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Original Research

Wintertime Variation of Black Carbon in $PM_{2.5}$ Aerosols Over an Urban Industrial City in East-Central India

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Abstract

$PM_{2.5}$ black carbon aerosols in ambient air of Raipur, India, were monitored for the first time during the peak winter season 1-31 December 2014. The monthly (December) average of BC mass concentration was 23.03±7.50 µgm^{-3} and the hourly average varied from 2.55 to 81.81 µgm^{-3}. Daily average maximum was observed on 23 December (35.25 µgm^{-3}) and minimum (5.76 µgm^{-3}) on 31 December. Daytime average was 17.40 µgm^{-3} and nighttime average was 26.99 µgm^{-3}, with 1.55 being the monthly average ratio for nighttime and daytime concentrations. Monthly average diurnal variation showed two distinct peaks, at morning with 39.84 µgm^{-3} (7:00-8:00 IST (Indian Standard Time)), and at night with 35.55 µgm^{-3} (00:00 -1:00 IST); and two distinct valleys, with lowest BC concentration (7.11 µgm^{-3}) during late afternoon (15:30-16:30 IST), and during early morning (3:30-4:30 IST) with BC mass concentration of 27.04 µgm^{-3}, consistent with the nighttime average. A clear negative correlation between BC mass concentration and wind speed (r = -0.75), relative humidity (r = -0.30) and temperature (r = -0.19) was observed. The back-trajectory at surface level (50 m and 500 m) mostly originated from northeastern India, and for higher altitude (5,000 m), from Middle-Eastern and Arabian countries, except during the last week of December (Indian Ocean). The ratio and difference between BC mass measured at 370 nm and 880 nm showed biomass burning being the dominant cause at night.

Keywords: atmospheric aerosols, black carbon, inversion, Raipur, winter

Introduction

Atmospheric aerosol affects local, regional, and global climate by directly absorbing and scattering the incoming and outgoing solar radiation [1], indirectly by altering the cloud microphysical properties [2], and semi-directly by reducing cloud coverage [3]. Aerosols also have an adverse impact on human health – particularly regarding respiratory and cardiovascular diseases [4]. Black carbon (BC) is a strongly light-absorbing component of the atmospheric aerosol, mainly $PM_{2.5}$ [5] formed during incomplete combustion of fossil fuels, biofuels, and biomass burning [6]. BC contributes to global warming [7-8] by possibly altering precipitation patterns [9] and melting glaciers due to snow albedo reduction [10]. Due to rapid urbanization and industrialization during the last decade, India and China are the largest BC emitters in the world, contributing 25 to 30% of global emissions of BC

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[11]. In South Asia, BC residential emissions from bio-fuel cooking (wood, crop residue, cow dung) are the largest source of BC concentration [12]. To assess the regional and global climate change, a detailed study of spatio-temporal distribution of BC concentration is required; BC being inert with around five days of residence time can be transported over long distances [13]. Although several studies have been carried out in different cities around the world (including Beijing [14], Lahore [15] and, in India, Delhi [16], Trivandrum [17], Ahmedabad [18], Pune [19], Vishakhaptnam [20], and Agartala [21]), few studies are available for the eastern-central plain region of the country such as Nagpur [22]. In most of the studies, BC mass concentration has been reported to be the highest in winter season, particularly in December.

Raipur is the capital city of Chhattisgarh state, with urban, industrial, and trade activities. Its economy is generally based on agriculture and mineral deposit-based industrial activities such as coal-based thermal power plants, metal processing, and cement manufacturing. The World Health Organization [23-24] reported in 2014 and 2016 that Raipur ranked fourth and seventh in the world, respectively, on the list of most-polluted cities as regards ambient air $PM_{2.5}$ concentrations. Chhattisgarh, with its agriculture-based economy directly depending on the monsoon pattern, is one of the most vulnerable areas in South Asia, and any climate change will adversely affect it. Hence, the assessment of BC-induced climate change is very important for this region. In the present work, we have continuously monitored BC mass concentration with the help of a next-generation aethalometer (AE-33-7) in ambient air of Raipur city during the winter month of December, which presents adverse meteorological conditions for dispersion of pollutants; and analyzed the BC mass variation (diurnal, daily, weekly, weekends, daytime nighttime) and meteorological parameters influencing BC concentrations. We also identified possible distant sources using the NOAA-HYSPLIT model, and also by light absorption spectral dependency of BC aerosol at 370 nm and 880 nm. This type of work has been done for the first time for this east-central area of India.

Site Description and General Meteorology

Raipur, situated between 22° 33‘N to 21°14‘N latitude and 82°6‘ to 81°38‘E longitude, is one of the leading industrial and commercial cities of central India. The Raipur district is industrially developed, and it has further scope for new and up-scaling of existing industries as well.

BC mass aerosol in $PM_{2.5}$ sampling was performed during December. The sampling site was at a height of 14.0 m above ground level, on the rooftop (21°15'00"N, 81°36'16.50"E, and 303.360m a.m.s.l.) of the main building of the National Institute of Technology Raipur, and around 200 m away from an urban arterial corridor with annual average daily traffic of about 25,000 vehicles, with several industrial clusters like Gogaon, Urla, and Siltara located to the northeast and northwest about 8 km from the sampling station.

Raipur has a tropical wet and dry climate; temperatures remain moderate except from April to June, when the highest temperature reaches 48ºC. Winter is generally during November to January, with a minimum temperature of 5ºC. As per the atlas of wind roses (1971-96) published by Indian Meteorological Department [25] (2012), the winds are from the southwest (SW) 32% of the time, and 23% of the time are from the northeast (NE). Fig. 1 shows the site location map.

Instrument Details

Continuous, near real time measurement of $PM_{2.5}$ BC aerosol mass concentration was done at a flow rate of 2 LPM at an average time interval of one minute throughout the month of December 2014 using a next generation aethalometer (AE-33-7) with patented dual spot technology (aerosol make). The operating principle of the aethalometer is based on the measurement of optical attenuation of a light beam (at seven wavelengths: 370, 470, 520, 590, 660, 880, and 950 nm) transmitted through a deposited sample collected on filter tape (Teflon-coated glass fibre). Measurements from the aethalometer are reported in terms of BC aerosol concentrations in μgm^{-3} using the attenuation coefficient recommended by the manufacturer, which converts the attenuation to BC mass concentration [26]. The next-generation aethalometer incorporates the patented dual spot technology measurement method with two additional benefits: elimination of the change in instrumental response due to loading effect, and a real-time calculation

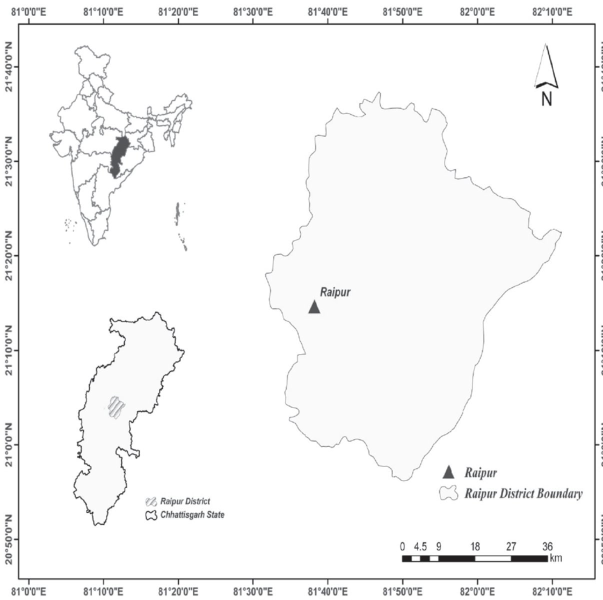

Fig. 1. Site Location.

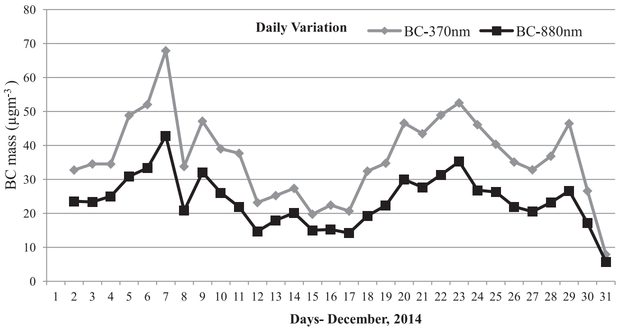

Fig. 2. Daily hourly average BC mass concentrations during the study period.

of loading effect compensation parameter [27] providing additional information about the aerosols, like aerosol origin and age.

Results and Discussion

Ground-Based BC_{370nm} and BC_{880nm} Mass Concentrations

Hourly variations of BC_{370nm} and BC_{880nm} mass concentrations during December 2014 at Raipur, India, are shown in Fig. 2. Daily mean BC_{880nm} mass concentration during the study period was 23.03±7.50 µgm^{-3}, varying within the range of 5.76 (31 December 2014) and 35.25 µgm^{-3} (23 December 2014). Hourly average BC_{880nm} mass concentration varied within the range of 2.55 (14:00 IST on 31 December 2014) and 81.81µgm^{-3} (07:00 IST on 23 December 2014). Apart from other reasons, the variations in the concentration of BC_{880nm} for the above-mentioned days may be due mainly to meteorological parameters (discussed in a later section). The ratio between nighttime (18:00-06:00 IST) and daytime (06:00-18:00 IST) BC_{880nm} concentrations was 1.55 during the study period. Monthly daytime average was 17.40 µgm^{-3}, and nighttime average was 26.99 µgm^{-3}, clearly indicating the impact of a stable shallow boundary layer and emission pattern. Tiwari et al. [28] have reported similar value (1.53) at Delhi during the winter. Daily mean BC_{370nm} mass concentration during the study period was 35.51±10.93 µgm^{-3}, with variation range of 7.91 to 52.54 µgm^{-3}. Hourly average BC_{370nm} mass concentration varied within the range of 3.24 (14:00 IST on 31 December 2014) and 154.64 µgm^{-3} (around 11:30 IST on 25 December 2014). The highest hourly average $BC_{370\ nm}$ concentration was observed on the night of 25 December, indicating biomass burning as the dominant cause.

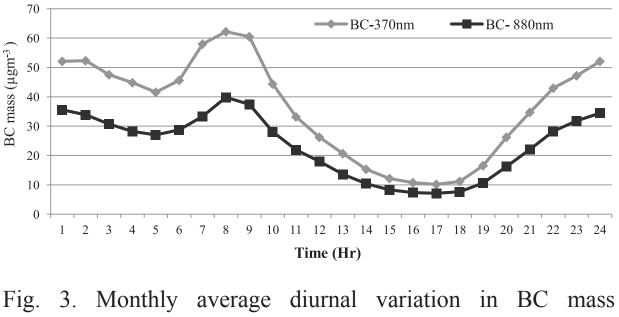

Fig. 3. Monthly average diurnal variation in BC mass concentrations during the study period.

Diurnal Variation Of Bc Mass Concentration

Monthly average of BC_{370nm} and BC_{880nm} diurnal variation is shown in Fig. 3, with two distinct BC_{880nm} peaks, one in the morning with 39.84 µgm^{-3} (07:00-08:00 IST) and the other at late night hours with 35.55 µgm^{-3} (00:00-01:00 IST); and two distinct valleys with lowest BC concentration (7.11 µgm^{-3}) during the late afternoon (15:30-16:30 IST), and the other during early morning (03:30-04:30 IST), with BC mass concentration of

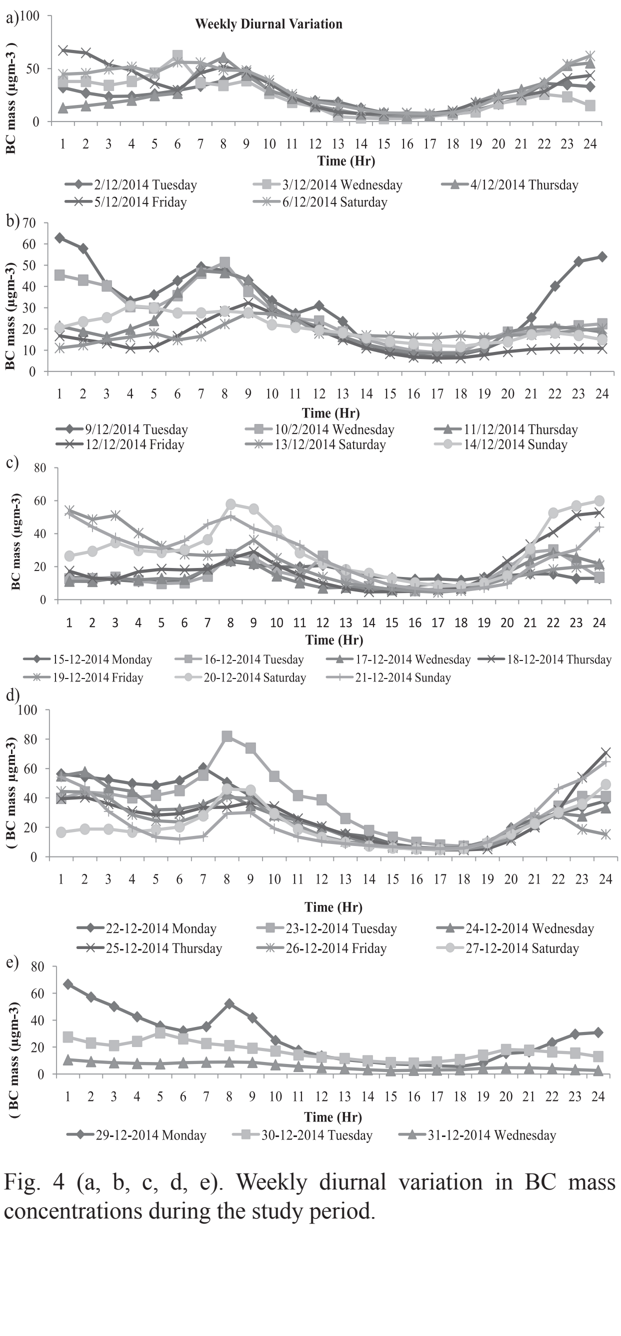

Fig. 4 (a, b, c, d, e). Weekly diurnal variation in BC mass concentrations during the study period.

27.04 μgm^{-3} – consistent with the nighttime average during the study period. Such significant peaks in mornings may be due to early-morning anthropogenic activities, breaking off the nocturnal boundary layer and convection turbulence after sunrise [29]; and at night due to the formation of a stable shallow boundary layer due to thermal inversion after sunset. Lower values during noontime throughout the month of December indicate the impact of mixed layer depth and atmospheric ventilation. The diurnal variations of BC in winter days in the present study are similar to those observed by others at different locations in some Indian cities during winter, like Hyderabad [30] and Dhanbad [31].

Weekday and Weekend Variation

Fig. 4 shows weekly diurnal variation of BC mass concentrations during the study period, depicting less variation in the month of December except for the final week. During 12-17 December, we observed less daily average BC concentrations (which may be due to prevailing meteorological conditions), and we also observed low BC mass concentrations on 31 December 2014 (discussed in a later section).

Daytime and Nighttime Averages

We analyzed available data for daytime and nighttime BC concentrations and found the ratio between nighttime (18:00-06:00 IST) and daytime (06:00-18:00 IST) of BC concentrations as 1.55 during the study period, clearly indicating the impact of the stable shallow boundary layer and emission pattern. Aruna et al. [32] found about two times higher nighttime value than daytime at Chennai. As per available literature, some cities also have high BC mass loading during nighttime: Dhanbad [31] and Agra [33].

Comparison with Other Studies

Ambient air aerosol variability depends upon the atmospheric conditions, the location of sources, and prevailing meteorological parameters. During the past two decades many studies have been carried out to monitor BC mass concentration over the Indian region and other parts of the world. The mean BC mass concentration during winter at Raipur is compared against values reported for several other stations (Table 1). Kolkata [34] and Delhi [35] have been reported to have higher

Table 1. Winter BC mass concentrations in various cities.

Location	Period	Winter BC mass (μgm^{-3})	Author
Hyderabad, India	2010-12	13.12	Jose et al. [30]
Darjeeling, India	Jan 2000-Dec 2011	3.90	Sarkar et al. [39]
Agartala, India	Sep 2010-Sep 2012	17.80	Guha et al. [21]
Dhanbad, India	1 Jan-31 Dec 2012	8.20	Singh et al.[31]
Kolkata, India	Dec 2009	36.00	Pani and Verma [34]
Nagpur, India	2011-12	4.57	Kompalli et al. [22]
Dibrugarh, India	June 2008-March 2012	13.20	Pathak et al. [37]
Bhuvaneshwar, India	Jan 2010-May2012	5.60	Mahapatra et al. [41]
Pune, India	2005-10	3.58	Safai et al. [19]
Delhi, India	1 Jan to 31 Dec 2011	17.90	Tiwari et al. [28]
Chennai, India	March 2011-Aug-2012	11.19	Aruna et al. [32]
Anantpur, India	Dec 2010	5.05	Reddy et al. [40]
Ahemdabad, India	Dec 2008	14.00	Ramchandran and Kedia [38]
Visakhapatnam, India	Dec 2005-Sep 2006	8.01	Sreekanth et al.[20]
Trivandrum, India	Jan 2000-Dec 2003	5.68	Moorthy et al. [17]
Agra, India	Dec 2004	20.60	Safai et al. [33]
Nainital, India	Dec 2004	1.36	Pant et al. [42]
Delhi, India	Dec 2004	29.40	Ganguli et al. [35]
Beijing, China	Jan 2014	6.10	Wang et al. [14]
Shenzhen, China	Fall and winter 2009-2010	4.10	Huang et al. [44]
Xian, China	Dec 2004	20.30	Cao et al. [36]

BC mass concentration as compared with the present study. The reason being obvious multifold differences in population size and spread, urbanization, and industrial activities of these cities compared to Raipur. Several other locations – including Agra [33] and Xian, China [36] – were found to have quite equal and comparable BC mass concentrations to Raipur during winter months. At Hyderabad [30], Dibrugarh [37], and Ahemdabad [38], the BC concentrations were sufficiently lower than the values reported in Raipur. For some places, like Darjeeling [39], Anantpur [40], and Bhuvaneshwar [41], BC mass concentrations were about 5-6 times lower than the value reported at the present location. High-altitude (naturally pristine) regions like Nainital [42] were reported to have about 90% lower concentrations. Looking to the large-scale spatial variations in BC concentration, a large BC dataset with spatio-temporal distribution should be available before developing any model to predict BC concentration for any location.

Relationship with Meteorological Parameters

In this study we tried to investigate the influence of meteorological parameters over BC mass variation in December, when prevailing meteorology plays an important role in surface movement and vertical dispersion of pollutants. Local meteorological parameters such as air temperature, relative humidity, visibility, and wind speed and direction were obtained from the archives at Weather Underground (wunderground.com). Statistical analysis of available data was performed for finding the correlation between BC mass concentration and meteorological parameters for the study region and duration. We found negative correlation between BC mass concentration and wind Speed (-0.75), relative humidity (-0.30), and temperature (-0.19). Tiwari et al. [28] reported a clear inverse relationship between BC and wind speed, visibility, and temperature at Delhi. Other locations also have an inverse relationship among BC mass and temperature, including Bhuvaneshwar [41], BC mass and rainfall at Pune [19], and BC mass and wind speed at Anantpur [43]. Other parts of the world, like Shenzen, China [44], found inverse correlation between BC and wind speed, and at Xian, China [36], a clear inverse relationship between BC mass and wind speed and temperature was reported. Fig. 5 shows the daily mean BC variation with meteorological conditions prevailing during the study period. The lower value (14:00 IST on 31 December 2014) may be due to high wind speed, wind direction, and rainfall/showers during the period, and the higher value of 81.81 μgm^{-3} (07:00 IST on 23 December 2014) may be due to the fumigation effect after sunrise, breaking of the nocturnal boundary layer, and dispersion of pollutants trapped during nighttime inversion conditions [29]. The third week of December saw a lot of variation in meteorological parameters such as relative humidity, wind speed, and wind direction due to this irregular diurnal pattern in BC mass concentrations.

Identification of Possible Sources

Long-Range Transport: Back-Trajectory Analysis Based on the NOAA-HYSPLIT Model

To examine the long-range transport of BC to the sampling site, five day air mass back trajectory ending at 50 m, 500 m, and 2,000 m above ground level were carried out for December over Raipur based on the NOAA-HYSPLIT (hybrid single-particle lagrangian integrated trajectory) model [45-46]. Analysis of back-trajectory for different weeks of December 2014 revealed that most of the back trajectory at surface level (50, 500 m) originated from northern and northeastern India and at higher altitude (2,000 m) from Middle Eastern and Arab countries, except during the last week of December, when back trajectory was shown to be from the Indian Ocean (Fig. 6). Sarkar et al. [39] also found that air mass trajectory arriving at 500 m a.s.l at Darjeeling originated from Middle Eastern countries and passed over the Indo-Gangetic Plain.

Wavelength Dependence of BC: BC_{370nm} and $BC_{880\ nm}$

The aethalometer (AE-33-7) is capable of analyzing aerosol absorption at seven different optical wavelengths (370-950 nm). Several studies have been performed to identify biomass burning by spectral dependence of light absorption by BC aerosol [47-48]. Organic components present in wood smoke absorb light at 370 nm more effectively than 880 nm, thus BC_{370nm} may be used for the identification of UV-absorbing organic species (wood smoke marker) emitted during biomass burning.

In the present study, the ratio between BC mass measured at 370 nm and 880 nm is consistently more than unity (1.31-1.75) during the study period, indicating that UV-absorbing organic components were consistently higher than the BC mass measured at 880 nm. Sarkar et al. [39] found a comparable ratio (1.11-1.33) between BC_{370nm} and BC_{880nm} in winter at Darjeeling during January 2010-December 2011. Tiwari et al. [28] also found a higher $BC_{370\ nm}/BC_{880\ nm}$ ratio during the post-monsoon and winter of 2011 at Delhi, indicating the enhancement of biomass burning. Wang et al. [49] identified residential wood

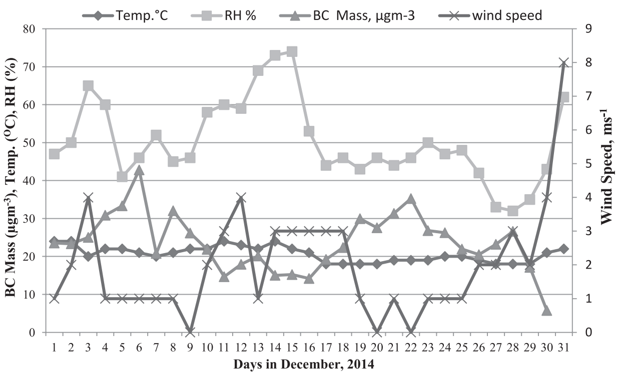

Fig. 5. BC mass variation versus meteorological factors.

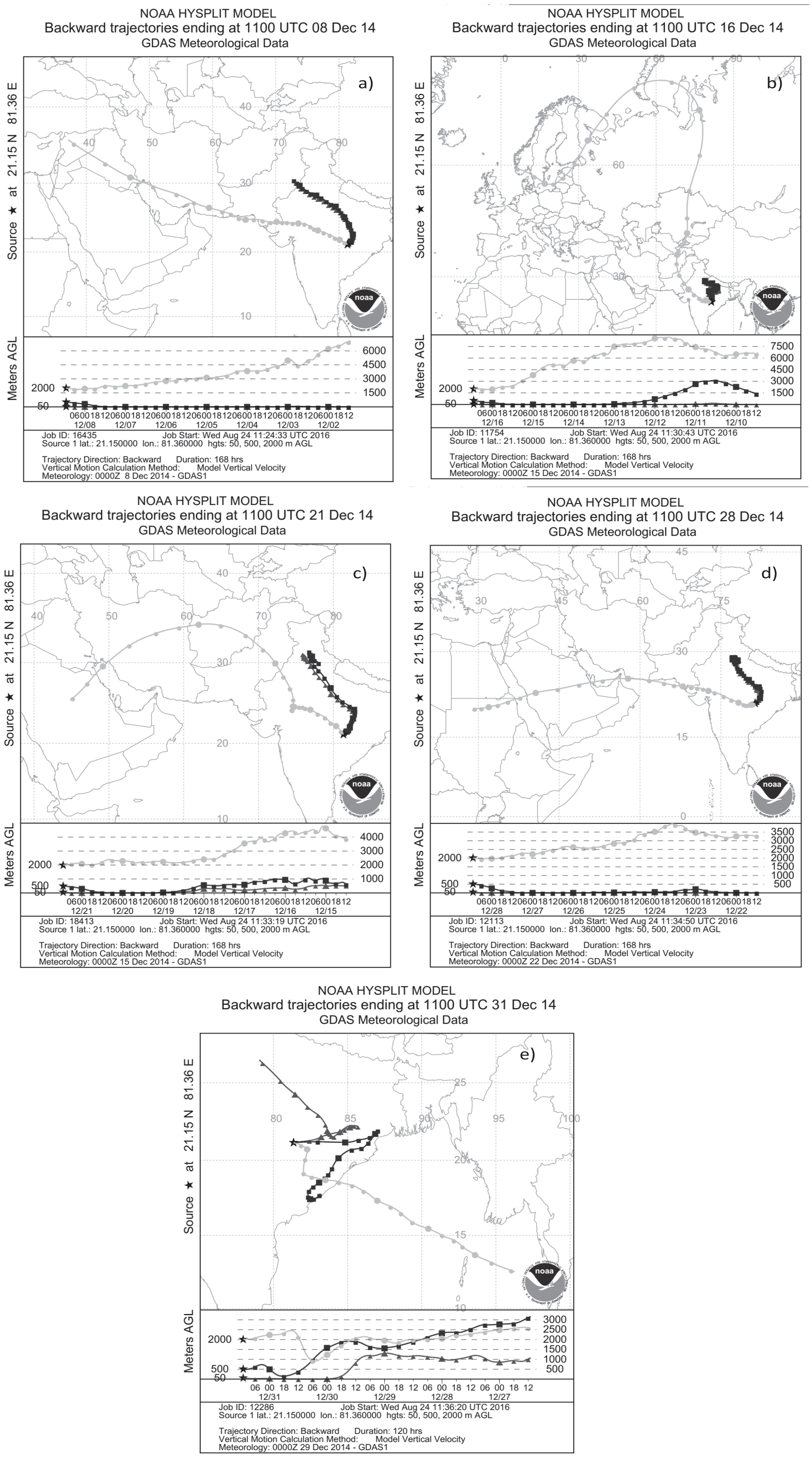

Fig. 6 (a, b, c, d, e). Five-day air mass back trajectories (NOAA-HYSPLIT) ending at 50 m, 500 m, and 2,000 m above ground level at Raipur.

combustion by calculating differences between BC_{370nm} and BC_{880nm} (Delta-C (indicator of wood combustion) = BC_{370nm}-BC_{880nm}) and found increased values of Delta-C during winter and in nighttime in Rochester, New York. We also found enhanced Delta-C values throughout the study period, and about 1.72 times higher diurnal averaged Delta-C value in nighttime (19:00-07:00 IST) than daytime.

Conclusion

BC concentrations in $PM_{2.5}$ aerosol were measured over the urban-industrial city of Raipur (289.0 m a.m.s.l) in east-central India during December 2014 (1-31 December). Some important conclusions and findings of the present study are:

– Hourly average BC_{880nm} mass concentrations varied within the range of 2.55 (14:00 IST on 31 December 2014) and 81.81 μgm^{-3} (07:00 IST on 23 December 2014).
– Daily mean BC_{880nm} mass concentration during the study period was 23.03±7.50 μgm^{-3}, varying within the range of 5.76 (31 December 2014) and 35.25 μgm^{-3} (23 December 2014).
– The nighttime and daytime ratios of BC mass concentration were found to be 1.55 during the study period, clearly indicating the impact of prevailing meteorological conditions during the winter months.
– Monthly average diurnal variation showed two distinct peaks, one at morning with 39.84 μgm^{-3} (07:00-08:00 IST), and the other at night with 35.55 μgm^{-3} (00:00-01:00 IST); and two distinct valleys, with lowest BC concentration (7.11 μgm^{-3}) during late afternoon (15:30-16:30 IST), and the other during early morning (03:30-04:30 IST).
– We found negative correlation between BC mass concentration and wind speed (-0.75), relative humidity (-0.30), and temperature (-0.19).
– Three major regions – Middle Eastern countries, Arab countries, and the Indian Ocean – were identified for long-range BC mass transport at higher altitude; and at the surface level, the back trajectory mostly originated from the northern northeastern parts of India.
– The ratio and difference between BC mass measured at 370 nm and 880 nm showed biomass burning during the nighttime to be the dominant cause.
– BC mitigations during winter/worst season may be possible by educating and facilitating the population to shift to alternative clean fuels and to avoid uncontrolled open biomass burning.

Acknowledgements

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