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Technical Information

Perturbation of Background Atmospheric Black Carbon/PM_1 Ratio during Firecracker Bursting Episode

Deepanjan Majumdar* and Ashok Gangadhar Gavane[1)]

Kolkata Zonal Centre, CSIR-National Environmental Engineering Research Institute, i-8, Sector C, EKDP, EM Bypass, Kolkata-700107, India
[1)]Air Pollution Control Division, CSIR-National Environmental Engineering Research Institute, Nehru Marg, Nagpur-440020, India

*Corresponding author. Tel: +91-33-24421988, E-mail: d_majumdar@neeri.res.in / joy_ensc@yahoo.com

ABSTRACT

Perturbation in ambient particulate matter (PM_1, $PM_{2.5}$, PM_{10}) and black carbon (BC) concentrations was studied during a firecracker bursting episode in Diwali (Festival of Lights) celebrations in Nagpur, India. Firecracker bursting resulted in greater escalation in fine particulates over coarse particulates while $PM_{2.5}$ was found to be dominated by PM_1 concentration. On the Diwali day, daily mean concentration of $PM_{2.5}$ and PM_{10} exceeded Indian National Ambient Air Quality Standards by over 1.8 and 1.5 times, respectively, while daily mean BC concentration on the same day was almost two times higher than the previous day. The BC/PM_1 ratio reduced remarkably from about 0.26 recorded before firecracker bursting activity to about 0.09 during firecracker bursting on Diwali night in spite of simultaneous escalation in ambient BC concentration. Such aberration in BC/PM_1 was evidently a result of much higher escalation in PM_1 than BC in ambient air during firecracker bursting. The study highlighted strong perturbations in ambient PM_1, $PM_{2.5}$, PM_{10} concentrations and BC/PM_1 during the firecracker bursting episode. Altered atmospheric BC/PM_1 ratios could serve as indicators of firecracker-polluted air and similar BC/PM_1 ratios in local and regional air masses might be used as diagnostic ratios for firecracker smoke.

Key words: Aerosol, Air pollution, Atmosphere, Diwali, Firework

1. INTRODUCTION

Black carbon (BC) is one of the primary components of airborne particulate matter (PM) (http://www.ccacoalition.org/en/files/blackcarbonsasiafinalreport52212pdf; accessed on 04.04.2016). Black carbon, which is presumed to be mainly elemental carbon (EC) (Babich *et al.*, 2000), is generated by combustion of carbon containing materials (Hansen *et al.*, 1988) and is mostly ultrafine in nature (Gong *et al.*, 2016). Atmospheric BC concentration is generally low in remote areas e.g. in South Pole (Hansen *et al.*, 1998; Andrae *et al.*, 1995) and Manua Loa (Bodhaine, 1995) but higher concentrations (1-30 $\mu g\ m^{-3}$) are observed near its sources in urban areas and regions witnessing biomass burning (Ruellan *et al.*, 1999).

Firecrackers are known to be a major source of particulates and their bursting causes substantial air pollution during popular public festivals, namely the 'Festival of Lights', known as 'Diwali' in India and the 'Lantern Festival' in China (Barman *et al.*, 2008; Wang *et al.*, 2007; Ravindra *et al.*, 2003). Firecracker bursting results in emissions of smoke laden with heavy load of fine particulates (Barman *et al.*, 2008; Ravindra *et al.*, 2003) that are reported to carry various metals, anions and cations (Wang *et al.*, 2007; Kulshrestha *et al.*, 2004). On a Diwali day in Lucknow in India, 24-h average concentration of PM_{10} was found to be higher by almost 2.5 and 5.7 times than pre-Diwali and a normal day, respectively, and on Diwali night, the 12 h mean level of PM_{10} was 4 times higher than its respective daytime concentration (Barman *et al.*, 2008). Thakur *et al.* (2010) recorded higher particulate concentration (6.44 times for suspended particulate matter (SPM), 7.16 times for PM_{10}, 5.35 times for $PM_{2.5}$) during Diwali over a typical winter day in India and particulate concentrations exceeded its respective 24 hour residential air quality standards by several times (11.6 times for SPM, 22.3 times for PM_{10}, and 34.3 times for $PM_{2.5}$). During fireworks on Lantern Day in China, PM_{10} levels showed an elevation of up to 183% over the previous day while $PM_{2.5}$ and PM_{10} were more than six and four times higher than a normal day (Wang *et al.*, 2007). During a firework episode in Milan, Italy, PM_{10} emitted by fireworks was found to constitute about 50% of total PM_{10} mass in air (Vecchi *et al.*, 2008). Peshin *et al.* (2017) reported that average surface PM_{10} and $PM_{2.5}$ concentration levels showed an average increase of 387 and 278% in 2013; 123 and 145% in 2014; 42 and 43% in 2015 on Diwali day over a normal day at a site in India. During Chinese New Year in 2016, $PM_{2.5}$ were found to be significantly higher than Grade I National Standard of 35 $\mu g\ m^{-3}$ and contributions of secondary water soluble inorganic ions (WSII) to total WSIIs were lower in rural sites than urban ones in Central and East China (Zhang *et al.*, 2017).

Firecracker bursting is also responsible for BC emissions (Ramachandran and Rajesh, 2007; Suresh Babu and Moorthy, 2001). As per various reports, proportion of BC or EC in ambient particulate matter varies widely (Lim *et al.*, 2011; Fromme *et al.*, 2008). Lim *et al.* (2011) have reported that average mass fractions of elemental carbon (EC) were 10.4% in PM_1, 9.8% in $PM_{2.5}$ and 6.0% in PM_{10} at a site in Jeju Island during a 1-year study. The Indian Ocean Experiment (INDOEX) measurements have reported a 14% share of BC in fine aerosol dry mass in marine boundary layer (Lelieveld *et al.*, 2001).

Studying escalation in ambient particulate and BC concentrations and evaluating the alterations in BC/PM_1 ratios could be crucial to understand and characterize atmospheric perturbation during firecracker bursting episodes. We report the results of a study on temporal variation of particulates (PM_1, $PM_{2.5}$ and PM_{10}) and BC in ambient air within a residential area observed before, during and after firecracker bursting during the Festival of Lights (Diwali) in Nagpur City, India. Local background data on ambient air for the same parameters was collected for three-and-a-half months leading to the Diwali day in October. The extent of short-term escalation in particulate matter and BC and changing BC/PM_1 were studied and evaluated. Possible use of BC/PM_1 ratios as a diagnostic ratio for firecracker-polluted air is discussed. Instigating scientific debate, paving for future studies and development of firecracker-smoke related diagnostic ratios for future air pollution studies underline the importance of this study.

2. RESEARCH METHODS

The air quality monitoring activity was conducted in 2012 at a site located inside a residential colony in the city of Nagpur in India. Particulate matter (PM_1, $PM_{2.5}$ and PM_{10}) and BC concentrations in ambient air were studied continuously for three-and-half months to trace their temporal variation, finally leading to the Diwali night. The 4-day monitoring activity during Diwali celebration was divided into a few sampling periods designated as 'Background' (period before firecracker bursting started), 'Pre-Diwali Night' (period during sporadic firecracker bursting activities the night before Diwali), 'Interim Background' (period after the sporadic firecracker activities stopped up to Diwali evening), 'Diwali Night' (period of actual firecracker bursting during Diwali night) and 'Post Diwali Background' (few hours period after firecracker bursting ended on Diwali night) to track short-term perturbations in ambient particulates. Ambient particulate concentration was monitored by a portable aerosol spectrometer (Model 1.109, Grimm Aerosol Technik GMBH & Co., Germany) installed on a raised platform of approximately 4.7 meter height at a distance of approximately 10 meters from the principal firecracker bursting site. Particulate mass concentration ($\mu g\ m^{-3}$) in ambient air was measured in *real time* in three different size channels viz. 0.25-1.0, 0.25-2.5 and 0.25-10 μm at 1-minute time interval (time-base) during Diwali celebrations (for 4 days) while pre-Diwali background data for three-and-a-half month period was captured at three-hour intervals. A shorter sampling time-base for Diwali celebrations was selected to capture short-lived particulate generation events during firecracker bursting activities. The PM_1 mass concentration values were used for calculation of BC/PM_1.

A factor called 'C factor' which is a ratio of the actual mass of particulates deposited on the filter paper fixed within the instrument and mass of particulates reported by the instrument, was calculated. The C factor is used as a correction factor that is multiplied with the instrument reported particulate concentration values to get the corrected particulate concentrations in ambient air. The instrument reports particulate mass concentrations based on its calibration with monodisperse polystyrene latex particles and therefore multiplication of 'C factor' with instrument generated mass concentration values is essential to arrive at the actual ambient particulate mass concentrations.

Measurement of *real time* ambient BC mass concentration was made by an Aethalometer (Model AE-42, Magee Scientific Inc., USA), configured for a time base of 1-minute during the 4-day Diwali celebrations while the pre-Diwali background data of three-and-a-half months was collected at three-hour intervals. The shorter time-base was selected for the same reason as cited earlier in case of aerosol spectrometer. The monitoring activity during Diwali celebrations was divided into the same sampling periods discussed in one of the earlier sections. The sampling inlet of Aethalometer was fitted with a size-selective cyclone with 1-μm cut off diameter (BGI Inc.) to allow the measurement of BC in PM_1 only and to get realistic measures of BC/

PM_1. The Aethalometer was installed on the same platform along with the spectrometer to simultaneously sample ambient air at the same point. The proportion of BC in PM_1 (i.e. BC/PM_1) was calculated from the mass of BC reported by the Aethalometer fitted with 1 μm size selective inlet and simultaneous mass concentration of PM_1 reported by the spectrometer, as shown below:

BC/PM_1 = [(Average BC conc. in μg m^{-3} reported by Aethalometer fitted with a cyclone with 1 μm cut off diameter)/(Average PM_1 conc. in μg m^{-3} reported by Spectrometer)]

The observed data were subjected to descriptive as well as advanced statistical analysis. Two different regression analyses with prediction and confidence intervals were performed between ambient PM_1 and BC mass concentrations that were observed during (i) the entire monitoring duration and (ii) without mass concentrations of the values recorded during firecracker bursting period on Diwali night, to understand whether firecracker bursting led to disproportionate emissions of BC and PM_1 and disturbed their *business as usual* relationship. Pearson correlation analysis were also performed on the same data to evaluate if the *business as usual* correlation between PM_1 and BC was disturbed by the firecracker bursting or not. Mann-Whitney Rank Sum Test, which is used to compare two sample means that come from the same population and used to test whether two sample means are equal or not, was performed on ambient PM_1 and BC mass concentration values observed during firecracker bursting period on Diwali night and no-firecracker activities prior to Diwali, to examine if these groups of data under two different circumstances were statistically different from each other or not. The Mann-Whitney Rank Sum Test has been used earlier in air pollution studies for comparison of sample data sets (Röllin *et al.*, 2004; Di Giorgio *et al.*, 1996). The above statistical analyses and tests were performed by Sigmaplot (Version 13, SysStat) and Statistica (Version 13, Dell). Further, to examine statistically significant difference amongst various values in the data sets, Duncan's Multiple Range Test (DMRT) was conducted by MSTAT-C statistical program (Michigan State University, USA).

3. RESULTS AND DISCUSSION

3.1 PM_1, $PM_{2.5}$ and PM_{10} Concentrations

There was a sharp increase in the concentration of ambient particulate matter during bursting of firecrackers, both on the pre-Diwali day and on the night of Diwali itself. The escalation can be appreciated when the time series of background PM_1, $PM_{2.5}$ and PM_{10} concentrations in the preceding three-and-a-half months are compared (Fig. 1). There were marginal differences in between PM_1 and $PM_{2.5}$ time-series concentrations recorded during Diwali celebrations, indicating predominance of PM_1 in fine particulate fractions. On the day of Diwali and the preceding day, several BC and particulate peaks were recorded, which levelled off and reappeared a few times when firecracker bursting activities were at their peak. Larger peaks of particulates in the time-series plot marked the Diwali night while smaller perturbations were visible in the time-series plot the day before Diwali due to sporadic

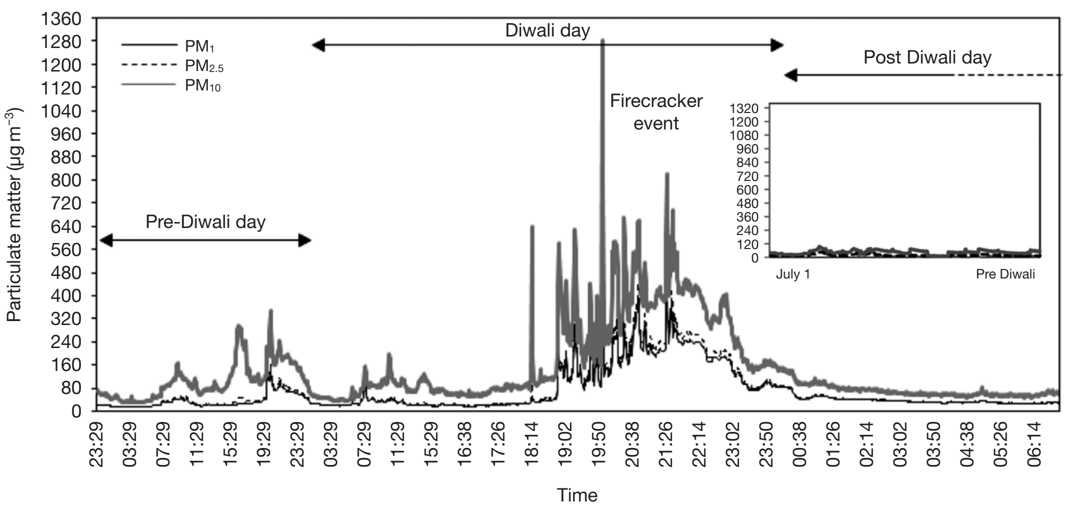

Fig. 1. Temporal variation in particulate matter (PM_1, $PM_{2.5}$ and PM_{10}) concentration in ambient air during Diwali celebrations [Inset: Trend of background PM_1, $PM_{2.5}$ and PM_{10} concentration in ambient air during three-and-a-half months before Diwali].

Table 1. Summary of average particulate and BC concentrations during designated time-periods during Diwali celebrations.

Sample name	Date	Time interval	Duration	Mean BC ($\mu g\ m^{-3}$)	Mean PM_1 ($\mu g\ m^{-3}$)	Mean $PM_{2.5}$ ($\mu g\ m^{-3}$)	Mean PM_{10} ($\mu g\ m^{-3}$)
Background	15-16th October	23:24-17:30	18 h 6 min	5.8 a	22.4 a	28.8 a	87.5 a
Pre Diwali night	16-17th October	17:39-00:12	6 h 33 min	17.8 b (+206.9%)	61.9 c (+176.3%)	72.9 c (+153.1%)	161.7 b (+84.8%)
Interim background	17-17th October	00:19-18:51	18 h 32 min	5.9 a (−66.5%)	26.2 a (−57.7%)	31.7 a (−56.5%)	83.1 a (−48.6%)
Diwali night[a)]	17-17th October	18:58-23:16	5 h 18 min	19.6 b (+232.2%) **(+9.18%)**	198.5 d (+657.6%) **(+220.7%)**	222.0 d (+600.3%) **(+204.5%)**	376.8 c (+353.4%) **(+133.0%)**
Post Diwali background	17-18th October	23:20-05:51	6 h 49 min	6.8 a (−65.3%)	44.4 bc (−77.6%)	49.7 b (−77.6%)	86.7 a (−76.9%)
Daily average[b)] (pre Diwali day)	16th October	24-hourly	00:00 to 24:00 hrs	8.4	33.2	40.9	73.0
Daily average[b)] (Diwali day)	17th October	24-hourly	00:00 to 24:00 hrs	14.3 (+70.2%)	97.6 (+193.7%)	110.5 (+170.2%)	155.4 (+112.9%)

Values in parentheses represent increase (+) or decrease (−) over the immediately preceding value
[a)]Bold letters indicate increase over Pre-Diwali night
[b)]Daily averages were calculated by from the minute-wise BC and PM concentrations reported by the instruments within each 24-h period
Values followed by same letters are not statistically different from each other as per Duncan's Multiple Range Test (DMRT) at 5% level of significance

and less intense firecracker bursting activities. On Diwali night, there were surges in average ambient PM_1, $PM_{2.5}$ and PM_{10} concentrations over the immediately preceding period (i.e. interim background) which were statistically significant. The escalation ranged from 353% to 658% and was higher for smaller size groups, indicating generation of predominantly fine particulates by firecrackers (Table 1). The increase on Diwali night over the pre-Diwali night was less pronounced (133-221%). Earlier, on the pre-Diwali night as well, there was a substantial increase in ambient particulate concentration, ranging from 85-176%, due to the sporadic bursting of firecrackers, and in this case, too, the finer particulate concentration had a higher escalation over the immediately preceding background. However, the escalations in 24-hourly PM_1, $PM_{2.5}$ and PM_{10} concentrations on Diwali day over pre-Diwali day were much lower, ranging from 113-194%. The escalated particulate concentrations declined quickly within a few hours after the bursting of firecrackers stopped and, by early next morning, particulate concentrations had dropped towards the background levels.

Mean 24-hourly concentrations of PM_1, $PM_{2.5}$ and PM_{10} were calculated after taking into account the particulate concentrations over the entire Diwali day and it was observed that daily average $PM_{2.5}$ and PM_{10} concentration on the Diwali day exceeded Indian National Ambient Air Quality Standards (http://cpcb.nic.in/National_Ambient_Air_Quality_Standards.php; accessed on 04.04.2016) by over 1.8 and 1.6 times, respectively. The Indian National Ambient Air Quality Standard prescribes PM_{10} limits of 60 and 100 $\mu g\ m^{-3}$ on annual and 24-h basis for all types of areas (industrial, residential, sensitive) and 40 and 60 $\mu g\ m^{-3}$ on annual and 24-h basis for $PM_{2.5}$ for all types of areas (http://cpcb.nic.in/National_Ambient_Air_Quality_Standards.php). Firecrackers are composed of inorganic salts and carrier materials, which would escape and add to atmospheric particulate load during firecracker bursting while the combustion process, inclusive of explosion, would help accelerate the generation of fine particulates from base material mixtures. Mann-Whitney Rank Sum Test revealed that the difference in median between two groups of ambient PM_1 concentrations observed with and without firecracker bursting was greater than what would be expected by chance and therefore was statistically significant ($P = < 0.001$). This implied that ambient PM_1 concentration levels reached during firecracker bursting activity were statistically higher than its levels when no firecracker was burst.

Perturbation in atmospheric particulate load during firework displays has been studied by several researchers. Chang *et al.* (2011) has reported that during a fireworks display, hourly concentration of PM_{10} and $PM_{2.5}$ at Yanshui Primary School in Taiwan reached about 429 and 250 $\mu g\ m^{-3}$, respectively, which was about 10 times the normal level, and 6-second values even escalated to as high as 1046 and 842 $\mu g\ m^{-3}$, respectively. The number of particles with < 100 nm diameter

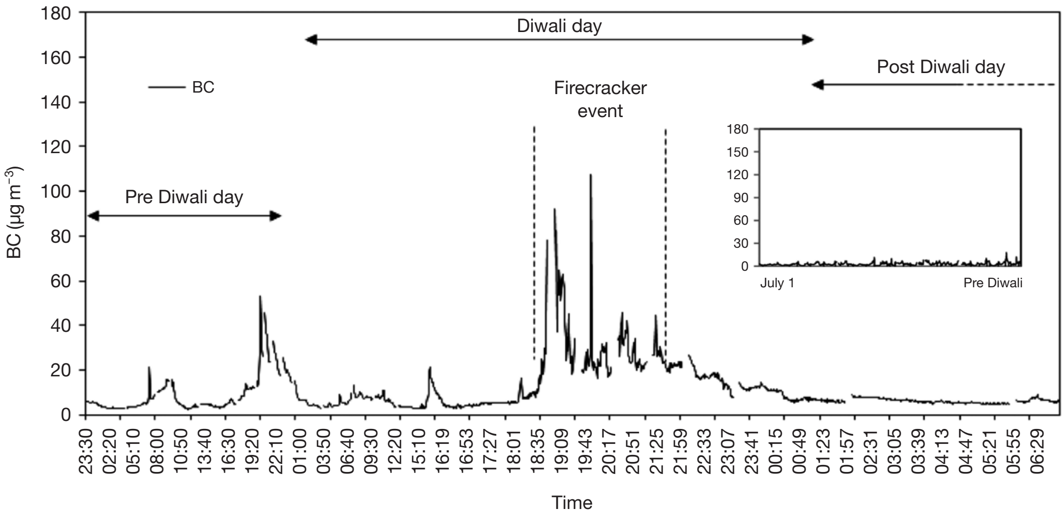

Fig. 2. Temporal variation in black carbon (BC) concentration in ambient air during Diwali celebrations [Inset: Trend of background BC concentration in ambient air during three-and-a-half months before Diwali].

increased abruptly during the event period. In another study in Taiwan (Tsai, 2012), concentration of Mg, K, Pb, and Sr in $PM_{2.5}$ were found to be 10 times higher during firework periods than during the non-firework periods. Additionally, the Cl^-/Na^+ ratio was approximately 3 during the firework display periods as Cl^- came from the firework powder with the OC/EC ratio increasing up to 2.8. Duo *et al.* (2015), reported that the average hourly ratio of $PM_{2.5}/PM_{10}$ reached 0.67, almost 1.5 times higher than the average ratio of $PM_{2.5}/PM_{10}$ of 0.42 during firework display on the New Year in Lhasa in Tibet, indicating fine particle generation by firework display. In Malta, Religious festivals (Festas) are observed during summer when 86 of them are celebrated between June and October, each involving fireworks over a period of 3 days or longer per festival. The PM_{10} and its content of Al, Ba, Cu, Sr and Sb that are used in pyrotechnic compositions, were found to be significantly higher during July-August than September-October and a strong correlation between PM_{10} and total metal content was observed. Authors concluded that fireworks had a significant negative impact on air quality (Camilleri and Vella, 2010). In a study conducted in Korea, firework events led to elevated concentrations of trace metals (K - 1.72 folds, Sr - 2.64 folds, As - 2.86 folds, Pb - 2.91 folds and Al - 5.44 folds) and levels of some metals did not drop to background level even after a day of the firework event. The contribution of fireworks to trace metal levels in 2011 event was negligible compared to 2012 and 2013 events due to different precipitation levels (Shon *et al.*, 2015).

3.2 Concentrations of Black Carbon

The escalation in ambient BC concentration on Diwali night over interim background and pre-Diwali night concentrations were to the extents of 232% and 9%, respectively, which were much less pronounced than the increase in ambient PM_1, $PM_{2.5}$ and PM_{10} concentration under similar circumstances. The escalation was, however, substantial over the background concentration of BC (Fig. 2). On the other hand, the 24-hourly average BC concentration on Diwali day was 70% higher than the pre-Diwali day, which was again much lesser than PM_1, $PM_{2.5}$ and PM_{10}. Escalation in BC concentration during fireworks is presumably a result of combustion of carbonaceous materials used in firecrackers as propellants and reducing agents. Escalation in BC concentration by a factor of over 3 above the background level has been reported by Suresh Babu and Moorthy (2001). Significant influence of fireworks on OC and EC concentrations are also reported in urban areas like Bhilai in India (a 7- and 2.9-times increase during Diwali compared to pre-Diwali days for OC and EC, respectively) (Pervez *et al.*, 2016), Milan in Italy and Lucknow in India (a 1.6-fold increase for OC and 3-fold for EC) (Barman *et al.*, 2008; Vecchi *et al.*, 2008).

Interestingly, BC/PM_1 reduced remarkably by about 3 times during firecracker bursting on Diwali night as compared to previous sampling periods, in spite of escalation in actual ambient BC concentration during the former. The strong BC/PM_1 perturbation for about 5-6 hours during firecracker bursting in Diwali night was instrumental in altering 24-h average BC/PM_1 ratio of the entire Diwali day to 0.15 against the pre-Diwali

Table 2. Observed BC/PM_1 in different time periods *vis a vis* activities related to firecracker bursting.

Sample name	Time period	BC/PM_1[a)]
Background	23:24-17:30	0.26
Pre Diwali Night	17:39-00:12	0.29
Interim Background	00:19-18:51	0.23
Diwali Night	18:58-23:16	0.09
Post Diwali Background	23:20-05:51	0.15
Daily Average (16th October): Pre Diwali day	24-hourly	0.25
Daily Average (17th October): Diwali day	24-hourly	0.15

[a)][(Average BC conc. in μg m^{-3} reported by Aethalometer fitted with a cyclone with 1 μm cut off diameter)/(Average PM_1 conc. in μg m^{-3} reported by Spectrometer)]

day average of 0.25 (Table 2). This was evidently a result of much higher increase in PM_1 concentration than BC in ambient air during firecracker bursting on Diwali night (Table 1). The background BC/PM_1 values varied within a very narrow range (0.23-0.29) before the initiation of firecracker bursting on Diwali night and returned to a value of 0.25 within a few hours after the firecracker bursting was over. Interestingly, the same ratio had reduced significantly to 0.09 during the firecracker bursting period. Similar low BC/PM_1 ratios in ambient air might indicate encroachment by air mass polluted by firecracker-smoke, raising the possibility of making it a diagnostic ratio to indicate such pollution. Further studies and confirmation of similar BC/PM_1 ratios during firecracker bursting could be necessary before taking a final call on the practicality of such a ratio. It may be noted that future studies must look into background BC/PM_1 ratios to examine the extent of their regional variability and in case of substantial variability, region-specific firecracker-specific diagnostic ratios may have to be generated. Zhang *et al.* (2017) reported that the ratio of Water Soluble Organic Carbon/Organic Carbon decreased during fireworks displays than during non-fireworks periods, implying that direct OC emissions from fireworks may have been water-insoluble. Liu *et al.* (2016) reported that the OC/EC (organic C / elemental C) ratios and the percentages of secondary organic C / Organic C declined during the Chinese Spring Festival when large-scale fireworks were under display. The difference in origin of thermal POC (primary organic C) and thermal SOC (secondary organic C) led to a low correlation ($R^2 = 0.39$) of the two. During the festival, thermal POC dominated thermal OC generation.

Mann-Whitney Rank Sum Test revealed that ambient BC concentration levels observed during firecracker bursting were significantly and statistically higher than BC levels with no firecrackers burnt ($P = <0.001$). The best-fit curve of ambient PM_1 with BC concentration during the monitoring period is presented in Fig. 3a. Correlation of BC with PM_1 had a Pearson Correlation Coefficient of 0.73 when data over entire monitoring period was considered, but when the correlation was revisited after neglecting their ambient concentrations recorded on Diwali night, the same correlation coefficient increased to 0.86, indicating better correlation. Also, the relationship between PM_1 and BC, as indicated by regression analysis, improved with a much better R^2 value when their concentration on Diwali night was omitted, indicating that disproportionate cogeneration of BC and PM_1 during firecracker bursting on Diwali night was instrumental in changing the otherwise better relationship between PM_1 and BC existing in unperturbed atmosphere (Fig. 3b).

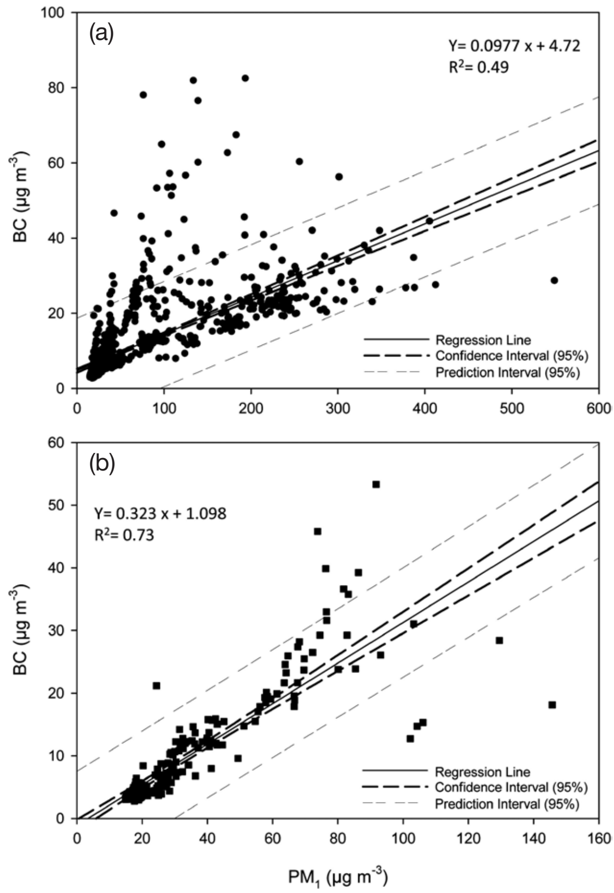

Fig. 3. Regression equation and Pearson Correlation of ambient black carbon (BC) concentration with ambient PM_1 concentrations (a) During entire monitoring duration i.e. before, during and after Diwali (b) Excluding firecracker bursting period on Diwali day.

4. CONCLUSION

Significantly, this study highlighted the alteration in BC/PM_1 ratio, apart from the perturbations in ambient particulate matter and BC concentrations during firecracker bursting activity. The following important observations were made: (i) Firecracker bursting led to much greater increase in fine particulate concentration over coarser particulates in ambient air; concentrations of $PM_{2.5}$ and PM_{10} were clearly dominated by PM_1 (ii) Firecrackers generated substantial amount of BC also but it was much lesser in magnitude than PM_1 (iii) BC/PM_1 ratio declined substantially from the background levels, by about 3 times, during the firecracker bursting episode but returned to background values within a few hours after the termination of firecracker bursting episode.

We hypothesize that such altered BC/PM_1 ratios during firecracker bursting could indicate atmospheric pollution by firecracker smoke and hence may be attempted as diagnostic ratios for such type of air pollution. Diagnostic BC/PM_1 ratios for firecracker-polluted air should preferably be arrived at after sufficient number of air quality studies on firecracker bursting and examination of similar ratios. This study may open up a scientific debate on the possible drawbacks, uncertainties and applicability of the proposed diagnostic ratio. Similar regional studies under different activities (e.g. firecracker bursting, traffic pollution, biomass burning, volcanic activity etc.) could lead to streamlining of signature diagnostic BC/PM_1 ratios, each specific to a particular activity.

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