad (1)$$

for the region $12° > \Lambda > -18°$; where A is the amplitude (concentration in ng m^{-3}), Y is the value of M_B at latitude Λ (deg) and Λ_D is the scaling length expressed in degrees (of latitude). The broken lines in Figure 2 show the fit to equation (1) through the points. A fairly good fit is apparent, with high values of the squared correlation coefficients (R^2), which are given in Table 1 along with other parameters of equation (1). Figure 2 and Table 1 show the presence of sharp gradients in the tropical oceanic region from 12°N to 20°S; with BC decreasing gradually as we move down south; the lowest value being at $\sim$20°S. Beyond 20°S, there is no perceptible latitudinal dependence of M_B.

[8] Another point that has emerged is the temporal change in Λ_D in the tropical region. As the initial and final phases of the expedition (between west coast of India and Mauritius) followed nearly the same track, the difference seen in the amplitude and gradient in Table 1 indicate that the concentrations are greatly reduced in the final phase and the growth rate have become quite shallow (Λ_D increasing from 7.49° in January to 9.25° in March) possibly associated with the change in synoptic conditions. Such seasonal

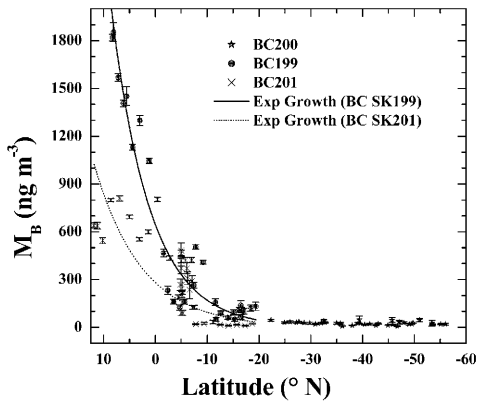

Figure 2. Latitudinal variation of the mass concentration of aerosol BC over the tropical and southern ocean. The dotted curves are best fitted to the measurements of the initial (short dot) and final (short dash) phases.

Table 1. Latitudinal Gradient of BC and Composite Aerosols

Aerosol Type	Cruise Phase	Period	A (ng m^{-3})	Λ_D (°)	R^2
BC	(Kochi - MRU)	Jan 2004	643.7 ± 5.2	7.49 ± 0.06	0.930
BC	(MRU - Karwar)	Mar–Apr 2004	286.49 ± 2.27	9.25 ± 0.08	0.695
Composite	(MRU - Karwar)	Mar–Apr 2004	1643	9.48	0.746

change in BC concentration is observed earlier also over the Arabian Sea/tropical Indian Ocean.

[9] Based on cruise observations over the Arabian Sea in 2003, *Babu et al.* [2004] have reported that the BC mass continuously decreases from winter monsoon, to inter-monsoon and summer monsoon seasons (SMS); a decrease by a factor of as much as 7. The initial phase in this study corresponds to the winter monsoon season considered by *Babu et al.* [2004], while the final phase to the inter-monsoon season. The results are in general conformity and the absolute magnitudes of M_B in the Arabian Sea observed during March 2004 are generally of the same order as seen in March 2003. The SMS values over the Arabian Sea observed by *Babu et al.* [2004] are only marginally higher than the values seen over SO. Thus the synoptic nature of the circulation has a very significant impact on the BC concentration over the oceanic regions down Asia. Significant decrease in M_B over Tropical IO from January to March is also reported by *Mayol-Bracero et al.* [2002].

[10] In contrast, the mass concentration of composite aerosols does not show such remarkable changes. In the top panel of Figure 3, the latitudinal variation of M_T is shown. Similar to M_B, M_T also decreases with latitude from the continental landmass up to ~20°S. In the SO, M_T was highly variable; being as low as ~15 μg m^{-3} on some days and going up to as high as ~60 μg m^{-3} at some locations; the high values being associated with the gusty conditions encountered during the cruise (during February 15 to 24 at ~45–50°S latitude). In the tropical oceanic region, north of 20°S, the latitudinal variation can be parameterised using equation (1) fairly accurately and the parameters are given in Table 1, 3rd row.

[11] The collocated measurements of M_B and M_T enabled us to compute the mass-mixing ratio of BC, a parameter quite important in assessing the radiative impacts. For this we estimated the average value of M_B corresponding to each sampling done using the HVS, by averaging the aethalometer data over the duration of HVS measurements. Thus the ratio M_B/M_T is estimated as the mass-mixing ratio F_{BC} of BC. This value is assigned to the mid point latitude (of the latitude range covered by the ship during the HVS sampling duration). In the middle panel of the Figure 3, we show the latitudinal variation of F_{BC}; the dotted line representing the fit to a peak function.

[12] The curve shows two important points

[13] 1. To the south of ~18°S, F_{BC} is too low (~0.2 to 0.3%) and does not show any significant latitudinal variation.

[14] 2. To the north, in the tropical oceanic region, F_{BC} is high; varying from ~1% to 4%. More interestingly, the higher share of BC does not over near the coast (where the anthropogenic sources are the strongest) but quite far, over the ocean near to equator. The data analysis shows the peak to be locate at 0.8°N, with an amplitude (A) of 0.033 (for the peak mixing ratio) and has a width (w) of ~5°.

[15] This observation is quite important in as much as that the absorbing nature of the aerosol is higher over the ocean. This might be arising from the rather longer residence time of BC (due to its smaller size and chemical inertness) compared to the composite aerosols. Owing to the presence of coarser particles and more hygroscopic components, the concentration composite aerosols deplete faster during advection to farther oceanic regions by sedimentation and cloud scavenging. Consequently, the share of BC to total mass increases initially, before the sea-salt aerosols start dominating the composite at large distances. It is interesting to note here that aircraft measurements by *Novakov et al.* [1997] during the TARFOX have also shown the fraction of carbonaceous aerosols to increase with altitude in the lower atmosphere and to reach a peak at locations where the composite aerosol mass is low. These observations, though made in a different context, fall in line with ours. Higher BC mass fraction implies increased atmospheric forcing efficiency for the aerosols. Thus even though BC is produced mainly over the continent, its impact is more at farther locations over the ocean. However, even the peak F_{BC} (~3.5%) in our measurements is lower than the mean fraction (~5%) reported by *Novakov et al.* [1997].

[16] The large variation in M_T in the SO, despite the absence of external sources, is striking. This indicates the possible impact of sea-surface winds and the consequent enhancement in sea-salt aerosol production. It is well known

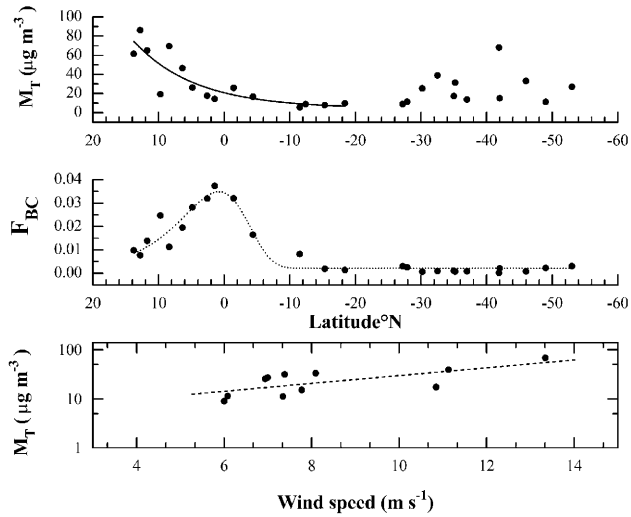

Figure 3. Latitudinal variation of the M_T (at the top); and the BC mass fraction, F_{BC}, (in the middle panel). The points are average of each latitudinal bin. The bottom panel shows the variation of M_T with wind speed over the southern ocean. The dashed line is regression fit to equation (2).

that over the open ocean (where sea-salt is the dominant aerosol species) the aerosol concentration increases with wind speed as

$$M_T(u) = M_{T0}\, e^{bu} \quad (2)$$

where b is the exponent for wind speed dependence and u is the mean wind speed. Several studies [e.g., *Lovett*, 1978; *Exton et al.*, 1985] have shown the value of b to be ~0.16. We have examined this for the SO in the bottom panel of Figure 3, which shows the variation of M_T (in log scale) with the mean wind speed during the period of HVS sampling. Despite the scatter, the figure shows a fit to the equation (2) with a correlation coefficient of 0.70 and slope of 0.18 ± 06; well in agreement with the values reported. This shows that the large variation in M_T over SO is due to wind-speed generated sea-spray.

4. Summary and Conclusions

[17] 1. Collocated measurements of composite aerosols and BC made over the Arabian Sea, tropical Indian Ocean, and the Southern Ocean as far as 56°S were made during a trans-continental cruise experiment.

[18] 2. The BC mass was extremely low (<50 ng m^{-3}) and remarkably steady (in space and time) in the Southern Ocean. In contrast, large latitudinal gradients were present north of ~20°S, with BC increasing exponentially to the north, reaching values as high as 2000 ng m^{-3} in the northern Arabian Sea.

[19] 3. In contrast, BC mass mixing ratio showed a distinctly different latitudinal variation, with a peak close to the equator and decreasing on either side.

[20] 4. Large variations in composite aerosol mass observed over Southern Ocean were found to be due to enhanced production of sea-salt aerosols in response to sea-surface wind speed.

[21] 5. The large latitudinal gradients and spatio-temporal changes in BC concentration and its mixing ratio may have significant implications to regional and global climate, which needs to be investigated separately.

[22] **Acknowledgment.** The Southern Ocean cruise was designed by the National Centre for Antarctic and Ocean Research, and the authors are grateful for the opportunity provided onboard the cruises for aerosol measurements and thank the chief scientist M. Sudhakar and officers for all the support.

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S. S. Babu, K. K. Moorthy, and A. Saha, Space Physics Laboratory, Vikram Sarabhai Space Centre, Trivandrum, Kerala 695 022, India. (krishnamoorthy_k@vssc.org)

S. K. Satheesh, Centre for Atmospheric and Oceanic Sciences, Indian Institute of Science, Bangalore, Karnataka 560 012, India.