Measurement of Black Carbon in Delhi: Evidences of Regional Transport, Meteorology and Local Sources for Pollution Episodes

A. Malik1,2, S.G. Aggarwal1,2,*, S. Ohata3,4, T. Mori5,6, Y. Kondo7, P.R. Sinha8, P. Patel1,2, B. Kumar1,2 K. Singh1, D. Soni1,2, and M. Koike6

1CSIR-National Physical Laboratory, New Delhi, India
2Academy of Scientific and Innovative Research (AcSIR), Ghaziabad, India
3Institute for Space–Earth Environmental Research, Nagoya University, Nagoya, Japan
4Institute for Advanced Research, Nagoya University, Nagoya, Japan
5Department of Applied Chemistry, Faculty of Science and Technology, Keio University, Yokohama, Japan
6Department of Earth and Planetary Science, Graduate School of Science, the University of Tokyo, Tokyo, Japan
7National Institute of Polar Research, Tokyo, Japan
8Department of Earth and Space Sciences, Indian Institute of Space Science and Technology, Thiruvananthapuram, India

*Corresponding Author. Tel: 91 11 45608331
E-mail: aggarwalsg@nplindia.org
Abstract

Measurement of particulate matter (PM) constituent such as black carbon (BC) over urban sites is critically important owing to its adverse health and climate impacts. However, the impacts associated with BC are poorly understood primarily because of the scarcity and uncertainties of measurements of BC. Here, we present BC measurement at an urban site of Delhi using a characterized continuous soot monitoring system (COSMOS) for a year-long period, i.e. from September, 2019 to August, 2020. This measurement period covers events, i.e. period of crop residue burnings from nearby states, festive events, e.g. Diwali and New Year, and first Covid-19 lockdown period. Effects of these events combining with local emissions and meteorological conditions on BC mass concentration ($M_{BC}$) are investigated to find the possible cause of severe pollution levels in Delhi. Mean $M_{BC}$ for the complete observation period was found to be 5.02 ± 4.40 µg m$^{-3}$. $M_{BC}$ showed significant seasonal as well diurnal variations. Winter season (December to February) is observed to be the most polluted season owing to increased local emissions and non-favourable meteorological conditions. Regional emission from crop burning in nearby states during October and November is the main contributing factor for increased pollution in this post-monsoon season. Furthermore, analysis reveals that cracker burning during festivals can also be considered as contributing factor to high $M_{BC}$ for a short period in post-monsoon and winter seasons. Significant decrease in $M_{BC}$ due to Covid-19 lockdown is also observed. $M_{BC}$ in summer and monsoon are lower as compared to other seasons but are still higher than mean $M_{BC}$ levels in several other urban cities of different countries. Also, the BC data obtained from nearby sites and Modern-Era Retrospective analysis for Research and Applications - version 2 (MERRA-2)”s surface black carbon (SBC) are compared against the $M_{BC}$ to evaluate coherency among the different datasets, and discussed in detail.
1. INTRODUCTION

Increasing air pollution is one of the major challenges of the recent time in most of the metropolitan cities across the globe. Air pollution has become a serious health issue, especially in densely populated countries like India. The global burden of disease study in 2017 (Balakrishnan et al., 2019), which was focused on analyzing the health effects of ambient air pollution in 195 countries found that 1.24 million (1.09 – 1.39 million) deaths in India in 2017 were attributable to air pollution. Another recent review study based on 59 studies (Rajak and Chattopadhyay, 2020) reported that long-term exposure to pollution can be directly attributed to life-threatening conditions like asthma and heart attack.

According to a recent report on air quality (IQAir, 2020), out of 30 most polluted cities in the world, 22 are from India, and Delhi is rated the most polluted capital city in the world. Previous report has also found Delhi to be among the most polluted cities in the world (IQAir, 2019). Besides being the nation’s capital, Delhi also harbors approximately 19 million people (16.8 million according to 2011 census), which further makes it an area of grave concern. Therefore, there is a long ongoing debate on causes and sources of high pollution levels in Delhi. Previous studies (Bikkina et al., 2019; Kulkarni et al., 2020; Liu et al., 2018) suggest that crop residual burning in nearby states may be responsible for increased pollution levels. In addition to this, local emissions and meteorological conditions are also considered the other contributing factors for Delhi pollution.
Delhi also receives long-range transported dust from Middle East and Thar deserts in summer season (Kumar et al., 2016).

Black carbon (BC) is an important air pollutant that forms a substantial part of PM mass and has its own spectrum of adverse health and climatic effects (Bond et al., 2013; Mori et al., 2020). Apart from undifferentiated PM mass, BC is considered as an additional standalone indicator of harmful particulate substance from combustion sources (Janssen et al., 2011). Global carbon project report (Friedlingstein et al., 2019) and various emission inventories (Bond et al., 2007; Klimont et al., 2017; Paliwal et al., 2016; Wang et al., 2012) have found India to be the second major contributor (after China) to global BC aerosol. However, these inventories contain large uncertainties in emission estimates of BC and there is a large discrepancy in BC emission estimates studies from India (Rana et al., 2019). Therefore, ground-based observations of atmospheric BC are of utmost importance and a preferable source for measuring changes in emission estimates and investigating health and climatic implications of BC.

In India, openly accessible data of BC mass concentration ($M_{BC}$) is still limitedly available and researchers have to rely on independent observations. A recent review summarized the $M_{BC}$ measurements in India based on the survey of 140 studies conducted in India spanning for a period from 2002 to 2018 (Rana et al., 2019). It was found that long-term data on $M_{BC}$ is scarce in India which makes it difficult to construct trend over the years and derive some meaningful information. Few multi-year (3-5 year) studies have been conducted across the country (Gogoi et al., 2013; Jose et al., 2017; Kumar et al., 2020; Sharma et al., 2017; Talukdar et al., 2015). Similarly, despite high relevance of long-term $M_{BC}$ measurement studies in Delhi, very limited BC studies have been conducted in the previous decade and fewer are based on long-term (at least for 1 year) real-time data (Bisht et al., 2015; Kumar et al., 2020; Surendran et al., 2013; Tiwari et al., 2013; Tyagi et al., 2020). Two of these studies were primarily focussed on realizing climatic effects of BC over Delhi.
(Bisht et al., 2015; Surendran et al., 2013). Kumar et al. (2020) evaluated the spatial and temporal heterogeneity of BC aerosol over India using three-year measurements from Indian Meteorological Department (IMD) BC observation network (one of the 15 observation sites, which is located in Delhi). Other two studies (Tiwari et al., 2013; Tyagi et al., 2020) were focussed on characterisation and comprising possible sources of BC aerosol in Delhi.

A recent review (Malik and Aggarwal, 2021) summarized the measurements technique used for real-time online measurement of BC levels in India and China. In India, most (>95%) of the previous studies (where \( M_{BC} \) was measured using light absorption technique and not the evolved carbon technique) have made use of different versions of widely used instrument, i.e. Aethalometer (Malik and Aggarwal, 2021; Rana et al., 2019). Multi Angle Absorption Photometer (MAAP) have been used in a single study (Hyvärinen et al., 2010). The uncertainties of the \( M_{BC} \) measurements done in India by these instruments have not been evaluated quantitatively.

Considering this, we made measurements of \( M_{BC} \) using Continuous Soot Monitoring System (COSMOS) for the first time in India. The accuracy of \( M_{BC} \) measured by COSMOS is about 15%, as discussed in detail in following section. Since this is the first measurement in Delhi using COMSOS, comparison between \( M_{BC} \) and simultaneous BC measurement data obtained from nearby location (using Aethalometer, denoted as “\( M_{BC} \) (AE)” hereafter) is also performed. Also, Modern-Era Retrospective analysis for Research and Applications - version 2 (MERRA-2)’s surface black carbon (SBC) concentrations are compared with \( M_{BC} \). MERRA-2’s reanalysis data (SBC) have been used previously to define the spatial distribution and trends of BC and other pollutants in India and worldwide (Bali et al., 2017; Evangeliou et al., 2021; Navinya et al., 2020; Qin et al., 2019; Rana et al., 2019; Sitnov et al., 2020). Also, SBC data have been checked and evaluated against ground observations of BC in previous studies conducted outside India (Qin et al., 2019; Xu et al., 2020). Malik and Aggarwal (2021) compared MERRA-2’s SBC data with the
ground observation’s BC data published in previous peer reviewed studies conducted in India and
China. However, a direct comparison between highly temporarily resolved datasets (as done by
Qin et al. (2019) in Beijing) of SBC and ground observation has not been done in India.
In this study, we report $M_{BC}$ measurements from September 2019 to August 2020 in Delhi, which
can be recognised as a representative of eventful period. Measurement period spans various events
like crop-residue burning in the months of October and November 2019 and a nation-wide
lockdown due to Covid-19 pandemic in the months of March and April 2020. Furthermore,
observation period covers major festive events (Diwali and New Year) which can be responsible
for sudden peak in pollution concentration. Therefore, in this study, possible effects of these events
and local meteorological conditions on the seasonal variations of $M_{BC}$ are realized. Table 1
summarizes the major events during measurement period and their effects on $M_{BC}$. Based on the
data analysis, we have tried to find the possible causes of severe air pollution in Delhi during the
observation period. Information discussed here can be useful for better understanding of the Delhi’s
increasing air pollution and accordingly formulation of control strategy of carbonaceous aerosol.

2. METHODOLOGY

2.1 Site Description and Measurement Period
Black carbon ($M_{BC}$) measurement by COSMOS was made on the premises of the CSIR-National
Physical Laboratory (NPL) located in New Delhi, India, (Fig. 1, 28.63 °N and 77.17°E). NPL is
surrounded by thinly populated Indian Agricultural Research Institute (IARI, PUSA institute) on
three sides and a traffic (major rush hours are in morning and evening) road and residential area on
one side. There is no major industry in nearby area which can affect local pollution levels. New
Delhi, the capital of India, forms a small part of Delhi metropolitan area. Delhi is located in Indo-
Gigantic Plain region spanning an area of 1,484 km². It is a landlocked city with the state of Haryana bordering it on three sides and Uttar Pradesh covering it from the fourth, i.e. East side. South-west monsoon is the most important feature controlling the Indian climate because about 75% of the annual rainfall is received during a short span of four months (June to September). So significant is the monsoon season to the Indian climate that the remaining seasons are often referred relative to the monsoon. According to the classification given by Indian Meteorological Department (Attri and Tyagi, 2010), India experiences four distinct seasons, i.e. winter, pre-monsoon, monsoon and post-monsoon. Monthly time span of each season is slightly different in different regions of country depending upon the arrival of monsoon and temperature variation. Year-round Mєc measurements were performed from September 2019 to August 2020 to cover all seasons in Delhi.

Fig. 2 shows monthly variation of meteorological parameters, i.e. ambient temperature (AT), relative humidity (RH), solar radiation (SR) and precipitation during measurement period. 24-Hourly averaged datasets of all parameters are taken from Central Pollution Control Board’s (CPCB) Central Control Room for Air Quality Management website (https://app.cpcbcr.com). AT, RH and SR datasets of two monitoring station (Pusa-DPCC, Shadipur-CPCB) situated within 1.5 km radius of monitoring site are used to plot monthly variation. Precipitation data of monitoring stations, which are located near the sampling site were not available. Therefore, average of precipitation data of all monitoring stations (Alipur-DPCC, DTU-CPCB, Sirifort-CPCB) situated in Delhi are used. Ambient temperature was minimum during the months of December – February. Precipitation levels were maximum in the months of June-August. Therefore, seasons in Delhi are classified in general as – winter (December - February), pre-monsoon/summer (March - May), monsoon (June - August), and post-monsoon (September - November).

2.2 Instrumentation
Details of the COSMOS are given in previous papers (Kondo et al., 2011; Miyazaki et al., 2008; Ohata et al., 2021, 2019). In brief, the COSMOS measures the extinction coefficient ($b_0$) of aerosols collected on a quartz-fiber filter at a given wavelength ($\lambda = 565$ nm). High-efficiency particulate matter (HEPA) filters rolls were used in COSMOS for particle collection in this study. The inlet of the COSMOS is heated to 300°C to remove volatile light scattering particles and coating on BC particles. This excludes the effect on $b_0$ of co-existing volatile components externally or internally mixed with BC particles. The COSMOS is equipped with a PM$_1$ cyclone to minimize the effect in coarse mode of refractory non-BC particles, such as dust particles. Therefore, the absorption coefficient for the COSMOS is given as

$$b_{abs} (\text{COSMOS}) = f_{fil} b_0$$  \hspace{1cm} (1)$$

where, $f_{fil}$ is a factor used to correct for the increase of absorption caused by multiple scattering in the filter medium (Bond et al., 1999; Ogren, 2010; Ohata et al., 2019). The mass absorption cross section (MAC) for the COSMOS ($\text{m}^2$ $\text{g}^{-1}$) has been determined as

$$\text{MAC (COSMOS, SP2)} \equiv \frac{b_{abs} (\text{COSMOS})}{M_{BC} (\text{SP2})}$$  \hspace{1cm} (2)$$

where, the numerator and denominator, respectively, are simultaneous measurements of $b_{abs}$ ($\text{Mm}^{-1}$) by COSMOS and $M_{BC}$ ($\text{mg m}^{-3}$) by a single particle soot photometer (SP2) for ambient air. SP2 is based on laser induced incandescence technique (Malik and Aggarwal, 2021) and measures $M_{BC}$ with an accuracy of about 10% (Kondo et al., 2011; Moteki and Kondo, 2010; Ohata et al., 2021). The MAC value for a HEPA filter at $\lambda = 565$ nm is about 9.25 ($\text{m}^2$ $\text{g}^{-1}$) (Irwin et al., 2015). $M_{BC}$ ($\text{COSMOS}$) ($\text{g m}^{-3}$) at standard temperature (0°C) and pressure (101 kPa) can be estimated as
\[ M_{BC}^\text{COSMOS} = \frac{b_{abs}^\text{COSMOS}}{\text{MAC}^\text{COSMOS,SP2}} \] (3)

A “Standard COSMOS (Std-COSMOS)” was calibrated by comparison with the SP2 in Tokyo. Because the MAC of the Std-COSMOS was determined by comparison with SP2 (Eq. (2)), it acts as a transfer standard for the SP2. The \( b_{abs} \) (COSMOS) of each COSMOS manufactured is compared with the Std-COSMOS by sampling ambient BC particles in Osaka, Japan, typically for 1–2 weeks. The temperatures of the optical detector unit and filter holding unit of COSMOS are actively maintained at 50°C to minimize any possible effects of changes in ambient relative humidity (Kondo et al., 2009; Miyazaki et al., 2008). \( M_{BC} \) (COSMOS) was also compared with \( M_{BC} \) (SP2) at Hedo, located at downstream of the East Asian continent and at Ny-Ålesund in the Arctic (Ohata et al., 2019). \( M_{BC} \) (COSMOS) and \( M_{BC} \) (SP2) agree to within about 10% at these sites, thus demonstrating the validity of using the Std-COSMOS to calibrate each of the COSMOS instruments to be used for field observations. It will be appropriate to estimate the absolute accuracy of \( M_{BC} \) (COSMOS) to be about 15%, including the 10% uncertainty of \( M_{BC} \) (SP2). This 15% uncertainty also covers the range of agreement between \( M_{BC} \) (COSMOS) and \( M_{BC} \) (SP2) previously reported by other groups at Ny-Ålesund (Zanatta et al., 2018) and at Fukue, located downstream of the East Asian continent (Miyakawa et al., 2017). Hereafter we denote \( M_{BC} \) to represent \( M_{BC} \) (COSMOS) for simplicity.

### 2.3 Additional Data Set

Black carbon data of a nearby monitoring station (station name “PUSA, Delhi-IMD”, location denoted as PUSA in Fig. 1(b)), available at CPCB’s website (ecpcbccr.com) are used for comparison with \( M_{BC} \) measured at NPL in this study. Mass concentrations of black carbon at monitoring station of PUSA are measured using Aethalometer (Hansen, 2005; Sedlacek, 2016) with PM\(_{2.5}\) cutoff size...
inlet. Absorption coefficient measured by Aethalometer (AE) at a wavelength of 880 nm was
converted to BC mass concentration $M_{BC}$ (AE) by assuming a mass absorption cross section (MAC)
of 7.7 m$^2$ g$^{-1}$. It is important to note that information about Aethalometer data, i.e. whether the data
are at STP as it is done for COSMOS is unknown.

NASA’s MERRA-2 reanalysis data (SBC) over Delhi available through publicly accessible
website (https://disc.gsfc.nasa.gov/datasets?project=MERRA-2) are also used for the comparison
with $M_{BC}$. MERRA-2 is the latest long-term global atmospheric reanalysis to assimilate space-
based observations of aerosols and represent their interactions with other physical processes in
the climate system initiated by NASA’s Global Modeling and Assimilation Office (GMAO).

Details about MERRA-2 dataset can be accessed from previous studies (Bali et al., 2017;
Bosilovich et al., 2016; Buchard et al., 2015; Kuo, 2017). Different type of aerosols (including
BC) is simulated using the Goddard Chemistry Aerosol Radiation and Transport module in
GEOS-5 (Bali et al., 2017; Buchard et al., 2015). Surface black carbon data obtained from
MERRA-2 of coordinates 28.5°N and 77.25°E, which are closest to measurement site’s
coordinates, i.e. 28.63°N and 77.17°E are used for comparison.

Hourly data of boundary layer height (BLH) was obtained from the climate data store

Ventilation coefficient (VC) is an important parameter to define the dispersion of a pollutant at a
place. It is calculated as the product of average BLH and average wind speed (WS) over a given
area. The spatial resolution of gridded BLH data source is 0.25° × 0.25° (approximately 30 km$^2$).
Therefore, hourly averaged WS data of 28 monitoring stations in Delhi is used to cover the area
spanned by BLH data grid. This monitoring station network is jointly operated by the Central
Pollution Control Board (CPCB), Delhi State Pollution Control Committee (DPCC), the Indian
Institute of Tropical Meteorology (IITM), and the IMD. Open source archived fire map data from
NASA’s Fire Information for Resource Management (FIRMS, Data source-VIIRS) website is utilized for fire spots analysis. Furthermore, Global Data Assimilation System (GDAS) meteorological data files (1° × 1° resolution) are used to calculate the air mass back trajectories.

3. RESULTS AND DISCUSSION

3.1 Data Comparisons

3.1.1 Comparison between $M_{BC}$ and BC concentration of nearby monitoring station

Fig. 3(a) shows the scatter plot of 24-hour averaged $M_{BC}$ (AE) at PUSA site (Fig.1) against $M_{BC}$. $M_{BC}$ (AE) data correlate very well with the $M_{BC}$, where correlation coefficients ($r$) increase with the increase in averaging time period. For hourly, daily, and monthly averaged data, $r$ values are 0.81, 0.89, and 0.96, respectively. Although $M_{BC}$ at NPL site correlate well with the $M_{BC}$ (AE) of PUSA site, there is a bias between both the datasets. Furthermore, the bias between two data set is larger at higher concentrations. Slope of the linearly fitted line for $M_{BC} < 8 \mu g m^{-3}$ is 0.88 which is closer to 1 than that for all data points (0.67). This might be due to systematic bias between different instruments used (Aethalometer at PUSA and COSMOS at NPL) for deriving BC concentrations.

Ohata et al. (2021) made direct comparison of COSMOS and Aethalometer in the Arctic and determined MAC there. However, no such comparisons have been made, especially for urban site and MAC of Aethalometer is poorly characterized. Because, it is difficult to estimate the uncertainties of $M_{BC}$ (AE) at PUSA site, the comparison of Aethalometer with COSMOS is rather qualitative this time. We used currently available $M_{BC}$ (AE), although direct and systematic comparisons as done elsewhere (Ohata et al., 2021) are important and needed, especially under high BC mass loading condition (urban sites). Nevertheless, the good correlation between both
sites suggests that the NPL site can be considered as representative for the temporal variations of BC aerosol in Delhi area.

3.1.2 Comparison between $M_{BC}$ and SBC

The SBC data are compared with 24-averaged $M_{BC}$ for the complete observation period, i.e. September 2019 - August 2020. The correlations of the two data sets are shown in Fig. 3(b). SBC data correlate well with the $M_{BC}$, where correlation coefficients increase with increase in averaging time period. The $r$ values for hourly, daily, and monthly averaged data are 0.57, 0.79, and 0.93, respectively.

Mean SBC concentration for the complete observation period is 3.07 µg m$^{-3}$ which is about 60% of mean $M_{BC}$ (5.02 µg m$^{-3}$). Also, the scatter plot of 24-hour averaged SBC against 24-hour averaged $M_{BC}$ (Fig. 3(b)) shows that MERRA-2 underestimates the BC concentration, whereas the correlation between these data is fairly good, i.e. $r = 0.79$. As shown in Fig 3(a), the slope of the linearly fitted line of data between $M_{BC}$ (AE) and $M_{BC} < 8$ µg m$^{-3}$ is better (0.88) than that for all the data points (0.67), inset Fig 3(b) also suggests that the slope of linearly fitted line for $M_{BC} < 8$ µg m$^{-3}$ is 0.4, which is slightly better than that of the slope with all the data points, i.e. 0.3. This further indicates that underestimation of BC concentration by MERRA-2 is more pronounced at higher concentration. Similarly, Navinya et al. (2020) also showed that MERRA-2 generally underestimates the PM$_{2.5}$ in terms of both the mass concentration and the number of exceedance days, i.e. when 24-hour averaged air quality index (AQI) values exceeded the AQI limits of “Good” category as per national ambient air quality standards (NAAQS) of India. However, on the contrary, studies conducted in Beijing found MERRA-2 to be overestimating BC concentration (Qin et al., 2019; X. Xu et al., 2020).

One of the reasons for bias between two datasets can be the uncertainties in the emission estimates which are used as input to MERRA-2 for simulating the BC aerosol. Other reasons may be the...
difference in spatial resolution of both datasets (i.e., SBC and $M_{BC}$). The spatial resolution of
MERRA-2 data set is $0.5^\circ \times 0.625^\circ$, hence one rectangular grid (made up of 4 data points) of
MERRA-2 data set covers a large area (about $50 \times 50$ km$^2$). Therefore, it can be deduced that one
coordinate/point of MERRA-2 gridded dataset represents SBC levels roughly over an area of $15\times15$
km$^2$. On the other hand, $M_{BC}$ ideally represents BC levels measured at one site. As mentioned
earlier, SBC concentration of data point whose coordinates (28.5°N, 77.25°E) were nearest to the
NPL site are used for comparison in current study (Fig. 1(b)). The linear distance calculated
between the NPL site and MERRA-2’s data point is roughly 12 - 15 km and pollution levels at
ground can change considerably over such distance in Delhi where local pollution sources are
distributed heterogeneously.

Overall, the correlation between the MERRA-2’s data and $M_{BC}$ is moderate. A comparison
between mean $M_{BC}$ levels of several sites covering entire area under one grid of MERRA-2 dataset
would be more appropriate for further assessment and understanding the deviation between both
the data sets.

3.2 Year-Round Concentration of BC

Here, we discuss year-round concentration of $M_{BC}$ measured using COSMOS (i.e., temporal
variation in Delhi). Fig. 4 shows 1-hour averaged, 24-hour averaged and weekly averaged plots of
$M_{BC}$ measured for one-year starting from September 1, 2019 to August 31, 2020. However, data
for almost one month from April 9, 2020 to May 13, 2020 are not available due to Covid-19
lockdown. 24-Hour averaged $M_{BC}$ for the complete observation period varied from 0.7 to 29.1 µg
m$^{-3}$ with a mean value of $5.02 \pm 4.40$ µg m$^{-3}$. Table S1 summarizes short term as well as long-term
studies conducted in Delhi in the past decade (2011-2020). $M_{BC}$ observed in current study is
significantly lower than those reported by recent studies (12.73 µg m$^{-3}$ and 13.57 µg m$^{-3}$) conducted
in Delhi for the time period 2016-2018 (Kumar et al., 2020; Tyagi et al., 2020). Possible reasons
for the difference might be different study time period and use of different instruments (AE-33 in
tyagi et al. (2020) and kumar et al. (2020), and cosmos in current study), use of different PM
mass for BC measurement (PM<sub>2.5</sub> in kumar et al. (2020) and PM<sub>1</sub> in current study), and difference
in sampling sites as air pollution levels in delhi are not homogenous. However, observed
concentration is comparable to those reported by other studies conducted in the year 2011 (6.7 µg
m<sup>3</sup>) and 2011-2012 (7 µg m<sup>3</sup>) (bisht et al., 2015 and tiwari et al., 2013, respectively). It is
noteworthy that these studies were conducted at a site (IITM) which is ~ 1 km away from
measurement site of current study.

In this study, maximum value of 24-hour averaged $M_{BC}$ reached up to 29.1 µg m<sup>-3</sup> on January 2,
2020, i.e. one day after the New Year celebration. Minimum value of 24-hour averaged $M_{BC}$ went
down to 0.7 µg m<sup>-3</sup> on March 29, 2020, i.e. one week into the Covid-19 lockdown in India (Fig.
4(a)). Also, Fig. 4(b) shows several events (highlighted by colored bars) those influenced BC
concentration significantly during the observation period. These events are further discussed in the
following sections.

3.3 Diurnal Variation

For better understanding on the day- and night-time atmospheric chemistry, conditions and sources
of BC, here we discuss diurnal variation of $M_{BC}$ for the measurement period. Fig. 5 shows 1-hour
averaged time series of diurnal variation for complete observation period. Mean day-time $M_{BC}$
(0600 h to 1800 h, LT) is 3.99 ± 3.42 µg m<sup>-3</sup> which is lower as compared to night-time mean value
of 6.01 ± 5.8 µg m<sup>-3</sup> (1800 h to 0600 h). Wilcoxon rank test at significance level of 0.05 is
performed to test the significance of difference between day-time and night-time concentration.
Significantly higher $M_{BC}$ at night-time indicates a possible influence of increased emissions due to
heavy-duty diesel vehicles whose entry is allowed only after 2200 h in Delhi.
Diurnal plot (Fig. 5) shows a slight elevation in $M_{BC}$ during morning (0700 h to 0900 h) and evening (1900 h to 2100 h). Similar diurnal variation pattern has been observed in previous studies as well (Srivastava et al., 2014; Tiwari et al., 2013). Diurnal plots of boundary layer height (BLH), wind speed (WS) and ventilation coefficient (VC) over Delhi are given in Fig. S1. Diurnal variation of $M_{BC}$ during observation period showed negative correlation with diurnal variation of BLH ($r = -0.88$ for $n = 24$), WS ($r = -0.72$ for $n = 24$) and VC ($r = -0.86$ for $n = 24$) indicating the dominance of dispersion factor. Same is depicted from diurnal plots as distinctive rise in WS, BLH and VC plots during afternoon also coincide with dip in $M_{BC}$. However, during morning hours (0700 h - 0900 h), the mean $M_{BC}$ increases despite increase in dispersion parameters. This anomaly is possibly due to the effects of increased traffic on nearby road during these peak hours.

3.4 Seasonal and Monthly Variation

Year-round $M_{BC}$ data is further categorized for winter, summer, monsoon and post-monsoon seasons in Delhi. These seasonal as well as month-wise variations of $M_{BC}$ are shown using box and whisker plots in Fig. 6. The highest concentration of $M_{BC}$ is observed in winter (daily mean = 8.9 ± 4.94 μg m$^{-3}$) followed by post-monsoon (daily mean = 5.72 ± 3.96 μg m$^{-3}$), whereas monsoon is the least polluted among all seasons (daily mean = 1.76 ± 0.63 μg m$^{-3}$). $M_{BC}$ in summer and monsoon are significantly lower than post-monsoon and winter and comparable to other Asian urban cities (Chen et al., 2016; Kumar et al., 2020; Sahu et al., 2011; Zhang et al., 2020). However, these values are still higher than the yearly mean $M_{BC}$ levels reported for some developed countries (Crilley et al., 2015; Healy et al., 2017; Mousavi et al., 2018; Saha and Despiau, 2009).

This variation (trend) in $M_{BC}$ is in accordance with previous studies listed in Table S1 (Bisht et al., 2015; Srivastava et al., 2014; Tiwari et al., 2013; Tyagi et al., 2020). Low wind speed and shallow mixed layer were found to be main reasons for high levels in winter (Tiwari et al., 2013). Furthermore, dominance of local emission in winter of 2015-2016 has been shown by Dumka et
Non-local fire emissions were reported to be contributing to about 20% of particulate matter mass in October and November (post-monsoon) (Kulkarni et al., 2020). Moreover, India celebrates a number of different festivals owing to its diverse demography, and festivals are key to the respective cultures. Diwali is one of such festivals, which is celebrated throughout the country.

While it started off as a festival of lights, the celebration practice has taken a turn for the worse in the post-industrial era with the introduction of chemical fireworks (crackers). Various previous studies have highlighted the worse effect of cracker burning on air quality during the night of Diwali (Ghei and Sane, 2018; Mukherjee et al., 2018; Rao et al., 2012). Following these approaches and findings, further analysis and observation report of seasonal and monthly variation are discussed in following subsections.

### 3.4.1 Role of local meteorology

Here, we discuss the effect of local meteorological conditions on $M_{BC}$ at measurement site. As mentioned earlier, monthly variations of 4 meteorological parameters (i.e., AT, RH, SR and precipitation) near to measurement site are shown in Fig. 2. Further, monthly variations of critical parameters (BLH, WS, VC) are shown in Fig. 7, which suggest an idea about dispersion factor over Delhi. Table 2 summarizes the dependency of 24-hour averaged as well as monthly $M_{BC}$ on meteorological parameters. 24-Hour averaged $M_{BC}$ showed moderate negative correlation (ranging from -0.47 to -0.60) with all meteorological parameters except RH. Similarly, month wise $M_{BC}$ shows strong negative correlations (ranging from -0.51 to -0.84) with all meteorological parameters except RH. There exists negligible (24-hour averaged) to low correlation (monthly) between RH and $M_{BC}$. It was reported that variation in ambient RH can affect the BC measurements of filter-based instruments as MAC value increases with increase in RH (Wu et al., 2013). The temperatures of the optical detector unit and filter holding unit of COSMOS are controlled at 50°C (section 2.2).
This, together with the heated inlet stabilizes the moisture content in the aerosol at the time of
measurement, consequently no effect of RH in measurement as such is predicted.

Strong negative correlations between month wise variation of meteorological parameters and $M_{BC}$
show the dominance of local meteorology and local sources in defining the $M_{BC}$ over Delhi. High
values of meteorological parameters in monsoon and summer favors low $M_{BC}$, whereas unfavorable
meteorological conditions (i.e., low BLH, VC and AT) during winter can be directly attributed to
high $M_{BC}$.

Furthermore, Delhi’s topography also plays an important role in enhancing the impact of
meteorology. As visible in digital elevation model map (Fig.1(c)), Delhi’s surface is distinctively
uneven with elevation above sea level varying from nearly 200 m to >300 m. Most of the landmass
has elevation in the approximate range of 190 - 230 m which is surrounded by hills (significantly
elevated (280 - 320 m) surface) at South-East and Southern part. Sometimes during winters, the
24-hour averaged BLH is observed to be as low as 100 m above ground, and in such cases pollutants
accumulate in atmosphere over Delhi. Apart from regional pollution comes when winds are North-
Westerly, for local pollution also these hills play a role of wall to trap the pollutants over Delhi. So,
during these low BLH and calm conditions, the dispersion rate becomes very low which leads to
high pollution levels.

Strongly correlated (with $M_{BC}$) meteorological parameters (AT, SR, BLH, VC) show smooth
increment from January to mid of the year (May - June) and then smooth decrement from mid-year
to December. However, monthly variation plot of $M_{BC}$ is not so smooth and there are non-linearity
and deviations as compared to plots of meteorological parameters. All three months (September,
October, and November) in post-monsoon experiences similar meteorological conditions, whereas
$M_{BC}$ are significantly higher in October and November as compared to September. Similarly, May
has more favorable meteorological conditions as compared to June and April but $M_{BC}$ is higher
than June and April. Furthermore, there is a sudden increment in $M_{BC}$ from November to months of December and January (Fig. 6(b)), whereas there is smooth decrement in meteorological parameters from November to January (Fig. 7). These anomalies are indicative of change in local as well as regional emissions due to different events which are further explained in following subsections.

3.4.2 Regional emission transport

The 48 hours air mass back trajectories arriving up to 1000 m above sampling site for the month of September and October are shown in Fig. 8. Air mass back trajectories from GDAS meteorological files are calculated using geographic information system (GIS) based software, “TrajStat”. 48-Hour back trajectories are calculated for each day (refresh time of 0600 hours). Additionally, fire spots “shapefiles” from FIRMS are loaded using QGIS for combined fire spots and air mass back trajectory analyses.

In September (mean $M_{BC} = 2.49 \pm 0.89 \ \mu g \ m^{-3}$) most of the air masses at sampling site are arrived from South-East direction where there are very few fire spots. Whereas in October (mean $M_{BC} = 7.63 \pm 3.87 \ \mu g \ m^{-3}$), most of the air masses at sampling site arrived from North-West direction where large number of fire spots are visible as compared to the month of September. Similar maps are drawn for other months (Fig. S2). Using these maps, monthly mean number of fire spots per unit area are calculated for each month using Eq. 4.

\[
F_{av} = \frac{F_t}{AKN},
\]

where, $F_{av}$ is monthly mean number of fire spots per unit area ($km^{-2}$), $F_t$ is total number of fire spots on 48 hours back trajectories area for the complete month, $A$ is the total geographical area spanned
Fig. 9(a) shows the relation between number of monthly averaged fire spots per unit area \( F_{av} \) and corresponding \( M_{BC} \). In general, \( M_{BC} \) correlates very well with \( F_{av} \) except for the months of December and January (Table 2). From Fig. 9(a), it is evident that elevated \( M_{BC} \) during the months of October and November in post-monsoon despite having comparable meteorological conditions in September (Fig. 2 and Fig. 7) is due to increased fire spots in nearby areas/states of Delhi. Similarly, spike in \( F_{av} \) during month of May is also responsible for elevation of \( M_{BC} \). However, in May, effect of fire spots is less compared to that in October and November as meteorological conditions are favorable (relatively high BLH, VC and WS) in May. In the months of May, October and November, increased fire spots are due to crop-residue burning in nearby states (mainly Haryana, Panjab and Uttar Pradesh) of Delhi (CREAMS, ICAR, 2019).

3.4.3 Increased local emissions in winter

As discussed previously, there is a sudden increment in \( M_{BC} \) from November to months of December and January (Fig. 6(b)) whereas there is smooth decrement in meteorological parameters from November to December. Also, monthly mean fire spots per unit area in winter are less as compared to months of post-monsoon season (October, November) and are comparable to monsoon and summer seasons. Delhi faces severe cold during the months of December and January with average temperature drops as low as 2 - 3°C during night-time. The daily mean temperature for the months of December and January (Fig. 2(a)) near to measurement site was 12.5°C and 13.3°C, respectively. Moreover, Delhi receives cold waves from the nearby Himalayas resulting in lower apparent temperature as an impact of wind-chill. Therefore, open burning (including waste, wood, coal and biomass) to fight severe cold in winters is a common practice followed by people in Delhi (Kumar et al., 2018). Therefore, combined effect of increased local emission due to open
burning and non-favorable meteorological conditions can be the major reason for sudden increment in $M_{BC}$ in winter.

In Fig. 9(b), Non-Parametric Wind Regression (NWR) analysis results show that in winter very high $M_{BC}$ are associated with low wind speed ($< 4$ km s$^{-1}$) showing the dominance of local sources. Whereas, in post-monsoon high $M_{BC}$ are associated with high as well as low wind-speed showing the effect of regional transport of aerosols. Non-Parametric Wind Regression (NWR) analysis is performed using ZeFir (Petit et al., 2017), which is an IGOR-based package that provides user friendly coupling between pollutant concentrations and wind data. Non-parametric Wind Regression techniques couple wind data (direction and/or speed) with pollutant concentration to alternatively highlight wind sectors that are associated with high measured concentrations (Henry et al., 2009).

Furthermore, month wise variation plot of $M_{BC}$ (Fig. 6(b)) shows that in winter, $M_{BC}$ are significantly higher in December and January as compared to February. This observation is also indicative of increased emission due to open burning by local people in comparatively cold months of December and January in comparison to February. Qualitative impact of increased local emission during cold winters has been realized previously in Delhi (Dumka et al., 2018). Effect of residential coal burning and relatively stable meteorological conditions during winters were found to be the possible reason for increment in $M_{BC}$ in Beijing as well (Chen et al., 2016; Liu et al., 2016). However, a rigorous survey-based study is required to quantify and further investigate the effect of open burning in winter.

3.4.4 Impact of festive events

Cracker-burning during Diwali has been prevalent for a long time and its impact on pollution levels in Delhi have been observed in previous studies (Sateesh et al., 2018; Singh and Srivastava, 2020). During Diwali festival in 2019, also the crackers were observed to be burnt and weekly averaged $M_{BC}$ rose significantly around this festival (peak 1 in Fig. 4(b)). Fig. 10 shows the diurnal variation
of $M_{\text{BC}}$ on the days of Diwali and New Year. Diurnal characteristics for these two days are distinguishable from that of whole year (Fig. 5), where hourly averaged $M_{\text{BC}}$ throughout night hours (1800 h to 0600 h, LT) are similar. Unusual and nearly identical sharp peak around midnight on New Year as well as Diwali are clear indicative of cracker burning on these days. Therefore, in the recent years, the practice of cracker burning has transcended beyond the day of Diwali and events like New Year’s Eve also witness a significant amount of cracker burning.

During Diwali (post-monsoon), weekly averaged $M_{\text{BC}}$ (peak 1 in Fig. 4(b)) is comparable to nearby weekly averaged $M_{\text{BC}}$, whereas during New Year (winter), weekly averaged peak (peak 2 in Fig. 4(b)) is significantly higher from nearby weekly averaged concentrations. Comparable levels of weekly averaged $M_{\text{BC}}$ during Diwali in post-monsoon indicates that pollution from local emissions gets dispersed easily and doesn’t affect $M_{\text{BC}}$ significantly during post-monsoon. However, in winter, distinctive weekly averaged $M_{\text{BC}}$ peak around New Year shows the significant role of amplified effect of local meteorology on local emissions as dispersion of local pollution becomes non-favorable in winter (as discussed above). Furthermore, the number of weddings conducted in winters are significantly higher than other seasons as Indian culture relies heavily on astrology and most auspicious days fall in winter. Therefore, celebrations by cracker burning in weddings as well may also lead to increase the emissions.

3.4.5 BC in Covid-19 lockdown period

India went into first phase of nation-wide lockdown for 21 days from March 25, 2020 to fight the Covid-19 pandemic. All the industrial activity and traffic movement except essential services were prohibited. As a result, unprecedented low levels of pollution were seen in Delhi and throughout India (Dhaka et al., 2020; Mahato et al., 2020). In this study, the impact of Covid-19 lockdown on BC concentration at measurement site are analysed. Following a similar approach as in previous study where the effect of Covid-19 lockdown on PM$_{2.5}$ levels in Delhi has been analysed by
comparing data of 7 days before and 7 days into lockdown (Dhaka et al., 2020), in this study also $M_{BC}$ for continuous 15 days before (March 10-24, 2020) and 15 days after (March 25-April 8, 2020) the implementation of lockdown are considered for analysis.

Fig. 11 shows the day wise (one month) variation of $M_{BC}$ from March 10th to April 8th. The average $M_{BC}$ during 5th April night was about three times higher than the previous night due to nationwide light candles and firecrackers, and thus $M_{BC}$ data for these two days were excluded from our analysis. Averaged $M_{BC}$ for 15 days before lockdown ($3.42 \pm 1.06 \, \mu g \, m^{-3}$) is almost three times the averaged $M_{BC}$ for 15 days into lockdown ($1.18 \pm 0.94 \, \mu g \, m^{-3}$). Wilcoxon rank test at significance level of 0.05 is performed to check the significance of difference in mean. BC data from April 9, 2020 to May 13, 2020 could not be recorded due to instrument failure as it could not be attended in lockdown. Therefore, effect of complete lockdown period on $M_{BC}$ cannot be realized. Nevertheless, significant decrease in $M_{BC}$ during 15 days into lockdown can be considered a contributing factor to lower $M_{BC}$ in summer of 2020 in Delhi.

4. CONCLUSIONS

Year-round BC measurements were made at an urban site in Delhi using COSMOS. Seasonal and monthly variation of $M_{BC}$ is analyzed to find season- and month-specific causes and sources. Winter and post-monsoon are the most polluted seasons. It is found that meteorological conditions encompass with topography of Delhi play an important role in observed variation. Fire spot and air mass trajectory analyses revealed that in post-monsoon (October and November), the regional transport of pollution due to crop-residual burning in nearby states is important contributing factor for elevation of $M_{BC}$ in Delhi. However, in winter, combined effect of increased local emissions and non-favorable meteorological conditions are responsible for very high $M_{BC}$. Emission from
open burning by local people to keep warm themselves during winters could be the primary reason for increased local emissions. Furthermore, sudden increase in local emissions due to cracker burning on festivals is found to be the responsible factor for very dangerous levels of $M_{BC}$ (1-hour average $> 40 \, \mu g \, m^{-3}$). Covid-19 lockdown had a positive effect on air quality as $M_{BC}$ decreased up to three times due to traffic and local emission source reduction.

Monthly variation of BC concentrations showed strong correlation with meteorological parameters. Furthermore, BC concentrations were highest during winter season when topography of Delhi and meteorological conditions play a major role. Longer-term measurements will improve our quantitative understanding on the contributions of the factors controlling the observed high $M_{BC}$ (i.e., local sources, biomass burning in nearby states or geographical location/topography of Delhi together with meteorology). As per current analysis, it would be sufficed to point out that geographical location/specific topography together with meteorology is important factor for Delhi high BC concentration.

Proper mitigation steps should be taken to control high level of pollution especially in post-monsoon and winter seasons. These could be control over biomass burning (crop burning) in nearby states during October and November and controlling local emission sources (open burning to fight cold, vehicular emission) in winters. Several initiatives have been taken to control crop residual burning in nearby states. New agricultural machinery like happy seeder (https://www.ideasforindia.in/topics/environment/happy-seeder-a-solution-to-agricultural-fires-in-north-india.html) and super straw management system (Bhattacharyya et al., 2021) are promising innovations which can potentially alleviate the crop-residue burning issue. However, not much focus has been given to control local emission sources in winters. Furthermore, implementation of ban on crackers is required to control emissions during festivals like Diwali and New Year. More rigorous study primarily focused on quantification of local emission source
contribution to overall BC pollution (traffic, local open burning in winters and cracker burning) in Delhi is the need of the hour.

Further, to evaluate the representativeness of $M_{BC}$ data, we have compared these data with the Aethalometer data obtained from nearby monitoring stations, and also with satellite data, i.e., Modern-Era Retrospective analysis for Research and Applications - version 2 (MERRA-2)'s surface black carbon data of Delhi. Reasonably good correlations of $M_{BC}$ data with the other data suggest that $M_{BC}$ data reported in this study represent reasonably well the BC variations in Delhi for the study period.

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trends and source apportionment of fossil fuel and biomass burning black carbon (BC) in the

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simulated by nasa’s merra version 2 aerosol reanalysis over india and its relation to the air


Table 1. Summary of all major events during measurement period.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Event</th>
<th>Period</th>
<th>Source</th>
<th>Effect on $M_{BC}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Diwali</td>
<td>October 27, 2019</td>
<td>Fireworks</td>
<td>Sudden increase in $M_{BC}$ during celebration night leading to second largest peak in weakly averaged $M_{BC}$ plot (see Fig. 4(b)).</td>
</tr>
<tr>
<td>2</td>
<td>Regional transport</td>
<td>October - November 2019</td>
<td>Crop-residue burning</td>
<td>$M_{BC}$ in October increased nearly three times as compared to September (see Fig. 6(b)).</td>
</tr>
<tr>
<td>3</td>
<td>New Year</td>
<td>December 31, 2019</td>
<td>Fireworks</td>
<td>Sudden increase in BC levels during celebration night leading to largest peak in weakly averaged $M_{BC}$ plot (see Fig. 4(b)).</td>
</tr>
<tr>
<td>4</td>
<td>Cold winter</td>
<td>December 2019 – February 2020</td>
<td>Local open-burning</td>
<td>Increased local emissions along with non-favourable meteorological condition leading to highest daily $M_{BC}$ in December and January (see Fig. 6(b)).</td>
</tr>
<tr>
<td>5</td>
<td>Covid-19 lockdown</td>
<td>March 25, 2020 – April 9, 2020</td>
<td>Negligible sources</td>
<td>Averaged $M_{BC}$ for 15 days before lockdown (3.42 ±1.06 µg m$^{-3}$) is almost three times the averaged $M_{BC}$ for 15 days into lockdown (1.18 ± 0.94 µg m$^{-3}$).</td>
</tr>
</tbody>
</table>
Table 2. Correlation coefficient (r) between $M_{BC}$ and meteorological parameters and fire spots.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Correlation (r, for n = 307) with 24-hour averaged $M_{BC}$ (µg m$^{-3}$)</th>
<th>Correlation (r, for n = 12) with monthly averaged $M_{BC}$ (µg m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient temperature (°C)</td>
<td>-0.61</td>
<td>-0.83</td>
</tr>
<tr>
<td>Relative humidity (%)</td>
<td>0.08</td>
<td>0.30</td>
</tr>
<tr>
<td>Solar radiation (W m$^{-2}$)</td>
<td>-0.56</td>
<td>-0.84</td>
</tr>
<tr>
<td>Precipitation (mm)</td>
<td>NA</td>
<td>-0.51</td>
</tr>
<tr>
<td>Wind speed (m s$^{-1}$)</td>
<td>-0.47</td>
<td>-0.68</td>
</tr>
<tr>
<td>Boundary layer height (m)</td>
<td>-0.47</td>
<td>-0.68</td>
</tr>
<tr>
<td>Ventilation coefficient</td>
<td>-0.48</td>
<td>-0.74</td>
</tr>
<tr>
<td>Fire spots (km$^{-2}$)</td>
<td>NA</td>
<td>0.35 (all months)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.81 (excluding December and January)</td>
</tr>
</tbody>
</table>
Fig. 1. Inset of world map showing location of Delhi (a), location of measurement site, nearby Aethalometer measurement site, and MERRA-2 coordinates (b) which are used for comparison. Digital elevation model map (c) of Delhi is also included to show the role of its topography for low dispersion rate. Furthermore, surrounding of measurement site (d) is also shown to denote the urban location.
Fig. 2. Month wise variation of (a) ambient temperature (AT), (b) relative humidity (RH), (c) solar radiation (SR), and (d) precipitation. Mean values are denoted by circles and horizontal bold lines represents median value. For making these plots, 10, 25, 50, 75, 90 percentiles are used, i.e., 80% observations (10 to 90 percentile) lie between the whisker (vertical lines) and 50% (25 to 75 percentile) values lie within the box (same criteria are used for making all box plots in this paper).
Fig. 3 Scatter plot of 24-hour averaged $M_{BC}$ (AE) at PUSA (a) and SBC (b) against $M_{BC}$. Inset figure shows the relation between SBC and $M_{BC} < 8 \, \mu g \, m^{-3}$. 
Fig. 4. Temporal variation of $M_{BC}$: 1-hour, 24-hour (a), and weekly averaged (b). All the events are highlighted by color coded bars in weekly averaged plots and are discussed in details in text.
Fig. 5. Box and whisker plots showing diurnal variation of $M_{BC}$ throughout the observation period.
Fig. 6. Box and whisker plots showing seasonal (a) and monthly variation (b) of $M_{\text{BC}}$. 
Fig. 7. Monthly variation of boundary layer height (BLH) (a), wind speed (WS) (b), and ventilation coefficient (VC) (c) over Delhi.
Fig. 8. Maps showing air mass back trajectories and fire spots in nearby areas of Delhi for the months of September (a), and October (b).
Fig. 9. Monthly averaged fire spots per unit area in the trajectory path showing effect of fire counts in the months of May, October and November (a), also NWR analysis for post-monsoon and winter season showing dominance of local emission during winter (b).
Fig. 10. 1-Hour averaged $M_{BC}$ time series during Diwali and New Year festivals.
Fig. 11. $M_{BC}$ from March 10, 2020 to April 8, 2020.