

# Anthropogenic impact on aerosol black carbon mass concentration at a tropical coastal station: A case study

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**A case study is made on the impact of extensive anthropogenic activity, associated with the Indian festival event (Diwali), on the mass concentration of atmospheric aerosol black carbon (BC) at a remote coastal location. Observations show a large increase in the BC concentration (by a factor of over 3 above the unperturbed background level) associated with this. The sea breeze and the associated boundary layer dynamics appear to perform a cleansing job in the lower atmosphere during the daytime, apparently by re-distributing the aerosols. Nevertheless the effect of the impact persists for several days, decaying gradually in a matter of about a week.**

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AEROSOL black carbon (BC) is that fraction of atmospheric carbonaceous aerosols which is highly light-absorbing (or optically black); the other component is called organic carbon (OC). BC is a universal component of the effluent from the combustion of carbonaceous fuels and is a significant component of atmospheric aerosols. It is produced only by combustion processes (natural or anthropogenic) and not by any known atmospheric reactions. It is not degraded under atmospheric conditions and hence wet and dry depositions are the only important sinks of BC<sup>1,2</sup>. Consequently, atmospheric lifetime of aerosol BC is of the order of days to weeks depending on the meteorology. So medium- and long-range transports become important in its spatial distribution. Observations of BC in the Antarctic regions<sup>1</sup> are attributed to such long-range transport.

BC has become the subject of interest in the recent years for a variety of reasons. For both the direct and indirect effects of aerosols on the global radiative balance, BC has an important role, since BC is a strong absorber of radiation in the visible and near-infrared part of the spectrum, where most of the solar energy is distributed. The absorption cross-section of BC is very high in the visible spectrum, with reported values in the range 7 to 10 m<sup>2</sup> g<sup>-1</sup> (refs 3–5). This large optical absorption cross-section of BC leads to the extinction of radiation, which produces brown skies and reduced visibility. As the

absorbed energy causes heating of the atmosphere, BC complements to greenhouse warming too. Increase in the concentration of BC will lead to the reduction in the aerosol single-scattering albedo and if an aerosol layer rich in BC occurs above a reflective cloud or over land areas of large surface reflectivity, there will be a net positive atmospheric forcing. BC may also act as condensation nuclei, altering the size distribution and optical properties of clouds<sup>6</sup>. It is known that BC plays an important role in atmospheric chemistry because of its catalytic properties<sup>7</sup>. Its surface can act as a site for the catalytic oxidation of SO<sub>2</sub> to sulphate and for the destruction of ozone<sup>8</sup>. The great porosity of BC particles enhances its ability to adsorb other species in vapour phase. They may therefore act as carriers for the transport and localized deposition of harmful compounds to the human pulmonary system and cause toxic effects. The BC particles are frequently small enough (< 1 μm) to be readily inhaled, where they have a certain probability of getting deposited in the lungs or other airways. Thus study of atmospheric BC is very important. In this paper we present the results of a case study showing the impact of anthropogenic activities (associated with the famous Indian festival, Diwali) on the BC mass concentration at a tropical coastal location – Thumba, Thiruvananthapuram.

## Experimental details

### *Instrument*

Regular and near-real-time measurements of the mass concentration of atmospheric BC were carried out using a dual beam aethalometer (model AE-20 U of Magee Scientific Inc., USA). The aethalometer is a fully automatic instrument that uses a continuous filtration of ambient aerosols and optical transmission measurement technique to estimate the mass concentration of BC. The principle of operation and details are described in detail elsewhere<sup>9</sup> and are therefore not repeated here. In our study, the aethalometer was configured for a flow rate of 3 l min<sup>-1</sup> and the measurement cycle (time base) was set to 5 min. The instrument then provides BC mass concentration in

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$\text{ng m}^{-3}$ , every 5 min, round-the-clock on each measurement day.

### *The event and study period*

The specific 'event' of our study is the Diwali festival of India, which is a festival of lamps. In the year 2000, it fell on 26–27 of October. The festival is very widely celebrated throughout India, and is marked by the extensive use of fireworks of all sorts during the night of the festival (26 October in the year 2000) and early morning period of the following day. Even though the peak of the activity occurred on the night of 26 October and the following early morning, it persisted on the previous and next days, though at a much lower level (as is the usual practice). The celebrations are extensive in north India and Tamil Nadu, but only at a low level in and around Thiruvananthapuram. Nevertheless, extensive fireworks took place on the night of 26 October, particularly in the densely-populated urban areas (almost every house contributing its share). As all these fireworks

are invariably potent sources of soot also, it is logical to expect an impact on aerosol BC. To understand this we considered a period of study from 12 October 2000 to 2 November 2000, with the period from 12 to 24 October taken as the control representing the prevailing state and the rest of the days to monitor the growth and decay of the impact.

### *Measurement site and prevailing meteorology*

The studies are carried out at Thumba, Thiruvananthapuram ( $8.5^{\circ}\text{N}$ ,  $77^{\circ}\text{E}$ , 3 m msl) situated near the south-west coast of the Indian peninsula. The actual sampling site is a remote, plain, coastal area, not in proximity to any major industrial and/or urban activities, and is located  $\sim 500$  m due east of the Arabian Sea coast and  $\sim 10$  km north, north-west of the urban area. The human activity is highly subdued over an extent of  $\pm \sim 3$  km parallel to the coast and about 1 km across it about the measurement site, as the area is almost uninhabited. A schematic of the site is shown in Figure 1.

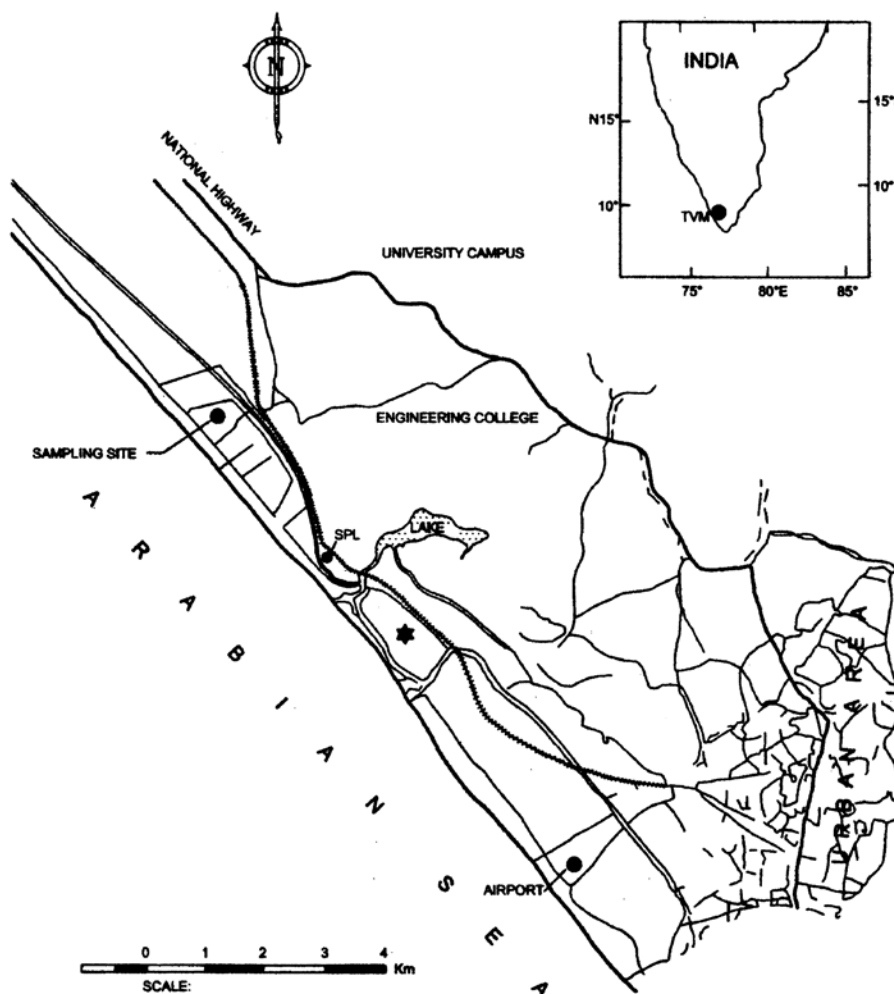


Figure 1. Schematic map of the sampling site.

The prevailing meteorological conditions during this period are characterized by the withdrawal of the S-W monsoon activity and the establishment of N-E monsoon activity. Consequently, the prevailing winds undergo a transition from a nearly north-westerly direction to north-easterly/easterly direction during this month. More details are given elsewhere<sup>10,11</sup>. The rainfall, however, is significant. The total rainfall during this month was 246 mm (which is close to its normal value for the month) and its distribution over the month is shown in Figure 2. Most of it occurred prior to 20 October 2000. Further, being a coastal site, the station also experiences a general diurnal variation in the wind speed and direction (close to the surface) owing to the land/sea breeze activity. The land/sea breeze circulations modify the prevailing winds within the boundary layer, almost daily during the period of study<sup>12,13</sup>. The surface winds are offshore (land breeze) during night and early morning hours, changing over to onshore (sea breeze) during the daytime. Referring to the local coastline geometry, winds arriving at angles between 145 and 325° measured clockwise (with respect to north) constitute a sea breeze. The sea breeze usually sets in between 0800 and 1000 h local time in the morning and land breeze onsets between 1800 and 2000 h in the evening. Around these periods of reversal, near-neutral conditions prevail for a long time, with the sea breeze component of the wind nearly zero or fluctuating between land and sea. Continuous data on wind speed, direction, RH and rainfall are obtained from the meteorological facility of TERLS (Thumba Equatorial Rocket Launching Station), located about 500 m due south-west of the sampling site.

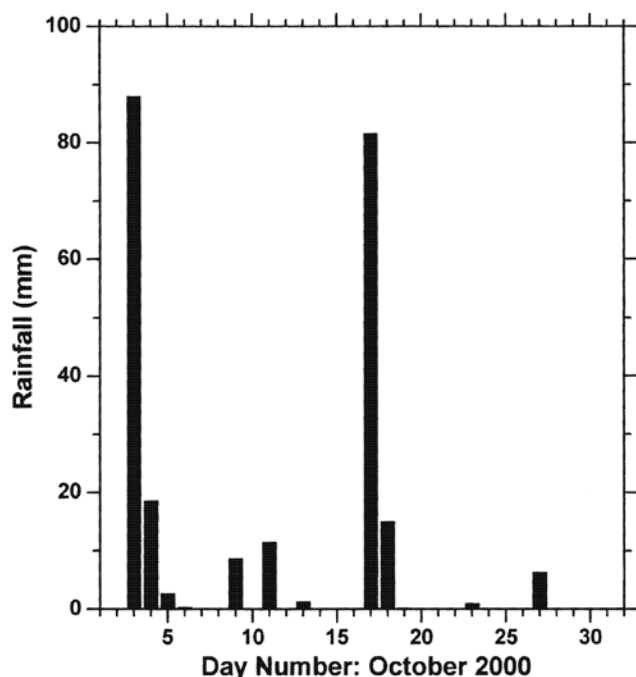


Figure 2. Distribution of daily rainfall for October 2000 (data from meteorological facility, TERLS).

Results and discussion

In order to estimate the impact of the event, it is required to have the information on the normal/average diurnal pattern of BC which prevailed over the location prior to the event. For the purpose of generating the unperturbed background aerosol BC mass concentration representative of the month, the database obtained during the period free from any major events, other than the normal anthropogenic activity typical to the region, was used. As such, the period from 12 to 24 October has been considered for this purpose. The aerosol BC mass concentration measured using the aethalometer on individual days of this period has been grouped together into identical time bins of the day (at 5 min interval) and the mean, standard deviation and standard error are estimated. This way of grouping and averaging smoothens out day-to-day variations in the diurnal pattern and brings out the average diurnal picture. However, this average still has a temporal resolution of 5 min (equal to the time base of the aethalometer).

The mean diurnal variation of BC for October 2000 under normal conditions is shown in Figure 3 (top panel) with vertical bars representing the standard errors. In the bottom panels are shown the corresponding variations in wind direction and wind speed, respectively for that month, averaged as in the case of BC and with respective standard errors represented by the vertical bars. (The wind speed and wind direction are obtained from a cup anemometer mounted at 17 m level on a tower located at the meteorological facility.) Temporal variation of BC depicts a broad nocturnal peak from ~ 2200 to ~ 0100

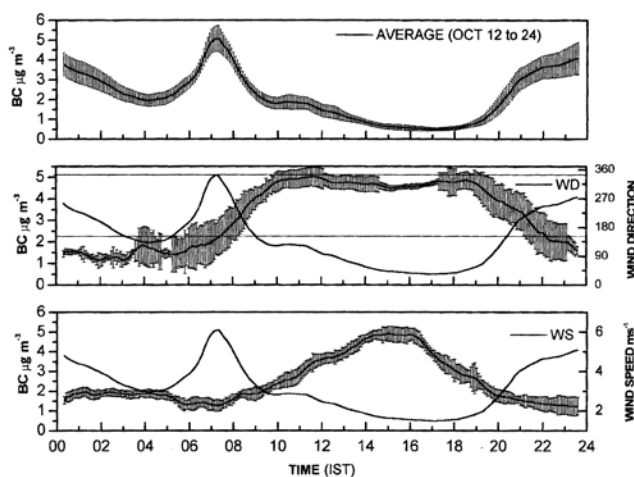


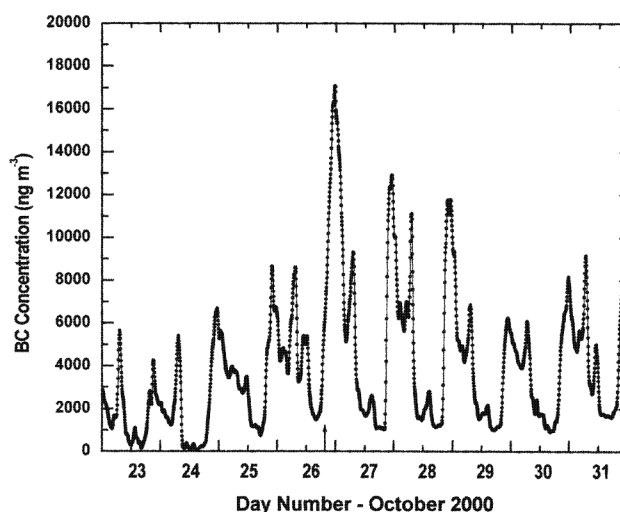
Figure 3. Mean diurnal variation of aerosol BC mass concentration for Thiruvananthapuram for October 2000 (top panel). The vertical bars are standard errors. The corresponding diurnal variation of wind direction and speed are shown (full lines) in the middle and bottom panel, respectively. The mean, BC variations are superposed over these by broken lines. Wind directions lying between the two horizontal dashed lines in the middle panel correspond to sea breeze. Note the larger variances in wind direction around the breeze reversal and the corresponding impact on BC.

local time, when land breeze (LB) prevails. Subsequently, BC concentration decreases and reaches a low value at early morning hours (0400 to 0500), when the local anthropogenic activities are the lowest. This nocturnal increase and the subsequent decrease is associated with the changes in the nocturnal boundary layer characteristics, which produce similar effects in the boundary layer aerosol concentrations also<sup>14</sup>. As the nocturnal boundary layer is shallower than its daytime counterpart by a factor of  $\sim$  three<sup>15</sup>, there is reduction in the ventilation coefficient of aerosols resulting in an accumulation/concentration near the surface. There is a gradual build-up from the morning and a sharp peak occurs between 0700 and 1000 LT. During the period of this peak, the mean BC has higher variances compared to later times of the day. This is due to the combined result of the morning increase in the routine anthropogenic activities and also the horizontal convergence of aerosols in the boundary layer associated with the neutral conditions prevailing prior to the breeze reversal<sup>13</sup>. This reversal does not occur abruptly on any day and not at a fixed time on all days and the wind direction around the morning reversal shows larger variations over the mean compared to other times of the day, as can be seen by the length of the error bars in the middle panel of Figure 3. This is similar to the phenomenon observed with BC also around this time (top panel). As the land/sea breeze circulation is a mesoscale motion set up by the land-sea thermal contrast, prior to the onset of the sea (land) breeze, it has to oppose the existing land (sea) breeze. This leads to shifting winds at the coast between land and sea and a neutral condition prevails similar to a dynamical equilibrium, before the breeze finally takes over and gets established. It is well known that during this neutral condition/period of shifting winds, a horizontal convergence occurs within the boundary layer leading to increase in the aerosol concentration<sup>16</sup>. Wind speeds (total) are also generally low ( $\sim 2 \text{ ms}^{-1}$ ) during this period. Because the land breeze is directed from the continent, particularly from the urban areas of Thiruvananthapuram, where most of the human activity is concentrated, it would be richer in BC aerosols. After the sea breeze has set in at  $\sim 1000$  LT, the speed increases and both the speed and direction stabilize (as can be seen from the shorter error bars in Figure 3, middle and bottom panels), and this brings in marine air which is rather free from BC aerosol (as there are no sources of BC over the sea). This causes a remarkable decrease in BC aerosol mass concentration and it remains low during the entire sea-breeze regime. This is because the BC-rich continental air is swept back by the sea breeze and the polluted air rises to higher levels and gets only partly recycled in the circulation cell. The continued heating of the landmass by the solar radiation results in development of thermal plumes and convective turbulence within the boundary layer and causes an elevation of the boundary layer to higher altitudes<sup>15</sup> (reaching up to 1.5 to 2 km as the

breeze moves quite inland from the coast) during the daytime. This also results in dilution of the aerosol BC concentration near the surface, as the BC burden is now spatially redistributed. The sea breeze speed increases towards afternoon and BC concentration further decreases as the marine air fetches deeper inland<sup>14</sup>. This continues till the breeze weakens and finally reverses. Now we examine the impact of the event (festival activity) over this background.

In Figure 4, we have shown the day-to-day variations of BC mass concentration for the period 22 to 31 October 2001 (each day having its diurnal variations with a nocturnal peak and daytime low). The variations were quite similar on 22 and 23 October and very much close to the mean pattern. However, from the 24 October onwards, there appeared to be a very small build-up in BC as evidenced by the increase in the peak value. On the event day (26 October), the BC concentration shot up to very large values in the early night itself, associated with the peak of the event when the fireworks are at the peak. The nocturnal peak value on this day was more than three times that on the previous days (or the mean of the previous several days); the peak was quite broad and the early morning trough on the next day was higher than the normal.

Another interesting feature seen from Figure 4 is that the decay of the impact (of the event) is clearly seen in the nocturnal peak value, which gets lower and lower progressively with days after the event. However, the daytime minimum does not recover to the values prior to the event (the values seen on 22 or 23 October); rather it stays somewhat higher or even shows a very weak increasing trend. This actually begins on the 25th itself (prior to the event day), thereby suggesting that it is not an effect of the event, but a part of a longer-term normal

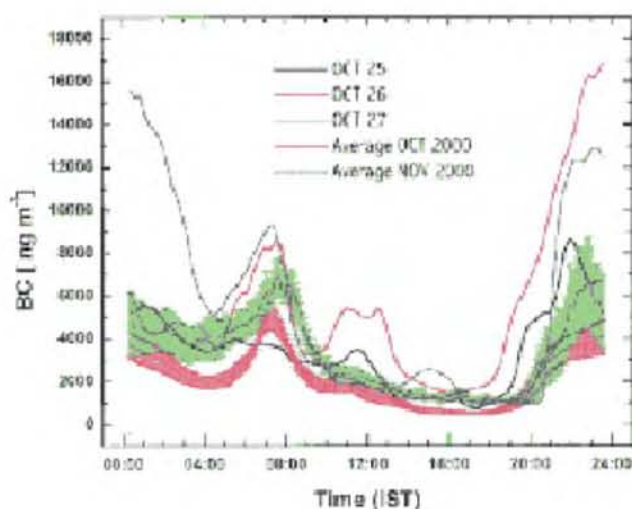


**Figure 4.** Time evolution of aerosol BC from 22 October to 31 October 2000. Vertical arrow on the abscissa shows the occurrence of the event.



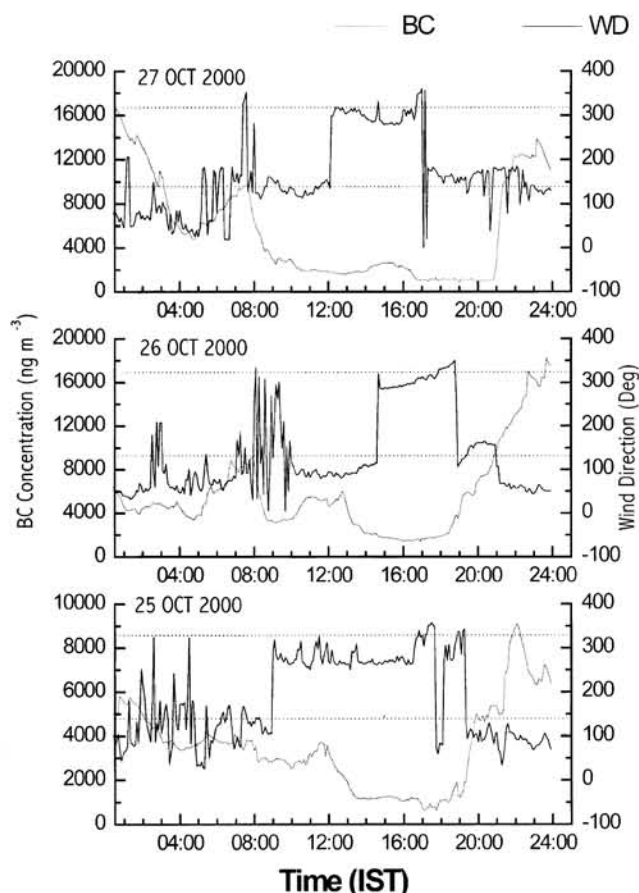
variation (similar to seasonal variation) in the BC concentration. Referring to Figure 2 it is seen that the period from 3 to 18 October had frequent and heavy rainfall (totalling to above 220 mm) and rainfall is the chief removal mechanism for aerosol BC<sup>4</sup>. However subsequently the rainfall was meagre (only an isolated event of ~6 mm on 27th). The strong rains prior to the event thus would have considerably cleansed the atmosphere of BC and the pre-event BC levels thus would be low. However, this is the normal pattern for October, which generally has a fairly good amount of rainfall (associated with the monsoon activity) at Thiruvananthapuram. This, of course, makes the event stand out. The near absence of rainfall subsequently for several days coupled with the continued generation of BC (by the normal activities pertinent to the region) and its longer lifetime result in a steady increase in the average BC level. This is corroborated by further data (not used here), which showed a steady increase in the BC concentration towards the drier months (December–January). It is well known<sup>4</sup> that the average lifetime of BC increases with decrease in the rainfall. This aspect is being investigated separately and hence is not dealt with here. For the present we are content to observe that the sharp rise following the onset of the event on 26th is almost entirely attributable to the anthropogenic impact.

For a detailed analysis of the event and to estimate its impact, the diurnal pattern of the BC mass concentration on 25th (pre-event day), 26th (event day), and 27th (post-event day) is shown along with the diurnal variation of the mean background in Figure 5. Further to the discussion in the preceding paragraph, in Figure 5 we have included the mean BC levels (along with the corresponding standard errors) for both October and November 2000. It is seen that the BC levels in November 2000 are consistently



**Figure 5.** Diurnal variation of aerosol BC for the pre-event, event and post-event days, superposed over the mean background levels (with error bars) for October 2000 and November 2000. Note the consistently higher BC levels in November.

higher than those of October 2000, in line with the earlier statements and as such we concentrate our attention to the mean diurnal pattern of October (obtained using data on days prior to the event) as the control day pattern. The diurnal pattern on 25th is more or less close to this average monthly diurnal pattern. On the event day also, the pattern remained similar to the average (October) pattern in the morning and during the daytime, except for a second short-lived enhancement observed between 1000 and 1300 IST. That short-lived enhancement was due to an unusual oscillation around that period between the land and sea breezes (a shift from sea breeze to land breeze after the initial onset of sea breeze), as can be seen from Figure 6 (middle panel). The initial land breeze started shifting to sea breeze by around 08:00 h and then underwent a series of shifts between land and sea until around 10:00 h and eventually shifted back to land breeze which then continued so till around 13:00 h, before the sea breeze finally got established. Being situated near the coast, the BC at our location has responded to this shift in winds producing a secondary peak (again possibly associated with the convergence, as the sea-breeze cell did not get established). The diurnal variations of BC on the previous/next days (shown respectively at the bottom

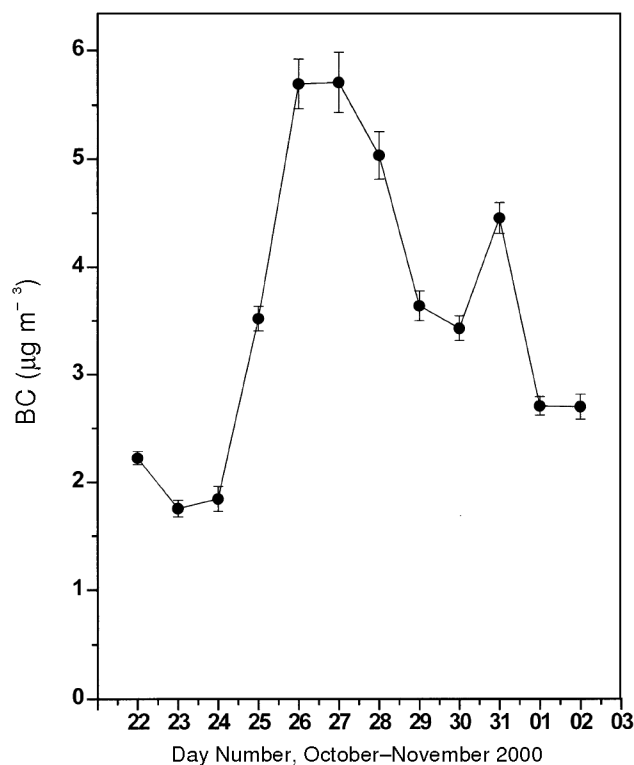


**Figure 6.** Diurnal variation of BC concentration (dotted curve) and wind direction (solid curve) for 25 to 27 October 2001. Directions lying between the dashed horizontal lines correspond to sea breeze.

and top panels) do not show such a pattern as the wind shift is rather smooth on these days, in line with the typical pattern at this station. Subsequent to the onset of sea breeze, though quite late on this day, the BC level started decreasing similar to normal days, but did not reach the normal daytime minimum, because the sea breeze activity was short-lived and not sufficiently strong for far inland advection of the aerosols. Land breeze set in at the early night-time itself ( $\sim 1800$  LT), when the event also starts picking up. There is a very large enhancement in BC mass concentration during the night, with peak value reaching as high as  $\sim 18 \mu\text{g m}^{-3}$  compared to the normal value of 4 to  $5 \mu\text{g m}^{-3}$ . This increase is very clearly caused by the BC emissions from the extensive fireworks that took place on that night in the urban area lying upwind. In fact, every house in the urban area took part in this festival and as land breeze had already set in, the conditions were favourable to advect these particles towards the remote coastal area within a short time. Further, the lowering of the nocturnal boundary layer<sup>15</sup> helps to confine these aerosols closer to the surface during the night. Thus the enhancement in BC mass concentration continued till 27th morning and resulted in the enhancement of the early morning trough also. After the onset of sea breeze on 27th, even though BC concentration was depleted, being swept away by the sea breeze and vertically re-distributed by the boundary layer dynamics, the BC levels still remained higher than that of the normal day values. Again during land breeze period (evening of 27th), the BC mass concentration increased to a high value ( $13 \mu\text{g m}^{-3}$ ), even though there was a very large reduction in the festival activity. This would be partly caused by the subsidence of particles carried aloft during daytime by the increased convective activity due to the reduction in the boundary layer height in the post-sunset period and following night-time and partly due to the longer residence time of atmospheric BC. Further, the particles which get trapped in sea breeze cell get recycled. The diurnal pattern on 28th was also similar to that of 27th, except in the early morning hours and the nocturnal peak value was further lower. This shows that the BC loading decreases gradually, superposed with the diurnal variations associated with breeze activity as well as trends associated with long-term changes.

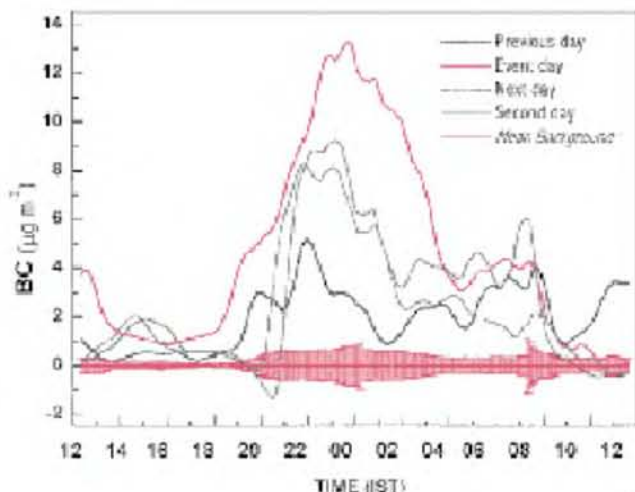
With a view to examining the evolution and decay, we estimated the daily mean BC concentration by averaging the individual values at 5 min intervals of each day. This is done primarily to smooth out the apparent effect of the land/sea-breeze circulation, causing a diurnal variation in BC and diurnal variation in the boundary-layer height, which would lead to a dilution of the BC during daytime (sea-breeze period) when the boundary layer is elevated to  $\sim 1$  to 1.5 km and a concentration of BC in the night-time, when the boundary layer is quite low ( $\sim 50$  to 100 m). This apparent change in the BC is not due to the removal of BC during daytime and production in the night; but

rather due to a spatial re-distribution of the species as it gets mixed in the boundary-layer circulation. Thus the daily mean value of BC will be the more representative one. The variation of this daily mean BC concentration for the period from 22 October to 2 November 2001 is shown in Figure 7. As can be seen from the figure, though the increase in BC associated with the event was almost instantaneous, the decay is gradual, and even after six to seven days the BC concentration has not recovered to the pre-event level. This is partly due to the normal increasing trend of background BC level from October to November (shown in Figure 5) and the long atmospheric lifetime of BC. Another possibility is that during the long residence time of BC aerosols in the atmosphere, a good fraction of the Diwali BC aerosols would have been distributed both horizontally and vertically in the atmosphere by the micro meteorological and mesoscale processes. Using a box model incorporating advection, dry deposition and wet removal, Reddy and Venkataraman<sup>4</sup> have found the lifetime of BC in the atmosphere to vary from  $\sim 3$  days (when wet removal is strongest) to as much as  $\sim 13$  days in dry months with no rainfall. They gave an annual average value of  $\sim 6.5 \pm 4.8$  days for the BC lifetime. Globally, average lifetime of BC is about 8 days. Our observations of about six to seven days are well in line with these values, particularly because there were no significant rains after the event. As the rainfall preceding



**Figure 7.** Evolution of BC associated with the festival. Each point is the daily mean mass concentration. Vertical bars are the standard errors.





**Figure 8.** Event impact on BC concentration as deviation from normal day patterns. Each day is depicted from midday to midday and the normal day values are subtracted from the daily values. Vertical bars are the standard errors.

the event days has made the BC concentrations reach a low level, the sudden increase following the event is a true measure of the anthropogenic impact, whereas the decay need not be a true measure of the lifetime, as it might be contaminated with the trend also. There is a small peak seen in the mean BC on 31 October which is unexplained, and we do not know the actual reason for this. No known major events took place around the study region on any of these days other than during Diwali.

To quantify the enhancement in BC mass concentration associated with the Diwali event, the normal mean values were subtracted from the individual measurements and the deviation from normal is shown as a function of time for four days, one preceding and two following the event day, in Figure 8. For examining the changes during high BC periods, the time axis in Figure 8 is taken from the noon of one day to the noon of the next day. The values obtained on pre-event day (i.e. midday of 25th to midday of 26th) are closer to the zero level. On the event day, there is a large increase (by  $\sim 14 \mu\text{g m}^{-3}$ ) from the zero level. This enhancement in BC persists till the next day morning 0400 (IST). The duration of this nocturnal high is almost 10 h as compared to the normal days, when it persists for only 5 to 6 h. The reduction in the BC concentration in the following daytime is the consequence of the boundary-layer dynamics and breeze reversal discussed earlier. The day-to-day variations in BC are also associated with the day-to-day changes in these. However, a more quantitative estimate of the impact would have been possible, if the amount of combustible matter that was burnt on the event day (similar to a source inventory) was known. But even an approximate estimate of this was not available.

## Summary

The extensive fireworks associated with the Indian festival Diwali, produced a 3- to 4-fold increase in the concentration of aerosol BC at a location quite far away from the centre of the activity. Even though the increase is nearly simultaneous, the decrease is gradual. The sea breeze and the associated boundary-layer dynamics, however, have a cleansing effect on this within the boundary layer, as they help to disperse the carbonaceous aerosols; nevertheless the overall decay of the impact appears to take about a week. At farther inland or non-coastal locations where the diurnal variation in wind direction is insignificant and also in large urban areas (like the Indian metropolitan cities) where the activity will be more vigorous and concentrated, the effect will be more significant and long-lasting. This is important from the radiative forcing angle also, as soot has more greenhouse potential over land whose surface reflectivity is higher.

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