MAUSAM, **56**, 1 (January 2005), 287-292

551.510.42 : 551.46.072 (267.37)

First observations of aerosol black carbon over coastal Arabian Sea during ARMEX

S. SURESH BABU, K. KRISHNA MOORTHY and S. K. SATHEESH*

Space Physics Laboratory, Vikram Sarabhai Space Centre, Trivandrum – 695 022, India

**Centre for Atmospheric and Oceanic Sciences, Indian Institute of Sciences, Bangalore – 560 012, India*

e mail : k-k-moorthy@eth.net, krishnamoorthy_k@vssc.org

सार – अरबसागर मानसून प्रयोग (आरमेक्स) के अंतर्गत अरबसागर के तटीय क्षेत्रों में इंटर मानसून और ग्रीष्मकालीन मानसून ऋतुओं (मार्च से जून 2003) के दौरान ब्लैक कार्बन और सम्मिश्र वायुविलयों के संहति सांद्रणों के परिमाण पहली बार व्यापक रूप से लिए गए। परिणामों से पता चला है कि मार्च में ~ 700 एन. जी. एम$^{-3}$ से ब्लैक कार्बन के औसत सांद्रण में (7 के फैक्टर से) उत्तरोत्तर तथा निरतंर ह्रास, जून में अत्यंत कम ~104 एन. जी. एम$^{-3}$ हो गया। मार्च में दैनिक परिवर्तन अत्यंत मंद पड़ गए, उत्तरोत्तर मंद पड़े और मई और जून के महीनों तक लगभग समाप्त हो गए। इसके अनुरूप सम्मिश्र वायुविलय तंत्र ब्लैक कार्बन के अंश में ह्रास तेजी से होता है।

ABSTRACT. Extensive measurements of the mass concentrations of BC and composite aerosols were made for the first time during the inter-monsoon and summer monsoon seasons (March to June, 2003) over coastal Arabian Sea region under the Arabian Sea Monsoon Experiment (ARMEX). Results showed a gradual and continuous decrease (by a factor of 7) in the mean concentration of BC from ~700 ng m^{-3} in March to reach as low as ~ 104 ng m^{-3} in June. The diurnal variations were very weak during March, weakened progressively and almost vanished by May and June. Consistent with this, there is a rapid decrease in the share of BC to the composite aerosol system.

Key words − ARMEX, Aerosol black carbon, Inter-monsoon, Summer monsoon.

1. Introduction

Black Carbon (BC) aerosol is the light-absorbing fraction of atmospheric aerosols, which is a by-product of all combustion processes and hence generally used as an indicator to human activities. Being chemically inert, BC has long residence times in the atmosphere and can be transported to long distances. Its high absorption in the visible and near IR region is capable of causing significant changes to the aerosol radiative forcing, both the direct and indirect; and even changing its sign. To date, numerous studies on the radiative effect of aerosols have focused on the role of anthropogenic sulfate [Charlson *et al*., 1991, 1992; Kiehl and Briegleb, 1993]. In contrast, quantitative estimates of direct radiative forcing due to black carbon have been given by a few researchers [Haywood and Shine, 1995; Haywood and Ramaswamy, 1998; Babu *et al*., 2002]. This is mainly due to the difficulty in obtaining data on black carbon needed to construct a reliable global emission inventory. In addition, the scarcity of field measurements of black carbon, particularly in remote regions, makes it difficult to verify global dispersion calculations.

Due to the primarily anthropogenic nature of its origin, concentrations of BC are expected to be substantially lower over oceanic regions than on landmass. However, several investigators have reported significant amounts of BC over even remote oceanic locations, which are attributed to advection from adjoining continents and long-range transport. Notwithstanding the above, there are no measurements of aerosol BC over the Arabian Sea adjoining Indian peninsula; particularly during the inter-monsoon period (March-April, when the prevailing synoptic winds are in transition from the NE winter circulation to the SW summer circulation) and in the summer monsoon season (June to September). The ARMEX field campaigns of 2003 provided a unique opportunity to make such measurements for the first time in India.

Extensive and collocated measurements of the mass concentration of total (composite) aerosols and BC were made onboard the last two scientific cruises (SK 190 and SK 193) of ORV Sagar Kanya under the ARMEX campaign of Indian Climate Research Programme (ICRP).

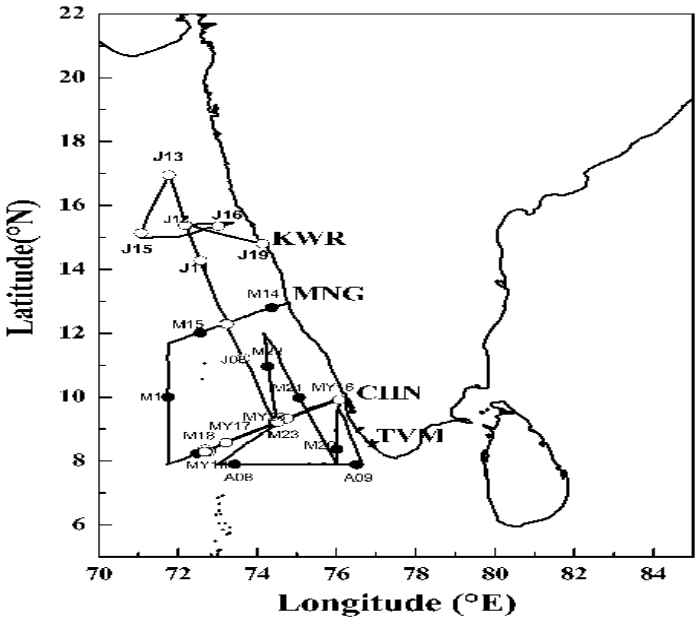

Fig. 1. Cruise tracks with the dotted lines joining the filled circles representing the inter-monsoon cruise and the solid line joining the open circles show the summer monsoon tracks. The location of the coastal station Trivandrum is also marked (TVM)

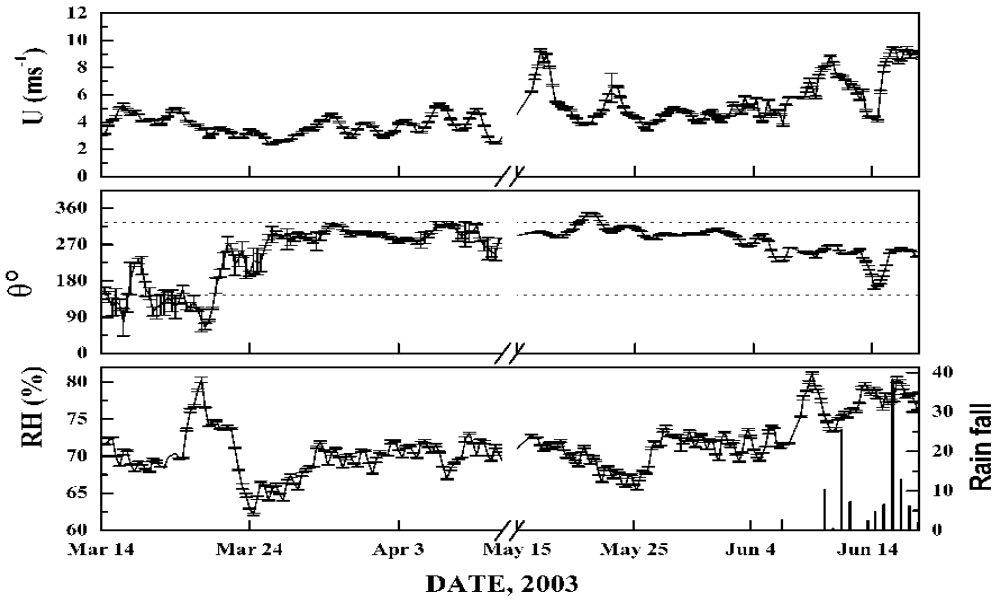

Fig. 2. The variation of surface meteorological parameters during the cruises, panels from top to bottom show the horizontal wind speed (U), direction (θ), relative humidity RH (%) and daily total rainfall (mm)

SK190 was during the inter-monsoon period (March-April) and SK 193 was during the summer monsoon (May-June) period. These measurements were compared with the long-term measurements of BC made as a part of Aerosol Climatology and Effect (ACE) project of ISRO-Geosphere Biosphere Programme, at a coastal location (Trivandrum, TVM) very close to the Arabian Sea.

2. Experimental details and database

The measurements onboard SK190 were from 14 March to 10 April, 2003 (inter-monsoon season) when the synoptic winds are in transition from the north easterly winter circulations to the north westerly/ westerly summer circulations, while SK193 were conducted during the

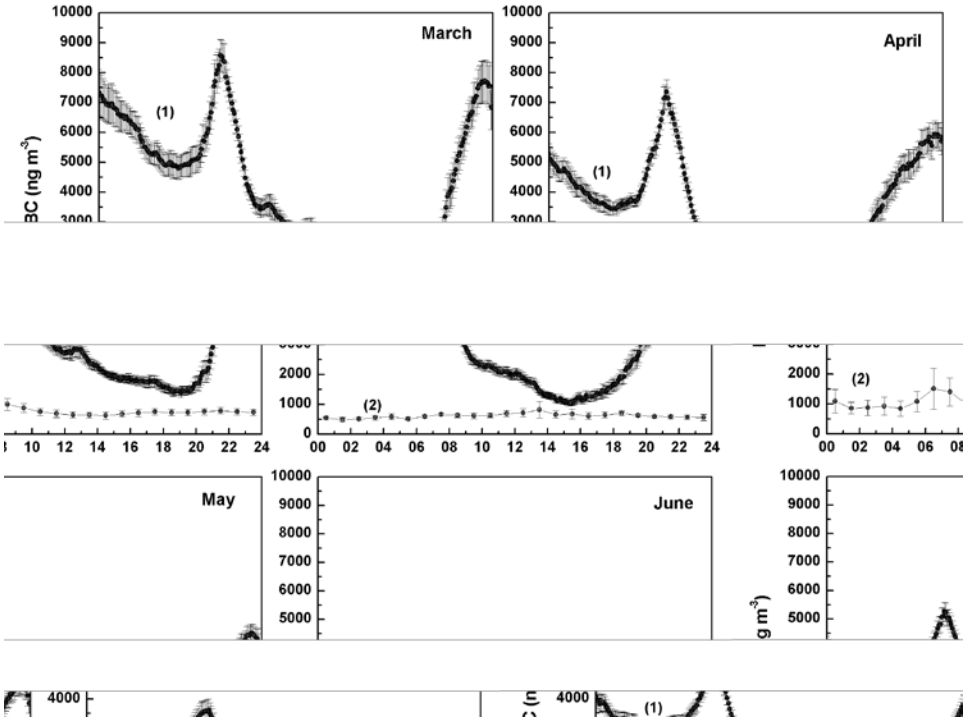

Fig. 3. Diurnal variation of BC mass concentration for March, April, May and June over TVM (1) and Arabian Sea (2). The points are monthly means and the vertical bars the standard deviations

beginning of the summer monsoon (15 May to 19 June, 2003) when the north westerly winds have established. The cruise tracks, shown in Fig. 1, are confined mostly to the coastal Arabian Sea adjoining peninsular India.

Estimates of the mass concentration (M_b) of BC aerosols were made using an Aethalometer; model AE-21 of Magee Scientific, USA [Hansen *et al*., 1984; Hansen, 1996; Babu and Moorthy, 2002], operated from the ship's cabin. The aethalometer measures the concentration of absorbing aerosol by the theory of optical attenuation of light. An aerosol sample is deposited continuously on quartz filter tape. It is assumed that aerosol absorption is mainly due to black carbon aerosol. A beam of light is directed at the aerosol deposit spot on the tape and the attenuation of light due to aerosol absorption is measured by detection of transmitted light through the sample on the filter. The measurement is made at successive time periods and during every measurement interval a light source of wavelength 880 nm shines on the aerosol deposited on the filter. A photocell beneath the filter detects the transmitted light and as the concentration of absorbing species increases, the transmitted light decreases. The tape is automatically advanced when the loading on the filter attenuates 25% of the original transmitted light. The lamp operates for about one minute during the measurement period. Measurements are also performed when the lamp is off to determine zero offsets. A reference beam measurement is performed to correct for small changes in the output of the lamp.

The sample flow rate is provided by an external pump and measured by a mass flow meter inside the instrument. Ambient air was aspirated through an inlet pipe from the front of the ship (sampling into the wind), at ~12 m above the water level. During the first cruise, the instrument was operated at a flow rate of 4l min^{-1} and at a time base of 5 min, while in the second cruise the flow rate was kept at 5l min^{-1} and the time base was increased to 30 min in view of the highly reduced BC concentrations.

A 10-channel quartz crystal microbalance (QCM) cascade impactor (model PC-2, California Measurements Inc., USA) was also used for near-real-time measurements of the total mass concentration (M_t) of ambient aerosols in the marine atmospheric boundary layer. The QCM makes measurements of aerosol mass concentration (μg m^{-3}) in its 10 size bins, with 50% cut off diameters at 25 μm, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1, 0.05 μm for the stages 1 to 10 respectively. More details of the instrument, the data collection procedures, precautions to be taken and error budget are given in an earlier paper (Pillai and

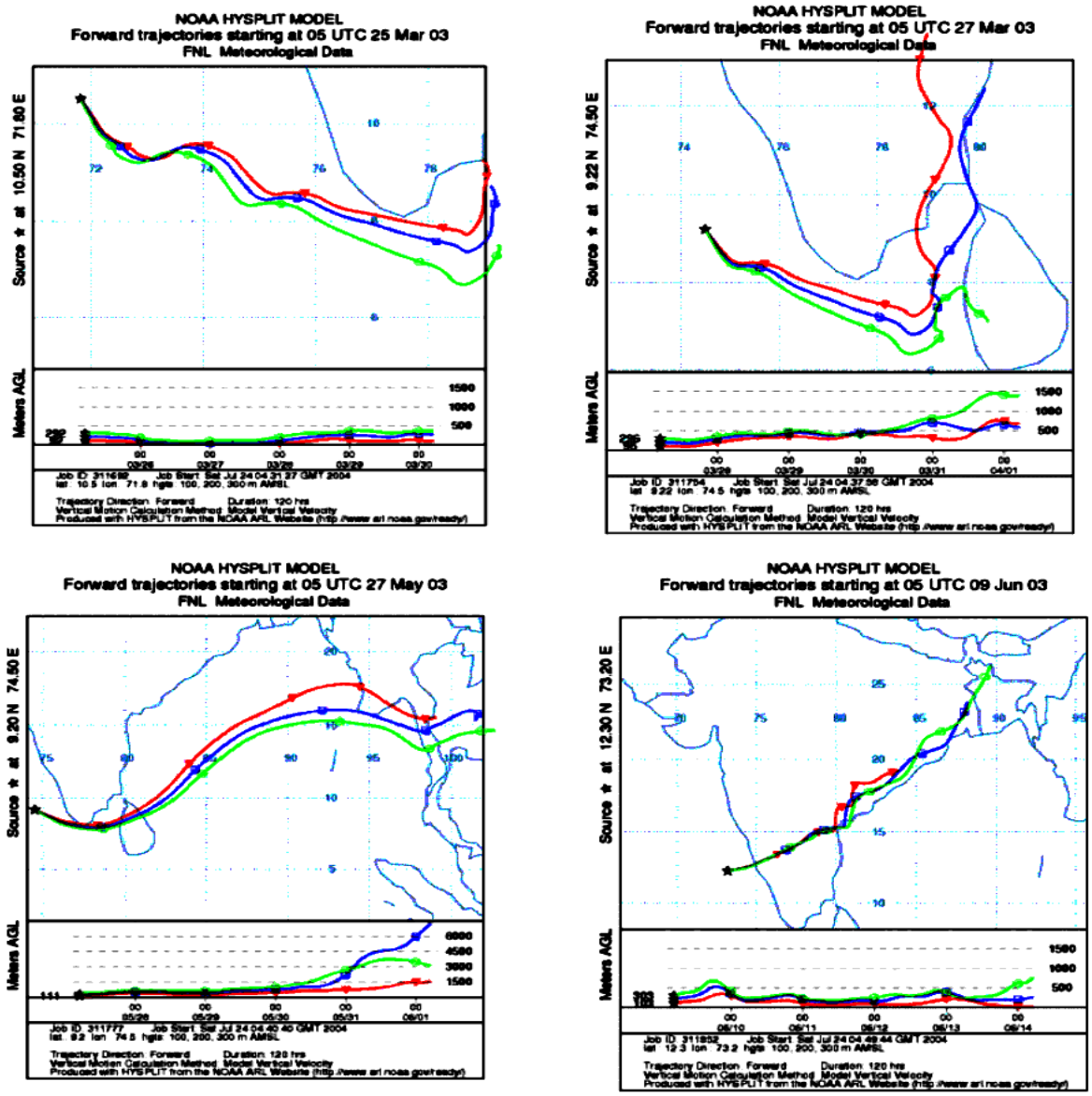

Fig. 4. Four typical examples of air back trajectories during the ARMEX period showing potential continental advection

Moorthy, 2001). The sampling duration varied from 5 to 8 min depending on the aerosol loading. The instrument was kept at the bow side ship deck, just below the bridge, at ~10 m above the water level and operated only when the ship was in motion at the normal cruising speed so that the instrument aspirates the clean oceanic air coming on to the ship, uncontaminated by the ship's exhaust or any possible ship-based emissions. In view of the sensitivity of the quartz crystals in the QCM to changes in RH at higher RH levels, the QCM was operated only when the RH <~78%. The typical uncertainty in the measured mass concentration was in the range 10 to 15% with the higher percentages for lower mass concentrations.

Information on the surface meteorological parameters was obtained from the shipboard measurements at every minute. These included wind speed and direction (corrected for ship's motion), relative humidity (RH) and temperature. These data are averaged at six-hour interval and then smoothed using a 4-point running mean to get the average characteristics as shown in Fig. 2. The bottom panel of Fig. 2 shows the variations of relative humidity (which was mostly below 75%) and rainfall. Except for an occasional drizzle on 19th and 20th March, there was no rainfall during the entire period of the first cruise, while rainfall was extensive during the second cruise.

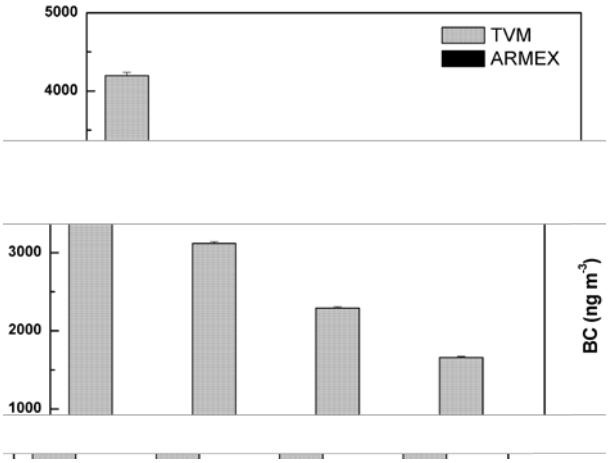

Fig. 5. The comparison of the monthly averaged mass concentration of BC observed over Arabian Sea with the climatological values (2000-02) of the coastal station, TVM

3. Results and discussion

3.1. *Diurnal variations*

Average diurnal variations of BC aerosols observed in four different months (March, April, May and June) along with the climatological monthly mean diurnal variations observed for the coastal station, Trivandrum [Babu and Moorthy, 2002], are shown in Fig. 3. In March, when the prevailing winds are mainly weak north-easterlies and the average wind speed is low, BC showed a weak diurnal variation with a morning peak, daytime low and a very small nocturnal high, even though the variations are much weaker than that seen over the land. This diurnal variation is due to the diurnal variation of the atmospheric boundary layer and morning peak is attributed to the fumigation effect in the boundary layer [Stull, 1988; Babu and Moorthy, 2002]. Generally, marine boundary layer is shallower (~500 m) and shows weaker diurnal variations than those over land; because due to its large heat capacity, ocean surface will not respond quickly to the changes in solar heating. By April, the winds have been established as north-westerlies and interestingly diurnal variation almost vanishes over the sea. In May and June, it can be seen that the winds are almost westerlies/ north-westerlies, directed from the open ocean and not only that the diurnal variations in M_b vanish completely, but the absolute values are also highly reduced. However, over land (at TVM, Fig. 1), well-defined diurnal variation for BC is observed, with seasonally changing amplitudes, which become smaller during the summer monsoon season. Similar diurnal variations are observed for BC at other continental sites also by several other investigators [Allen *et al.*, 1999; Chen *et al.*, 2001].

TABLE 1

Mean BC mass fractions over the Arabian Sea

Period	BC mass fraction (%)	
	To composite aerosol	To $PM_{2.5}$
March 2nd half	2.47 ± 0.33	4.3 ± 0.2
April 1st half	1.59 ± 0.22	2.69 ± 0.31
May 2nd half	0.81 ± 0.10	1.41 ± 0.06
June 1st half	0.50 ± 0.07	1.1 ± 0.09

3.2. *Comparison of monthly mean BC over Arabian Sea with Trivandrum*

One of the major sources of BC over Arabian Sea would be the anthropogenic air pollution from the adjacent continents. Both the prevailing winds during this period as well as the shipboard measurements indicate possibilities of such advection. However, this aspect was also examined in detail by computing air back trajectories using the NOAA – HYSPLIT model (http.//www/arl.noaa/ gov/ready.html) for the campaign period. Four typical examples are shown in Fig. 4, which all show potential continental advection (particularly form the east coast of India). Nevertheless, quantitative estimation of BC fluxes from land to sea, one needs BC concentration vertical profiles and horizontal distributions along the shoreline. Unfortunately, other than the measurements at TVM [Babu and Moorthy, (2002)] there were no BC measurements available so far in this region. Moreover, our measurements over the sea were for a limited period and were obtained intermittently. Nevertheless, Fig. 5 shows the comparison of the monthly averaged mass concentration of BC observed over Arabian Sea with the climatological values (2000-02) for TVM. Since BC is considered as an important tracer of pollutant transport, it is interesting and important to note the large difference in the monthly averaged value of BC mass concentration between TVM and Arabian Sea.

3.3. *Mass fraction of BC*

The total aerosol mass concentration (M_t) obtained from the QCM is plotted against the corresponding value of BC mass concentration (M_b) from Aethalometer as a scatter plot. The mass fraction of BC to the total mass is obtained by making a best fit to the data through the origin. The mean values of BC mass fractions thus obtained are given in Table 1. There is a gradual decrease in BC mass fraction from March to June. Babu and Moorthy (2002) reported that the share of BC to the total mass is very high (> 9%) during the dry months (December to March) and lowest during the monsoon months (~ 3 to 4%) over TVM. Though the mass fraction

of BC to the composite aerosols is important in aerosol modeling for assessing radiative impacts, the share of BC to $PM_{2.5}$ is important from the environmental and health perspectives. As the QCM measured size resolved mass concentration, it was possible to estimate the $PM_{2.5}$ concentration from each measurement and this was used to estimate the mass fraction of BC to $PM_{2.5.}$ These values averaged for each month are given in the last column of Table 1. It may be noticed that the share of BC to $PM_{2.5}$ is higher than that to the composite aerosols. As aerosol BC has deleterious health effects, higher concentration in the resperibale particle size regime is a matter of concern. Nevertheless this share also decreases considerably from inter-monsoon to summer monsoon season.

4. Conclusions

(*i*) The diurnal variations of BC are remarkably weak over ocean and vanish completely by May/ June.

(*ii*) The concentration of BC decreases continuously, from ~700 ng m^{-3} in March to ~104 ng m^{-3} by June. The share of BC to total mass concentration decreases from 2.5 % in March to 0.5 % in June.

(*iii*) The share of BC to $PM_{2.5}$ is much higher than that to the composite aerosols; nevertheless it also decreases from ~4.3% in March to 1.1% by June.

Acknowledgements

This work was done jointly under the I-GBP and ICRP. We thank the Department of Ocean Development (DOD) and chief scientists of the cruises for the shipboard facilities and Prof. R. Sridharan, for the support and useful discussions. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http.//www/arl.noaa/gov/ready.html) used in this publication.

References

Allen, G. A., Lawrence, J. and Koutrakis, P., 1999, "Field validation of a semi continuous method for aerosol black carbon (Aethalometer) and temporal patterns of summertime hourly black carbon measurements in south-western PA", *Atmos. Environ*., **33**, 817-823.

Babu, S. S. and Moorthy, K. K., 2002, "Aerosol Black Carbon over a Tropical Coastal Station in India", *Geophys. Res.Lett.*, **29** (23), 2098, 13-1 to 13-4, doi:10.1029/2002GL015662, 05.

Babu S. S., Satheesh, S. K. and Moorthy, K. K., 2002, "Aerosol radiative forcing due to enhanced black carbon at an urban site in India", *Geophys. Res Lett*., **29** (18), art. no. 1880.

Charlson, R. J., Langner, J., Rodhe, H., Leovy, C. B. and Warren, S. G., 1991, "Perturbation of the northern hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols", *Tellus*, **43AB**, 152-163.

Charlson, R. J., Schwartz, S. E., Hales., J. M., Cess., R. D., Coakley., J. A., Hansen, J. E. and Hoffman, D. J., 1992, "Climate forcing by anthropogenic aerosols", *Science*, **255**, 423-430.

Chen, L. W. A., Doddridge, B. G., Dickerson, R. R., Chow, J. C., Mueller, P. K., Quinn, J. and Butler, W. A., 2001, "Seasonal variations in elemental carbon aerosols, carbon monoxide and sulfur dioxide : Implications for sources", *Geophys. Res. Lett*., **28**, 1711-1714.

Hansen, A. D. A., Rosen, H. and Novakov, T., 1984, "The Aethalometer, an instrument for the real-time measurement of optical absorption by aerosol particles", Sci. Total Environ., **36**, 191-196.

Hansen, A. D. A., 1996, "Magee Scientific Aethalometer User's Guide", Magee Sci. Company, Berkely, Calif., p56.

Haywood, J. M. and Shine, K. P., 1995, "The effect of anthropogenic sulphate and soot on the clear sky planetary radiation budget", *Geophys. Res. Lett*., **22**, 603-606.

Haywood, J. M. and Ramaswamy, V., 1998, "Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols", *J. Geophys. Re*s., **103**, 6043-6058.

Kiehl, J. T. and Briegleb, B. P., 1993, "The relative roles of sulfate aerosols and greenhouse gases in climate forcing", *Science*, **260**, 311-314.

Pillai, P. S. and Moorthy, K. K., 2001, "Aerosol mass-size distributions at a tropical coastal environment: Response to mesoscale and synoptic processes", *Atmos. Environ*., **35**, 4099-4112.

Stull, 1988, "An introduction to boundary layer meteorology", Kluwer Academic Publishers, Dordrecht.