Fire Induced Particle Pollution


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Atmospheric Research 180 (2016) 78–91

Contents lists available at ScienceDirect

Atmospheric Research

journal homepage: www.elsevier.com/locate/atmosres

Fireworks induced particle pollution: A spatio-temporal analysis☆

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ARTICLE INFO

Article history:
Received 19 January 2016
Received in revised form 7 May 2016
Accepted 18 May 2016
Available online 20 May 2016

Keywords:
Aerosol
CALIPSO
Fireworks
Indo-Gangetic Plain
MODIS-AOD
OMI

ABSTRACT

Diwali-specific firework induced particle pollution was measured in terms of aerosol mass loading, type, optical properties and vertical distribution. Entire nation exhibited an increase in particulate concentrations specifically in Indo-Gangetic Plain (IGP). Aerosol surface mass loading at middle IGP revealed an increase of 56–121% during festival days in comparison to their background concentrations. Space-borne measurements (Aqua and Terra-MODIS) typically identified IGP with moderate to high AOD (0.3–0.8) during pre-festive days which transmutes to very high AOD (0.4–1.8) during Diwali-day with accumulation of aerosol fine mode fractions (0.3–1.0). Most of the aerosol surface monitoring stations exhibited increase in $PM_{2.5}$ especially on Diwali-day while PM_{10} exhibited increase on subsequent days. Elemental compositions strongly support K, Ba, Sr, Cd, S and P to be considered as firework tracers. The upper and middle IGP revealed dominance of absorbing aerosols (OMI-AI: 0.80–1.40) while CALIPSO altitude-orbit-cross-section profiles established the presence of polluted dust which eventually modified with association of smoke and polluted continental during extreme fireworks. Diwali-specific these observations have implications on associating fireworks induced particle pollution and human health while inclusion of these observations should improve regional air quality model.

1. Introduction

Regional distribution of airborne particulates, its composition, morphology and mixing states are highly heterogeneous which link particulates to multi-lateral negative impacts (Kumar et al., 2015a; Banerjee et al., 2011a, 2011b, 2015). Many of the factors that regulate a particle's chemical behaviour are region specific while some extraneous effects may also induce additional uncertainties by injecting multiple foreign elements into the regional environment. These external effects used to transform the entire aerosol-climate system into a far more complicated and uncertain one (Devara et al., 2015). One localized impact is pyrotechnic displays that modify local environment by introducing sudden flush of particulates and associated toxins, sometimes on a regional, and often extends to national scale. Collective impact of fireworks therefore, definitely be considered as an episodic pollution event as it poses potential to modify chemical nature of aerosols (Vecchi et al., 2008). Thus create the scope of initiating a comprehensive campaign on firework induced particle pollution at middle IGP for recognizing the origin of aerosols and its spatio-temporal distribution.

There are a number of instances when extensive pyrotechnic displays are made for recreational purpose most notably during New Year's Eve (e.g. Sydney), Lantern Festival (China), Bonfire Night (UK), Independence Day (e.g. USA), Bastille Day (France) and Diwali (India). These festive events possibly introduce a number of foreign species to lower troposphere and therefore, were subject to intense scientific investigation. There is ample evidence of the link between firework emissions and associated degraded environment most notably in United States (Licudine et al., 2012); Milan (Vecchi et al., 2008); Mainz (Drewnick et al., 2006); Delhi (Perrino et al., 2011); China (Wang et al., 2007); Spain (Moreno et al., 2007) and Pune (Devara et al., 2015). In most instances attempts were specifically made to explore variation in aerosol mass loading and chemical characteristics while there were only a few efforts (Devara et al., 2015; Vyas and Saraswat, 2012) to associate mass loading with aerosol optical properties and vertical distribution. Additionally, previous studies focused solely on exploring regional atmosphere without integrating particulate spatial nature which seems extremely relevant in identifying particulate flux and its potential impacts.

For the present analysis, efforts were made to identify the impacts of Diwali-specific pyrotechnic displays on the environment through concurrent measurements of both in situ aerosol physico-chemical

☆ Capsule: Polluted and dust aerosols in the entire Indo-Gangetic Plain were modified through Diwali-specific firework emissions with an association of smoke and polluted continental aerosol.

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http://dx.doi.org/10.1016/j.atmosres.2016.05.014

characteristics substantiated with aerosol columnar properties. Parallel employment of both surface and space based approach pose potential to provide better insight to firework emission chemistry. Initially attempts were made to demonstrate spatial nature of firework induced particulate emissions for different stations using surface monitored data complemented with satellite observations. By our understanding, such efforts were unprecedented in Indian context. Surface based aerosol observations were concurrently made with satellite based platforms to recognize interrelations of aerosol properties. Conclusively, aerosol vertical profiles were analysed to isolate gradual modification of aerosol type at different altitudes. The implication of such analysis may well be useful to understand consequences of pyrotechnic displays on regional environment and to recognize inherent chemistry that exists within the firework emissions.

2. Experimental methods

2.1. Experimental site

The entire experiment was performed at Varanasi (25°16′29″N, 82°59′46″E) which is one of the critical stations of IGP with typically very high aerosol loading throughout the year (Kumar et al., 2015b; Murari et al., 2015; Tiwari et al., 2015; Sen et al., 2014, 2016) (Fig. 1). Ground aerosol monitoring was continued on the premises of Institute of Environment and Sustainable Development, Banaras Hindu University (IESD-BHU) which represents an institutional set up surrounded by typical mixed urban environment characterized with congested roads, mixed residential and commercial activities. Monitoring site characterizes the IGP in terms of climatology, topography and aerosol type without having any localized effects of oceans or mountains. Regional climatology is governed by wide range of synoptic weather pattern with relatively flat topography that simplifies the planetary boundary layer over the region.

2.2. Micro-meteorology

Regional meteorology is characterized as humid sub-tropical climate which starts experiencing temperature downfall from the last week of October. Meteorological parameters associated with the study region including temperature, relative humidity (RH), wind speed (WS) and wind direction (WD) were procured from web based meteorological database wunderground.com for Varanasi station and were further validated with regional weather monitoring station. Atmospheric boundary layer depths (ABL) and ventilation coefficient (VC) were additionally procured from Global Data Assimilation System (GDAS, 1°, 3-hourly) collected from National Oceanic and Atmospheric Administration-Air Resources Laboratory (NOAA-ARL), Real-time Environmental Applications and Display System (READY) website (http://www.arl.noaa.gov/ready; Draxler and Rolph, 2003).

2.3. In-situ aerosol measurements

The in-situ measurement of ambient aerosol having aerodynamic diameter (d_{ae}) of ≤10 μm (PM_{10}), ≤2.5 μm ($PM_{2.5}$) and total aerosol loadings (TSPM) were continued during October 16–30, 2014 in correspondence to Diwali festival celebrated on October 23. The entire experiment was performed in a campaign mode identically 7 days before and after Diwali to precisely distinguish the effects of fireworks on regional pollutant loading. Particulate monitoring was continued at a height of 7.0 m above ground for 24 h (12:00–12:00 h) on each consecutive day. However, in order to track particulate variability in a finer time frame on the Diwali day, particulate monitoring was scheduled on 4 h basis.

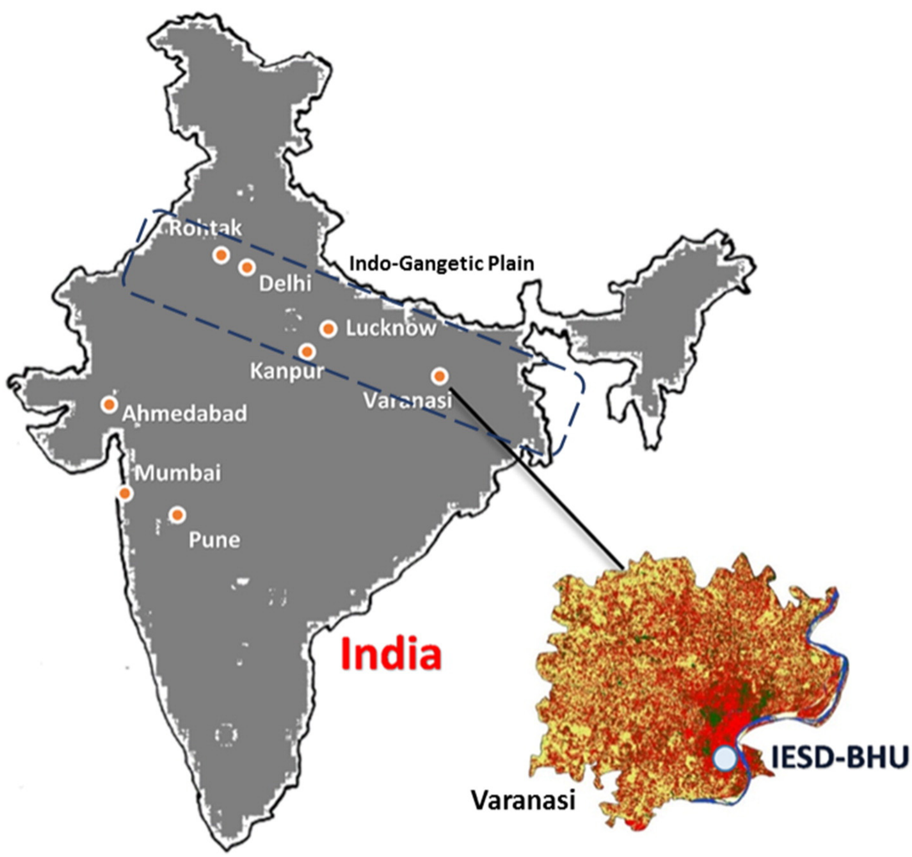

Fig. 1. Geographical location of aerosol ground monitoring stations. Note: The red, yellow and green pixels over Varanasi designate urbanization, open land and green cover, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Coarse particulates (PM_{10}) were collected on 20.3 × 25.4 cm sized glass fibre filters (GF/A Whatman) using Respirable Dust Sampler (APM-BL 460, Envirotech) having size selective inlet with an average flow rate of 1.2 m^3 min^{-1} (1.1–1.3 m^3 min^{-1}, accuracy ±2%). Total aerosol loadings were computed as an algebraic summation of PM_{10} and non-respirable particulates deposited in a separate collection device. $PM_{2.5}$ samples were collected with the help of fine dust sampler (APM-550, Envirotech) (1 m^3 h^{-1}, accuracy ±2%) using polytetrafluoroethylene filters (PTFE, 47 mm dia.). All filters were desiccated for 24 h before and after sampling and particulate mass concentrations were estimated gravimetrically. Exposed filters were stored under cool and dry condition (−20 °C) for chemical analysis. Details regarding particulate collection, analysis and quality control may found in the works of Murari et al. (2016). Concurrent particulate mass concentrations for other Indian cities were obtained from the central (cpcb.nic.in) and state pollution control board database available in public domain.

2.4. In-situ black carbon measurement

Black Carbon aerosols (BC) were measured concurrently in real time mode with a 7-channel Aethalometer (AE-42, Magee Scientific). The Aethalometer was continuously aspirated at a flow rate of 3.5 lpm from a height of ~5 m above ground level and BC particles were identified through observed attenuation at 880 nm. Details regarding black carbon collection and analysis may found in the works of Wang et al. (2012). Additionally, BC_{370} values were used for computing delta-C (BC_{370}–BC_{880} nm) for identifying specific anthropogenic contribution in terms of smoke (Yerramsetti et al., 2013) and particles emitted through wood combustion (Wang et al., 2012).

2.5. Particulate elemental analysis

The elemental composition of both PM_{10} and $PM_{2.5}$ collected on filters was determined by wavelength dispersive X-ray fluorescence spectrometer (XRF, X-Ray Analyser, ARL OPTIM-X, Thermo Scientific). The XRF provides precise analysis for selected major and minor elements or oxides in both inorganic and organic matrices. Details regarding working principles of XRF and particulate analysis may found in the works of Perrino et al. (2011).

2.6. Aerosol columnar properties

2.6.1. Surface retrieved AOD

Surface based measurements of aerosol optical depths were made with a portable multiband sun photometer MICROTOPS-II (MTP-AOD) (Solar Light, USA). MTP-AOD were measured at five (380, 440, 500, 675 and 870 nm) interface filters with a bandwidth of 2–10 nm at different channels and MTP-AOD at 550 nm were computed using Angstrom power law (Prasad et al., 2007). The MICROTOPS-II sunphotometer measures with a resolution of 0.01 wm^{-2}, 2.5° field of view, 0.01–0.03 associated uncertainties and measure aerosol optical thickness computing solar radiation extinction at a particular wavelength. Measurements were only performed during clear solar visibility conditions at a frequency of 30 min. MTP-AOD was additionally considered for validating satellite retrieved AOD. Angstrom exponent (α) explains the spectral dependence of AOD and was computed using Angstrom power law with the wavelength pair of 380 & 870 nm.

2.6.2. Satellite retrieved AOD

Satellite based daily columnar AOD were retrieved from Aqua MODIS (Moderate Resolution Imaging Spectro-radiometer) atmosphere level 2 products. The MODIS sensor on board AQUA satellite provides a spatial resolution of 250 m–1 km (at nadir) with high radiometric resolution (12 bits) links the columnar aerosol loadings from surface of Earth to the top of atmosphere by measuring reflected solar radiance and terrestrial emissions (Liu et al., 2007). For the current analysis, an area between 25°10′37″–25°19′47″N and 82°54′7″–83°4′30″E uniformly surrounding the ground monitoring site was selected for MODIS-AOD retrievals. Newly introduced Aqua MODIS 3-km atmospheric product (MYD04_3K; MODIS collection 6) was selectively retrieved through Atmosphere Archive and Distribution System (http://ladsweb.nascom.nasa.gov) and AOD in the pre-selected grid were further averaged and represented for the study site (Kumar et al., 2015b). Aqua MODIS 3-km aerosol product (MYD04_3K; Optial_Depth_Land_And_Ocean; quality flag-3) denotes AOD encompassing both finer and coarser aerosol and was deliberately chosen in order to achieve aerosol optical depths at finer resolution (3 × 3 km).

Terra MODIS Level 2 daily aerosol product (MOD04_L2; MODIS collection 6; 10 × 10 km) were processed from LAADS web for the entire Indian subcontinent (N: 25.0, S: 5.0, E: 90.0, W: 65.0) within similar timeframe. The intention was to recognize spatial and temporal distribution for a larger perspective both in terms of aerosol loading and aerosol fine mode fractions (FMF) in and around Diwali festival. Terra MODIS true granule images (RGB) with corresponding AOD (collection 51) for each aerosol loading episodes were also compared for Indian subcontinent to identify composite anomalies of synoptic aerosol transportation within the region.

In view of assessing fireworks induced aerosol loadings at middle IGP, it was essential to have parallel information about concurrent events of biomass burning or forest fires. Possible events of biomass burnings/wild fires were checked with Terra MODIS active fire location from FIRMS near-real time data source (http://firms.modaps.eosdis.nasa.gov/firemap/).

2.6.3. OMI aerosol index (near-UV)

The OMI aerosol index (OMI-AI) measures the wavelength dependence of backscattered UV radiation to discriminate dust and smoke from other aerosol types hypothesising high UV absorption (388 nm) properties of absorbing aerosol (e.g., organic carbon, smoke, and dust) (Torres et al., 2007). The OMI-AI is the product of ozone monitoring instrument (OMI), a high resolution spectrograph on board EOS-Aura platform. OMI pose a 2600 km wide swath having typical daily global coverage with a spatial resolution of 13 × 24 km at nadir. For the current analysis, OMTO3G_V003 UV aerosol index data product (OMI-Aura Level-2G Total Column Ozone Data Product OMTO3G_V003, Global 0.25° × 0.25° Lat./Lon.) were obtained from Goddard Earth Sciences Data and Information Services Centre (http://disc.sci.gsfc.nasa.gov/) to identify the presence and transport of mineral dust and smoke particles during pre-Diwali (October 16–22, 2014) and post-Diwali (October 23–30, 2014) event.

2.6.4. CALIOP aerosol vertical profile

Aerosol vertical profile was derived using Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) sensor on board CALIPSO satellite. CALIOP sensor has a fixed near-nadir view angle which carries a polarization-sensitive active lidar and specifically detects atmospheric aerosol profile measuring elastic laser backscatter (at 1.064 μm) with parallel and cross-polarized return signal (at 0.532 μm) (Kittaka et al., 2011). For the current analysis, CALIPSO altitude-orbit cross-section profiles (CAL_LID_L2_05kmAPro; level 2 version 3.30, 5-km ground horizontal resolution) were obtained from CALIPSO search and subsetting web application (https://www-calipso.larc.nasa.gov). For detail elaboration regarding CALIOP sensor and data profiles, readers are motivated to consult Kittaka et al. (2011).

3. Results and discussion

3.1. Spatial pattern of aerosol loading

Festival of light — Diwali is widely celebrated across India and possibly responsible for emitting huge amount of particulates and associated toxins into atmosphere. For the current analysis, spatial pattern of particulate origin and their subsequent evolution were initially analysed. Mostly, identical pattern was recognized with gradual increase in particulate loading during festive days (Fig. 2a–b). Entire nation exhibit sudden upsurge of Diwali-day specific (October 23, 2014) particulate concentration especially in Ahmedabad (PM_{10}: 2220 μg m^{-3}), Delhi (PM_{10}: 726; $PM_{2.5}$: 412 μg m^{-3}), Lucknow (PM_{10}: 741 μg m^{-3}) and Varanasi (PM_{10}: 432; $PM_{2.5}$: 280 μg m^{-3}). Characteristically, mass concentrations of both type of aerosols exceeded the 24-h average national standards (PM_{10}: 100 μg m^{-3}; $PM_{2.5}$: 60 μg m^{-3}; cpcb.nic.in), USEPA standards (PM_{10}: 150 μg m^{-3}; $PM_{2.5}$: 35 μg m^{-3}) (http://www.epa.gov) and the EU standards (PM_{10}: 50 μg m^{-3}, http://ec.europa.eu). Interestingly, all the surface monitoring stations exhibited increase in fine particulates loading especially on Diwali day or day after Diwali. In contrast, coarse particulate exhibited a consistent pattern of higher concentrations on day after Diwali which support particle aggregation which consequently enhance particulate effective diameter in subsequent days (Fig. 2). In absence of a collective datasets, evolution of particulate ratio ($PM_{2.5}/PM_{10}$) from pre-Diwali day (October, 22) to post-Diwali day (October, 24) were only compared in case of Mumbai (0.56 to 0.56), Delhi (0.50 to 0.73), Pune (0.53 to 0.64) and Varanasi (0.32 to 0.65). Particulate ratio demonstrated a gradual increasing pattern from pre-Diwali to day after Diwali signifying dominance of fine particulate in total particulate loading. However, such trend gradually modified in post-Diwali normal days with gradual increase in coarse particulates.

Entire monitoring period was purposely divided into four segments based on expected aerosol loadings in and around Diwali festival (Fig. 3). Spatial nature of particulate variability has been assessed by processing Terra MODIS Level 2 daily aerosol product using Dark Target algorithm (MOD04_L2; MODIS collection 6), aerosol fine mode fractions (Fig. 4a–e) coupled with true granule images (RGB) and corresponding AOD (collection 51) (Fig. 5a–b). For the current analysis, AOD profile before festive days typically identify IGP and few parts of Deccan province with moderate to high AOD (0.3–0.8) (Fig. 4a). Characteristically, most of the western and IGP region denotes presence of coarser aerosols specifically dust (0.01–0.40) while parts of eastern and south-eastern India signify presence of finer aerosols (0.61–1.00). However, the situation drastically transforms during pre-Diwali day to Diwali-day when intensive fireworks and crackers plunge substantial amount of aerosols throughout the continent (Fig. 4b–c). Terra MODIS aerosol fine mode fraction additionally validate accumulation of finer mode (0.3–1.0) particulate most certainly associated with firework emissions. On subsequent days, although AOD maintain its profile to an extent in the entire subcontinent (AOD: 0.2–0.9), airborne particles however, gradually orient from finer (0.3–1.0) to coarser (0.0–0.7) particulate. Rest of the period appeared to have persistent level of AOD except for Indo-Gangetic plain (Fig. 4e), which characteristically demonstrates incidence of a dense aerosol plume, mostly of finer particulates making the region a global aerosol hotspot. Terra MODIS true granule images (RGB) with corresponding AOD (Fig. 5) additionally validate initial observations by identifying high aerosol loading in post-Diwali days.

3.2. Aerosol surface mass loading over middle IGP

Particulate mass concentration anomalies in respect to normal day's average loading is illustrated in Fig. 6 with associated meteorological variables. Particulate background concentrations (PM_{10}: 129 ± 27.4;

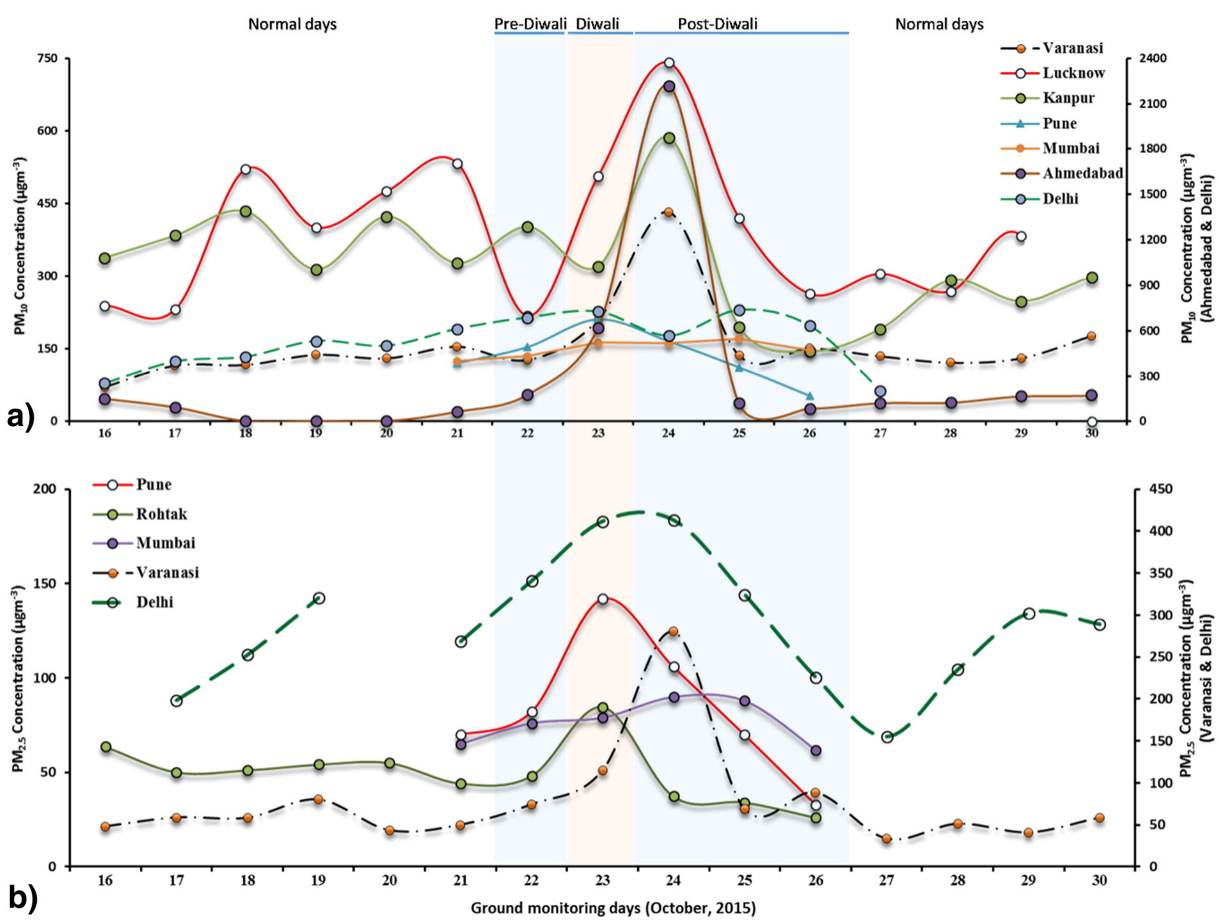

Fig. 2. (a–b) Time series of airborne particulates loading in and around Diwali festival.
Source: cpcb.nic.in.

Fig. 3. Aerosol ground monitoring time frame.

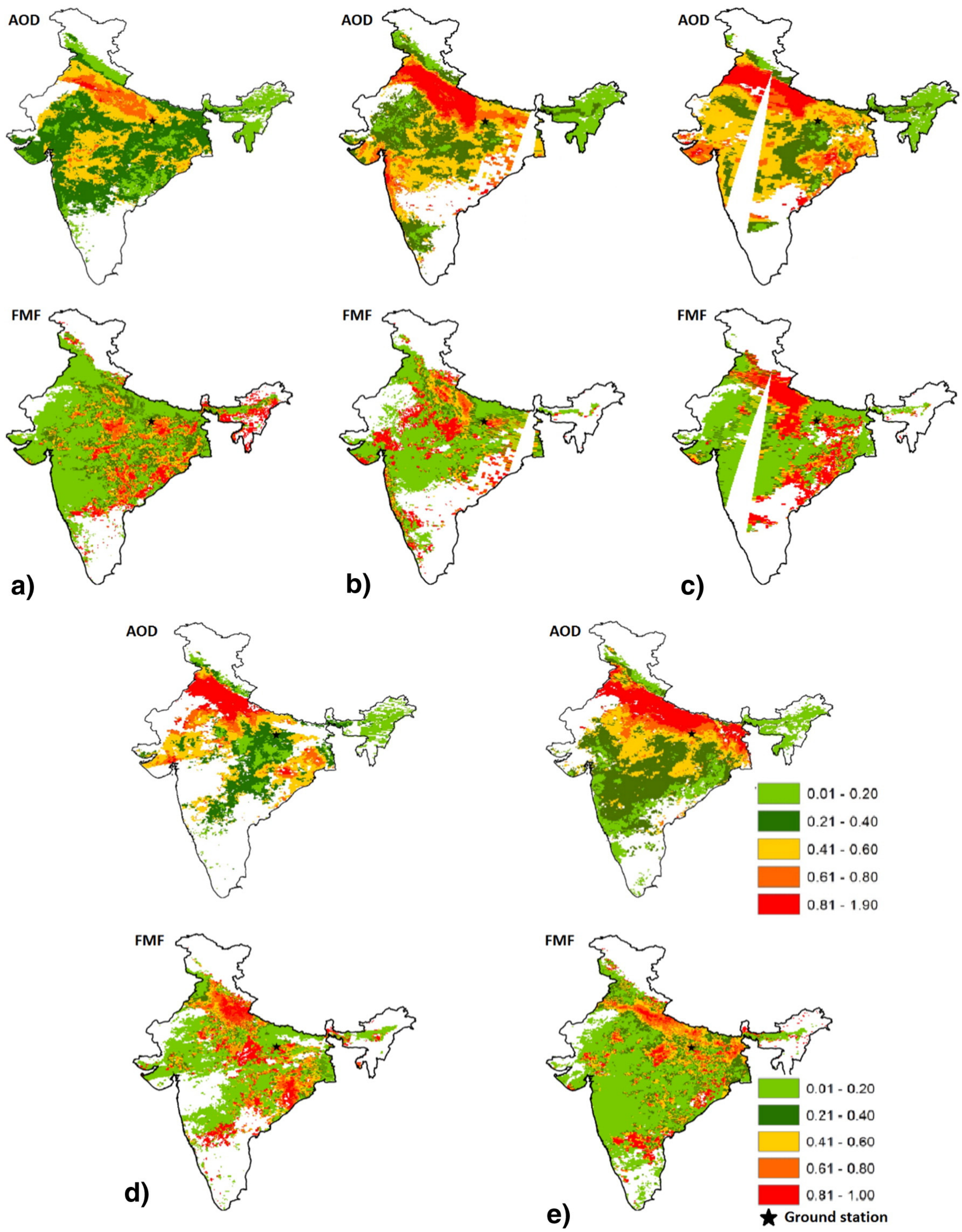

Fig. 4. (a–e) Spatio-temporal distribution of Terra MODIS AOD and corresponding aerosol fine mode fractions (FMF) (a) during pre-Diwali normal days, (b) pre-Diwali day, (c) Diwali day, (d) post-Diwali days and (e) post-normal days.

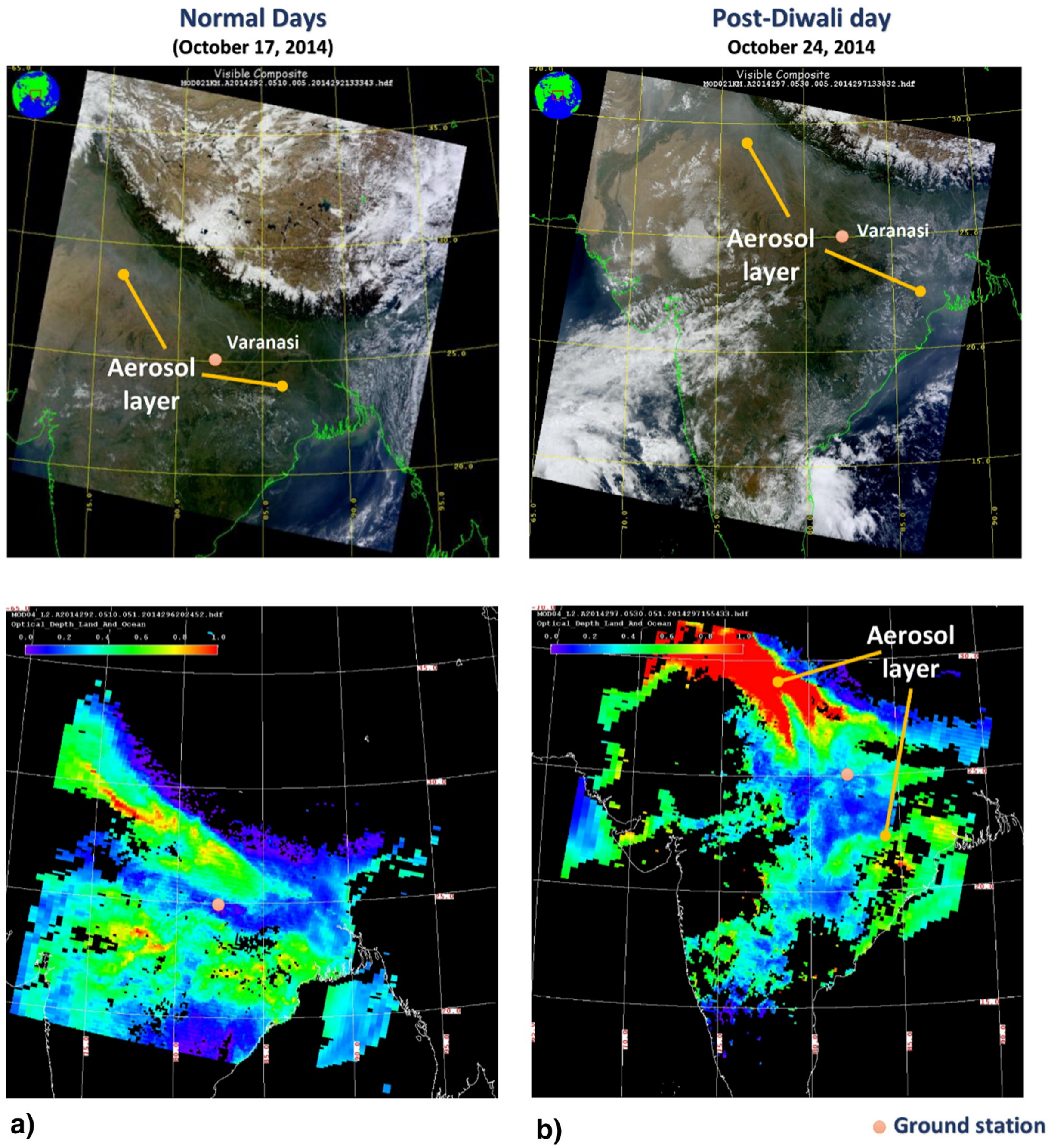

Fig. 5. (a–b) Terra MODIS true colour and corresponding AOD images over Indian sub-continent during different aerosol loading episodes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

$PM_{2.5}$: 52 ± 12.8; TSPM: 175 ± 34.0; BC: 11.4 ± 1.7 μg m^{-3}) were estimated by averaging mass concentrations for all the normal days (10 days, October 16–21 and 27–30, 2014). Fireworks usually commenced one day before the festival and was found to increase in $PM_{2.5}$ of about 42.3% over baseline concentration. Diwali-day specific aerosol loading also exhibited a substantial increase over pre-Diwali concentrations (PM_{10}: 64.1%; $PM_{2.5}$: 55.4% and TSPM: 62.5%). Now, considering Diwali-day specific particulate loading as an isolated event, we found a staggering increase in particulate concentrations over normal days (PM_{10}: 62.8%; $PM_{2.5}$: 121.2% and TSPM: 56.0%). Increment of such mass concentration may possibly associated to emissions of different metal oxides, metal salts, component of black powder and other inorganic species from fireworks. Typical characteristics of Diwali festival is the intensification of crackers and sparkles mostly at late evening to mid night and therefore, it was logical to consider more particulate mass escalation during post-Diwali days. Gas-to-particle conversations additionally substantiate particulate loading during post-Diwali days. The higher proportion increment in $PM_{2.5}$ in comparison to other particulates signifies the presence of more accumulation mode aerosols, a major part of which are mainly of secondary origin. Additionally, prevailing stable atmospheric conditions (stability class E to G; ABL: 404.8–447.9 m; ventilation coefficient: 91.8–170.2 m^2 s^{-1}) during post-Diwali period possibly reduced particle dispersion and consequently increased particulate concentrations in lower troposphere.

3.3. Black carbon mass loading

In contrast to particulate mass variability, atmospheric BC concentration reflects somewhat contrasting pattern (Fig. 6). Within IGP, vehicular emissions and biomass burning mostly constitute BC loading

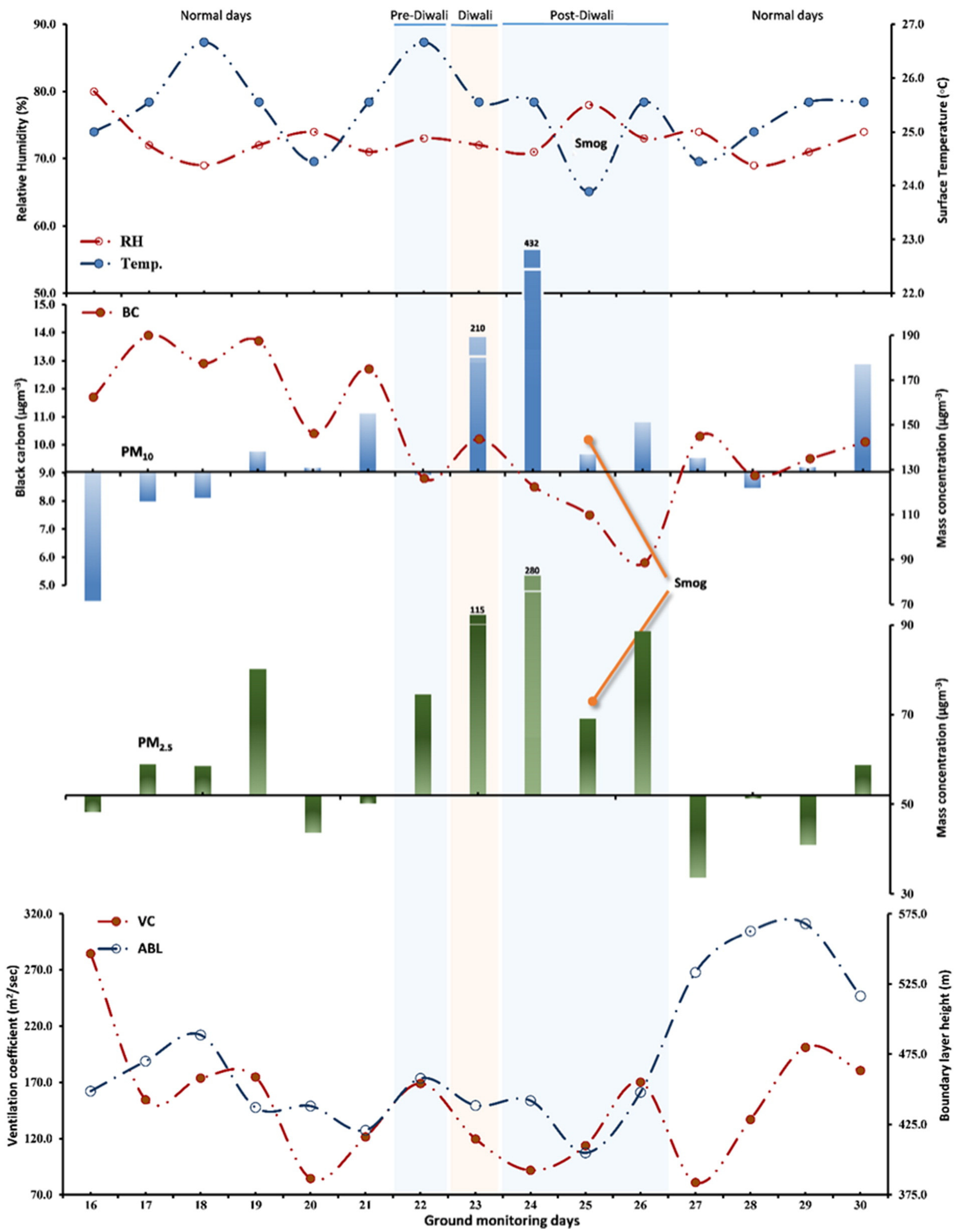

Fig. 6. Variation of 24-h average (1200–1200 h) aerosol and BC mass loading with meteorological variables. Note: PM_{10} and $PM_{2.5}$ mass concentration anomalies were plotted considering normal day's average concentration as baseline.

while for Varanasi, BC profile is mostly constitute by vehicular emissions (Murari et al., 2016). During normal days, 24 h (12:00–12:00) BC mass concentration (mean: 11.4 ± 1.7 μg m^{-3}) represent 21.9% of $PM_{2.5}$ loading. Gradually, BC concentration revealed a declining trend during pre-Diwali (8.8 μg m^{-3}; 22.8% reduction in respect to normal days), Diwali (10.2 μg m^{-3}; 10.5% reduction) and post-Diwali days (7.3 μg m^{-3}; 36.0% reduction) representing an overall 5.0–11.9% of $PM_{2.5}$. Decline in BC loading and its mass ratio with fine particulates possibly relate with reduced traffic emissions. Vehicular density during these festive days drastically reduced which consequently emit less BC

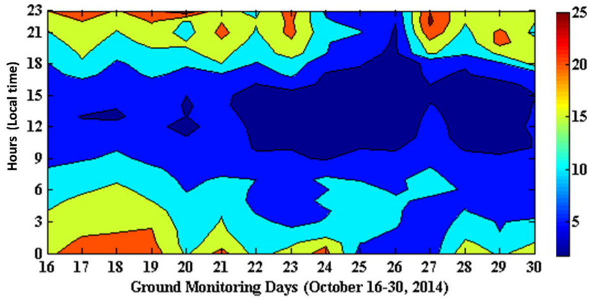

Fig. 7. Time series of BC concentrations (μg m^{-3}) during ground monitoring days.

into regional atmosphere. Similar pattern of reduced vehicular movements and hence decreasing BC concentrations during Diwali-day were also evidenced in Delhi (Sarkar et al., 2010).

Fig. 7 represents the diurnal variation in BC concentration at ground monitoring station. Interestingly, diurnal variation in BC loading was found to be higher in contrast to fireworks specific variation. Throughout the monitoring period, persisting high BC mass loading was observed during 20:00–02:00 (local time) before being gradually reduced during day-time (08:00–18:00). In absence of any specific night-time emission sources, such diurnal variation in BC profile represent effect of reduced boundary layer height which significantly regulate particulate dispersion. However, the trend was found reversing specifically during post-Diwali days where no such mid-night high BC loading was evident. BC-$PM_{2.5}$ mass ratio also revealed significant variation. BC-$PM_{2.5}$ mass ratio for pre- (0.119), Diwali day (0.089) and post-Diwali (0.050) revealed gradual declining trend before being increased at post-Diwali normal days (0.204). This may only be explained in terms of vehicle induced BC emissions, which mostly reduces during festive days before being gradually escalate in post-Diwali period.

3.4. Particulate elemental composition

In recent years demand for fireworks with multiple colours has drastically increased the use of metals as colour developers. Fig. 8 denotes particulate metal concentration anomalies in respect to normal day's baseline concentrations which reasonably indicate firework as potent source toxic chemicals. Most of the elements exhibited identical pattern of incremental peaks during the Diwali-day event before gradually declining on post-Diwali days. An amplified accumulation of firework specific tracers (P as main oxidizer in crackers and S as component of the black powder and firework's propellant; Licudine et al., 2012) in PM_{10} i.e. P (385% increase) and S (341%) during Diwali day (P: 1.11; S: 25.89 μg m^{-3}) in contrast to normal days (P: 0.23; S: 5.87 μg m^{-3}) confirmed the impact of pyrotechnic displays on aerosol chemical composition (Fig. 8a). Ba (green colouring agent), Sr (red), Cu (blue) (Perrino et al., 2011) also exhibited enhanced mass concentrations from normal (Ba: 0.09; Sr: 0.03; Cu: 0.09 μg m^{-3}) to Diwali-day (Ba: 2.29; Sr: 0.09; Cu: 0.29 μg m^{-3}) by 24.7, 2.98 and 3.18

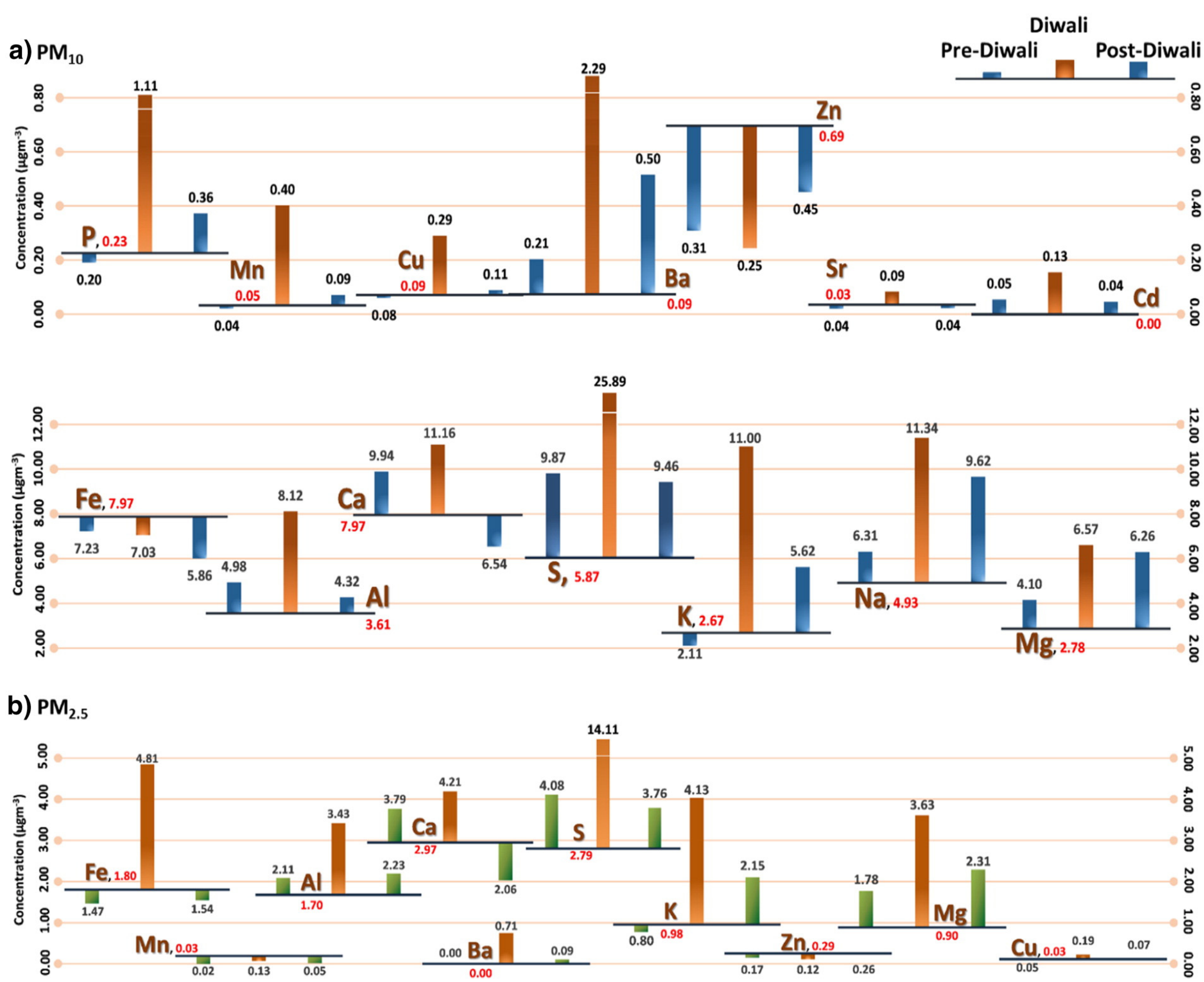

Fig. 8. (a–b) Variation in particulate elemental concentrations during pre-Diwali, Diwali and post-Diwali days. Note: Metal mass concentration anomalies were plotted considering average normal day's concentrations as baseline (in red) to demonstrate variation in respect to background. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

times, respectively. Mg and Ca contribute to regional PM_{10} loading possibly by crustal and road-dust re-suspension (Banerjee et al., 2015; Murari et al., 2015). However, these alkaline earth metals are used as fuel and sparkling agents (Moreno et al., 2007) and hence justify higher concentrations during Diwali-day (Mg: 6.57; Ca: 11.16 μg m^{-3}). However, increase in Mg and Ca possibly offset by certain magnitude through reduced contribution from dust re-suspension attributed to fewer vehicular movements. K and Mn (salts, like $KClO_4$) serve as potent oxidizer of firework mixture was found to be increased (313 & 639%) from background concentration (2.67 & 0.05 μg m^{-3}) to Diwali-day (11.00 & 0.40 μg m^{-3}) before ultimately declined during post-Diwali days (5.62 & 0.09 μg m^{-3}). Cadmium, a highly toxic element was only prominent during festival days (0.04–0.13 μg m^{-3}) before being reduced to non-detectable level in normal days. When firework tracers (K, Ba, Sr, Cd, S and P) were grouped together, they also exhibited stronger correlation coefficients (Ba–Sr: 0.90; K–Sr: 0.82; Cd–Sr: 0.98; Cd–Ba: 0.92; S–P: 0.95) establishing their co-genesis and constitute the effects of firecrackers on deteriorating regional air quality.

Elemental characteristics of finer aerosols ($PM_{2.5}$) were in identical to PM_{10} as observed variation firmly conclude the presence of some unusual chemical species (e.g. S, K, Ba, and Mn) not characterized during normal days (Fig. 8b). Exemplifying, Ba was measured as 0.71 μg m^{-3} on Diwali-day before being sharply reduced on post-Diwali in contrast to below detection level during normal days. Other firework tracers exhibited substantial loading especially during firework activity (K: 322%; Mg: 304%; Cu: 533%; S: 405%). Thus elemental analysis of

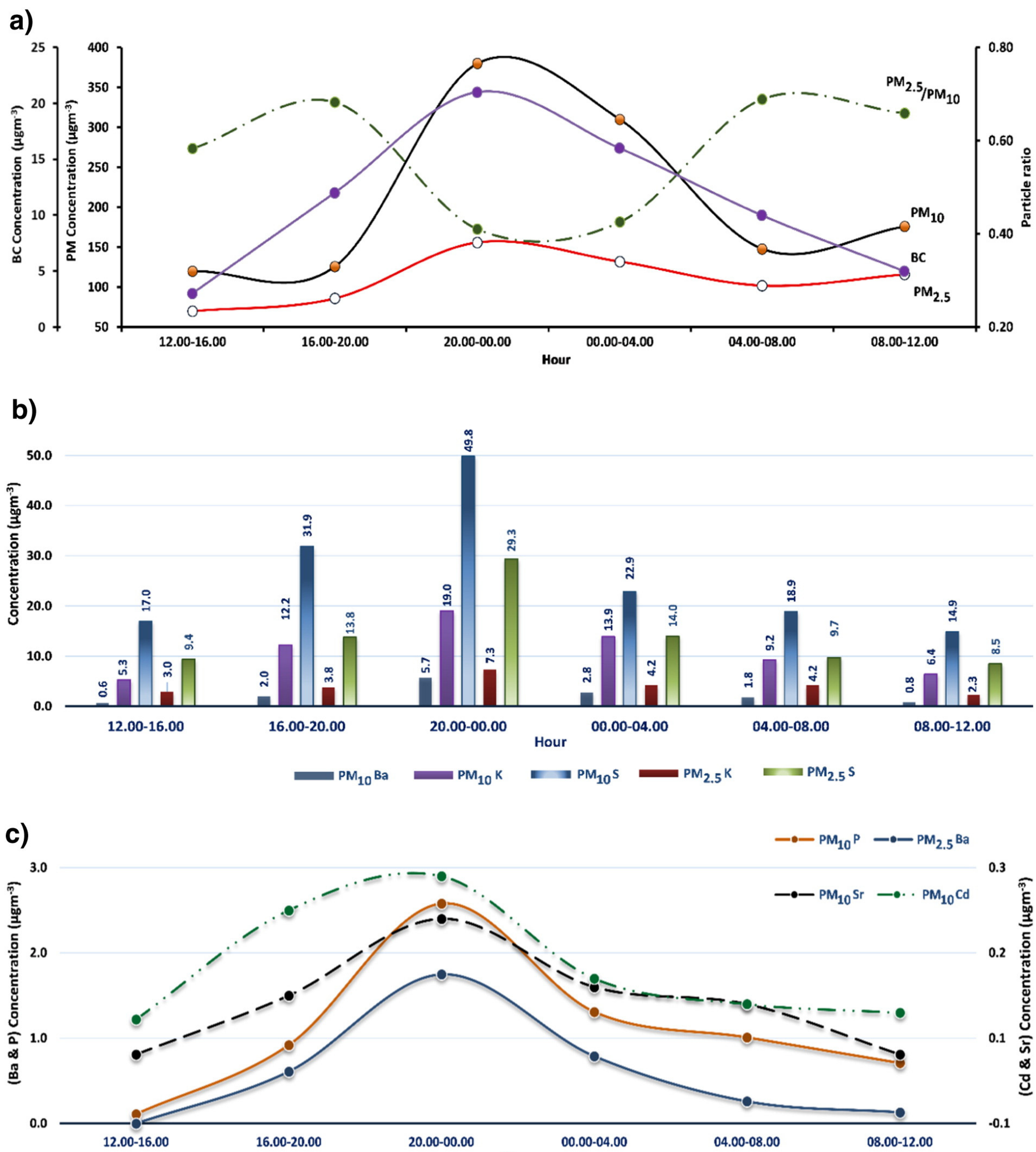

Fig. 9. (a–c) Variation of (a) airborne particulates, particle ratio and BC with (b–c) associated metals (μg m^{-3}) in Diwali specific day.

airborne particulates clearly establish significant deterioration of regional air quality which possibly induce negative health impacts to local population.

3.5. Diwali-day specific variation

Four hourly measurements on Diwali-day were specifically intended to delineate festival specific temporal impacts on the regional atmosphere (Fig. 9). Highest increment (PM_{10}: 202%; $PM_{2.5}$: 82%) in particulate mass concentrations in respect to normal day were observed between 20:00–00:00 h when evidence of fireworks was maximum (Fig. 9a). Afterward particulate concentrations revealed a declining trend before being exposed to daylight (08:00–12:00 h), which facilitates formations of secondary aerosols and thereby increases surface-level aerosol mass concentrations. However, aerosol loading may counter balance by corresponding increase in mixing height and vertical mixing which possibly reduce effective aerosol mass. Incidentally, similar trends were also observed for predominant monitoring stations (like in Delhi and Mumbai, available at cpcb.nic.in) with maximum particle loading at 20:00–00:00 h before being gradually reduced till early morning. Interestingly, during intense firework (20:00–00:00) evolution of coarser particulates were found to be higher than finer ones which may be associated to generation of some specific metals oxides (Ca, Mg and Ba Oxides) and by fireworks debris. Such abrupt increase in coarser particulates eventually reduce average particulate ratio for a time being (till 00:00 h), which otherwise exhibited contrasting pattern as explained in Section 3.1. Black carbon mass loading also exhibited maximum concentration (22.1 μg m^{-3}) during peak fireworks (20:00–00:00 h) with a temporal variation parallel to fine particulate (Fig. 9a). As carbon rich particles exhibit highest abundance within fine range (0.5–1.5 μm; Murari et al., 2016), these findings were expected. Daily averaged (12:00–12:00 h) Delta C values were computed higher (0.70) on Diwali-day compared to normal days (0.32). Elevated Delta C essentially identify sudden change in carbon sources and possibly indicates presence of smoke. MODIS Active fire locations confirmed absence of any such regional biomass burning events and nullified the possibility of adding excess smoke except fireworks (Fig. 1 in the Supplementary data).

Diwali-specific tracer elements were also exhibited higher prevalence (PM_{10}, K: 18.97; Ba: 5.72; Sr: 0.19; Cd: 0.24; S: 49.82 and P: 2.58 μg m^{-3}) specifically during peak Diwali hours (16:00–00:00 h) before declining to minimum during 08:00–12:00 h (Fig. 9c). Variations in elemental compositions follow their respective particulate mass pattern except few. Results strongly support K, Ba, Sr, Cd, S and P to be considered as tracers for firework induced airborne particulate. Fine particulate elemental compositions (Mn: 0.32; Al: 7.11; Ca: 8.83; S: 29.34; K: 7.34; Ba: 1.75 μg m^{-3}) were also reported to have highest during 20:00–00:00 h, many times above (3.0 to 18.0) than their normal day concentrations.

3.6. Variation in aerosol optical properties

3.6.1. Aerosol optical depth (AOD)

Day time daily variations of both space based MODIS-AOD and surface based MTP-AOD (at 550 nm) with corresponding particulate concentrations were plotted in Fig. 10. It is noteworthy to mention that MTP-AOD only corresponds to daytime average while MODIS-AOD indicate columnar aerosol loading specifically during satellite overpass time (12:00–14:00 IST). Therefore, projections of ground level particulate concentrations (24 h average) both with satellite and MTP-AOD will probably be arbitrary, however, was only intended to assess temporal variation in columnar aerosol properties in and around festival period (Devara et al., 2015).

For the current analysis, both MODIS-AOD and MTP-AOD recorded an excellent complement posing a strong correlation (r: 0.77; R^2: 0.59) for the entire monitoring period (Fig. 11). Daily variations of MODIS-AOD ranged from 0.36–1.46 (mean: 0.75 ± 0.30), while MTP-AOD prevailed within 0.44–1.15 (mean: 0.78 ± 0.24). Interestingly, both surface and space based observations demonstrate lower optical depth during festive days which were in contrast to surface based 24 h particulate mass loading. Specifically, in transition from pre-Diwali to Diwali and post-Diwali, surface based particulate exhibit a clear increasing pattern for both PM_{10} and $PM_{2.5}$ (Fig. 6). However, aerosol columnar loading revealed a contrasting pattern both in terms of MODIS-AOD (pre-Diwali: 0.77; Diwali: 0.39; Post-Diwali: 0.45) and MTP-AOD (pre-Diwali: 0.64; Diwali: 0.52; Post-Diwali: 0.48). This ambiguity may well be explained in terms of monitoring period and corresponding atmospheric dispersion potential. All AODs corresponds to day time aerosol loading which was invariably associated with relatively high atmospheric dispersive capability (ABL: 804.2–813.4 m; VC: 132.1–148.1 m^2 s^{-1}) and reduced cracker intensity. Ground level PM mass was obvious illustration of both day and night time reduced dispersive capability (ABL: 438.6–442.0 m; VC: 91.8–119.6 m^2 s^{-1}) additionally burdened with very high firework activities. Exemplifying the Diwali-day, Aqua-MODIS retrieved AOD (0.39) represent particulate columnar concentration at 13:30 local time which to an extent validate the minimum monitored particulate concentrations at 12:00–16:00 (PM_{10}: 120; $PM_{2.5}$: 70 μg m^{-3}). However particulate loading information specifically during satellite overpass time for all monitoring days were not available.

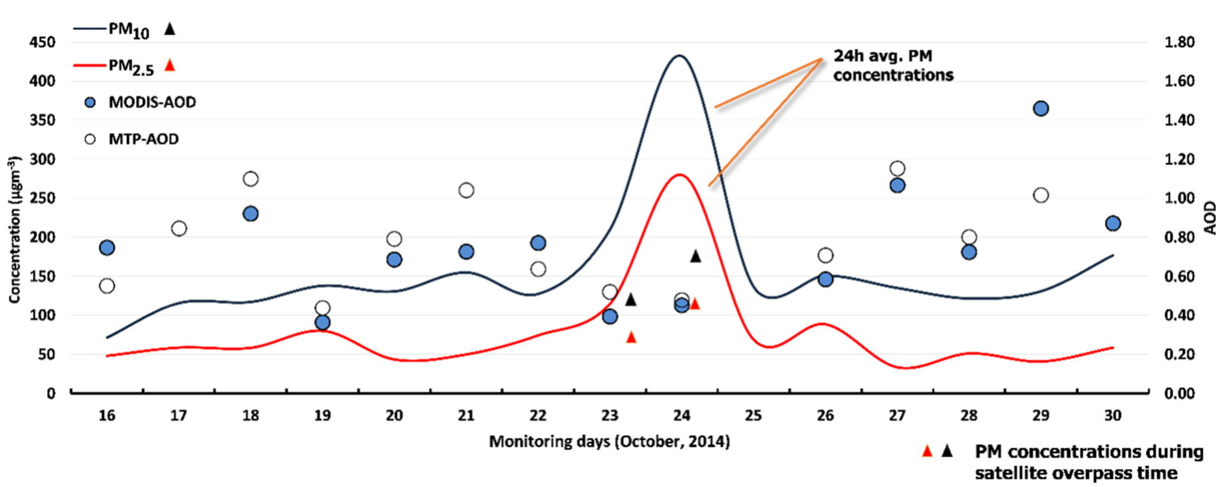

Fig. 10. Daily variations of Aqua MODIS and MTP-AOD in reference to surface aerosol mass loading.

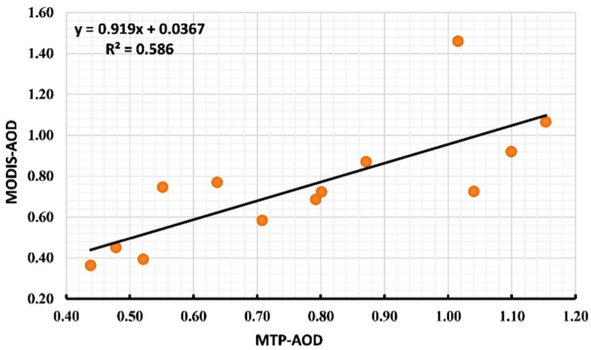

Fig. 11. Aqua MODIS-AOD as function of MTP-AOD.

In absence of any national database of Diwali-specific firework effect on ambient air quality in terms of multi-year ground based particulate concentrations, 10-year Aqua MODIS-AOD were selectively retrieved over Varanasi and analysed. Systematic observation over multiple years for large spatial region is helpful in minimizing the influence of year-to-year variation in particulate background concentrations, variations in magnitude and location of fireworks during festive period. Such analysis was not intended for other regions as information related to satellite retrieved AOD and ground particulate loading is sparse while for Varanasi, a fair bit of association (r: 0.46–0.54) was reported by Kumar et al. (2015b). Fig. 12 denotes 10-year average Aqua MODIS-AOD at Varanasi during Diwali and normal days. To assess statistical significance, one-tailed T-test was considered where a null hypothesis that respective AOD during normal days do not significantly differ against festive period was tested assuming $p < 0.05$ to reject the null hypothesis. The increase in Diwali-day and post-Diwali-specific AOD was found to be insignificant in respect to normal days. It justifies our initial assumption that satellite retrieved AOD only represent aerosol loading during its overpass time which may not identify the relative contribution of fireworks for a region.

3.6.2. Particulate ratio and Angstrom exponent (AE)

Ground observation of airborne particulate on Diwali-day expressed abrupt increase in $PM_{2.5}$ for most of the stations at IGP while PM_{10} exhibit higher proportionate loading only during post-Diwali days (Section 3.1). In order to establish the regional pattern, both $PM_{2.5}$ to PM_{10} ratio were computed against Angstrom exponent (AE) which explains quantitative measures of aerosol size distribution (Fig. 13). AE was in the order of 1.06–1.38 (mean: 1.22 ± 0.09) for the entire period suggesting dominance of fine particulates possibly of anthropogenic origin (like smoke). However, AE was higher during pre-Diwali (mean: 1.26) and Diwali-day (1.26) before slightly reduced to post-Diwali days (1.16). The AE readings well validate surface-level particulate ratio ($PM_{2.5}/PM_{10}$) (mean: 0.47 ± 0.14), which also describes presence of mixed aerosols during Pre-Diwali days (ratio: 0.50 ± 0.13), increase of $PM_{2.5}$ during Diwali-day (0.55) before reducing in post-Diwali days (0.44 ± 0.14). Both AE and particulate ratio in general describes dominance of coarser particulates in post-Diwali to post-normal days, which may possibly due subsequent formation of secondary aerosols and particulate hygroscopic growth.

3.6.3. OMI aerosol index (OMI-AI)

The AI observations derived from the Ozone Monitoring Instrument (OMI) on board Aura were analysed over Indian subcontinent to identify presence and transport of mineral dust and smoke particles during pre-Diwali (October 16–22, 2014) and post-Diwali (October 23–30, 2014) event. Airborne particulates have great significance in attenuating irradiance UV-A spectrum (315–400 nm) either by scattering and absorption and therefore, associate presence or absence of UV-absorbing particulates (specifically soot and desert dust) in the atmosphere. In general, AI is positive for absorbing aerosol and negative AI corresponds to non-absorbing ones (marine aerosols, sulphate aerosols and elevated clouds) while > 1.0 is typical to desert dust and smoke particles (Torres et al., 2007). For the present analysis, OMI-AI mean distribution (Fig. 14) critically identified the spatial pattern of aerosol loading especially induced by pyrotechnic activity. Before fireworks, entire continent was dominated by presence of absorbing aerosol (OMI-AI: 0.60–1.20) (dust or smoke). However, some parts of upper IGP, Rajasthan, Delhi, western Uttar Pradesh, Lahore, Punjab plain in Pakistan and Potwar plateau appear distinct with more pronounced presence of absorbing aerosols (1.60–2.80). Post-Diwali OMI-AI scenario clearly distinguished enhanced level of OMI-AI (0.80–1.40) for the entire continent, while it has more prominent (1.90–3.20) in upper and middle-IGP, parts of Gujarat and central India. The OMI-AI profile virtually established the effect of cracker and fireworks on gradual accumulation of absorbing aerosol over Indian subcontinent, however, it cannot certify individual aerosol type within a definite group. Therefore, in order to discriminate absorbing aerosol sub-types, altitudinal profile were further analysed by means of cloud-aerosol lidar data. Aerosol vertical profile is extremely critical for the surface monitoring station as long-range transboundary particulates are supposed to be a significant contributor of regional particulate loading (Kumar et al., 2015b; Sen et al., 2014).

3.6.4. Aerosol vertical profile

The AQUA-MODIS and AURA-OMI sensor based observations (Figs. 4 & 14) on aerosol profile were used to identify presence of diverse UV-absorbing aerosols (specifically soot and desert dust) which was required to be isolated based on aerosol subtypes. Aerosol vertical profile

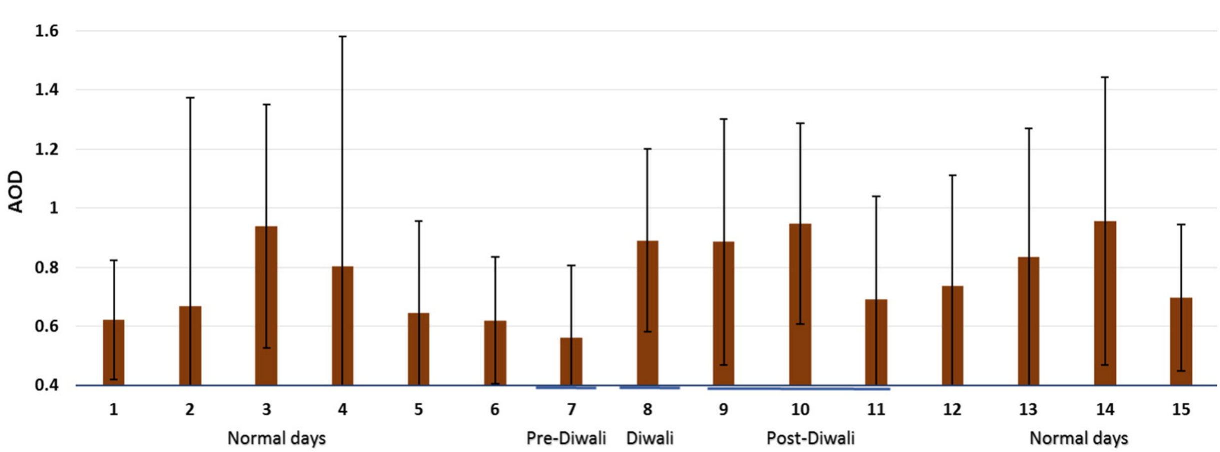

Fig. 12. Variation in 10-year average Aqua MODIS-AOD during pre- and post-Diwali days.

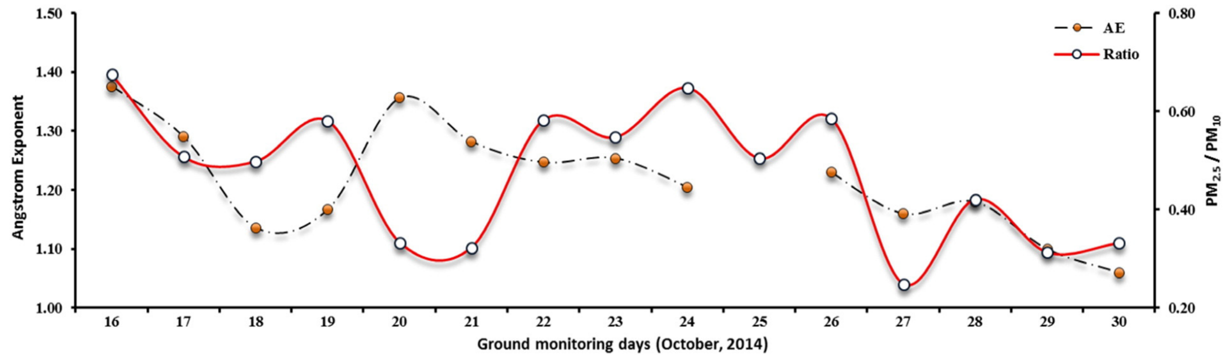

Fig. 13. Daily variations of Angstrom exponent in reference to airborne particulate ratio.

was therefore obtained for best suited CALIPSO overpass path for the selected transect and both aerosol vertical feature mask and subtype products were compared (Fig. 15) with corresponding aerosol extinction coefficient (Fig. 16). The entire CALIPSO profiles essentially validate both MODIS-AOD and OMI-AI observations by identifying huge aerosol mass (specifically dust and polluted dust representing dust mixed with biomass burning smoke) in Indian subcontinent with a change in aerosol type supposed to be influenced by Diwali-specific firework emissions.

In pre-Diwali event, aerosol vertical feature mask (VFM) (Fig. 15a) identified persistence of aerosol layers (high confidence) up to an altitude of 3.0–3.8 km with a peak in extinction coefficient (0.24 km^{-1}) at about 0.45 km. From the surface to 3.8 km, depolarization ratio (δ_a) slowly varied in the range of 0.16–0.30. However, presence of aerosol layer were found gradually declining above 2.0 km with a corresponding low magnitude extinction coefficient (<0.10 km^{-1}). Entire profile abruptly changed during pre-Diwali day with presence of very thick low altitude aerosol layer (0.40–0.50 km) characterized with high extinction coefficient (0.54 km^{-1}) at 0.45 km with corresponding high δ_a (0.15–0.40) (figure not shown). Enhanced extinction coefficient and corresponding δ_a invariably suggest association of polluted continental and smoke particles emitted through pre-Diwali fireworks. To a certain extent Diwali-day specific CALIPSO VFM profile (Fig. 15b) resemble pre-Diwali (Fig. 15a) while aerosol subtype product (Fig. 15e-f) clearly distinguished both by association of more absorbing type aerosols (polluted continental, polluted dust and smoke). Interestingly, aerosol type did not exhibit significant modifications during post-Diwali (Fig. 15g) except elevation of aerosol layer. During post-Diwali two relatively large peaks in aerosol extinction coefficient were seen at 0.2 km (0.56 km^{-1}) and 0.89 km (0.34 km^{-1}) associated with large δ_a values (0.15–0.42) which suggest relatively large proportion of non-spherical particles in total aerosol loadings. Presence of absorbing aerosol layer in such high elevation can easily trigger atmospheric inversion and induce radiative forcing. The whole system further transmutes to initial condition (prevalence of dust and polluted dust) within days leaving behind a thick layer of aerosols which essentially validate both MODIS-AOD (Fig. 4e) and OMI-AI observations (Fig. 14b). The entire Level-2 CALIPSO profile established presence of huge quantity of high altitude dust, polluted dust and polluted continental which only transmutes during Diwali through association of smoke and polluted continental and potentially modify the regional atmospheric dynamics and ultimately regional climate.

4. Conclusions

Fireworks induced modification in aerosol spatio-temporal characteristics were investigated using both remote sensing and ground based techniques. The top to bottom approach of analysing the spatial variations of fireworks induced aerosol characteristics at national level to their respective properties at a single station in middle IGP is novel in many aspects. Diwali-specific observations at middle IGP have larger implications on associating fireworks induced particle pollution and human health. Inclusion of these observations should improve regional

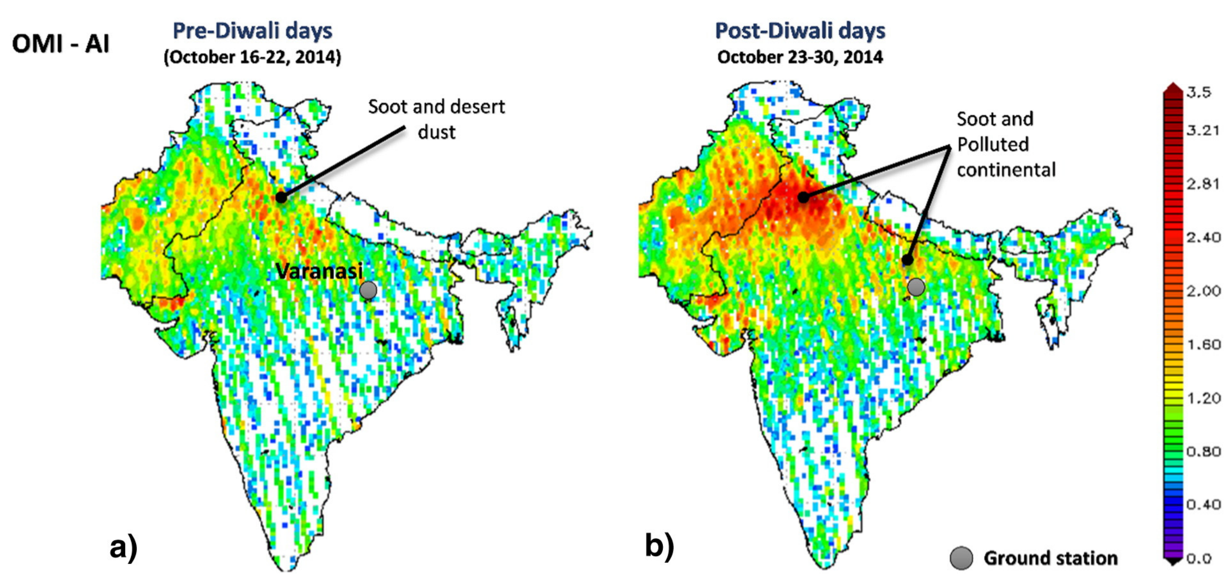

Fig. 14. (a–b) OMI-aerosol index for (a) pre- and (b) post-Diwali days identifying soot particle.

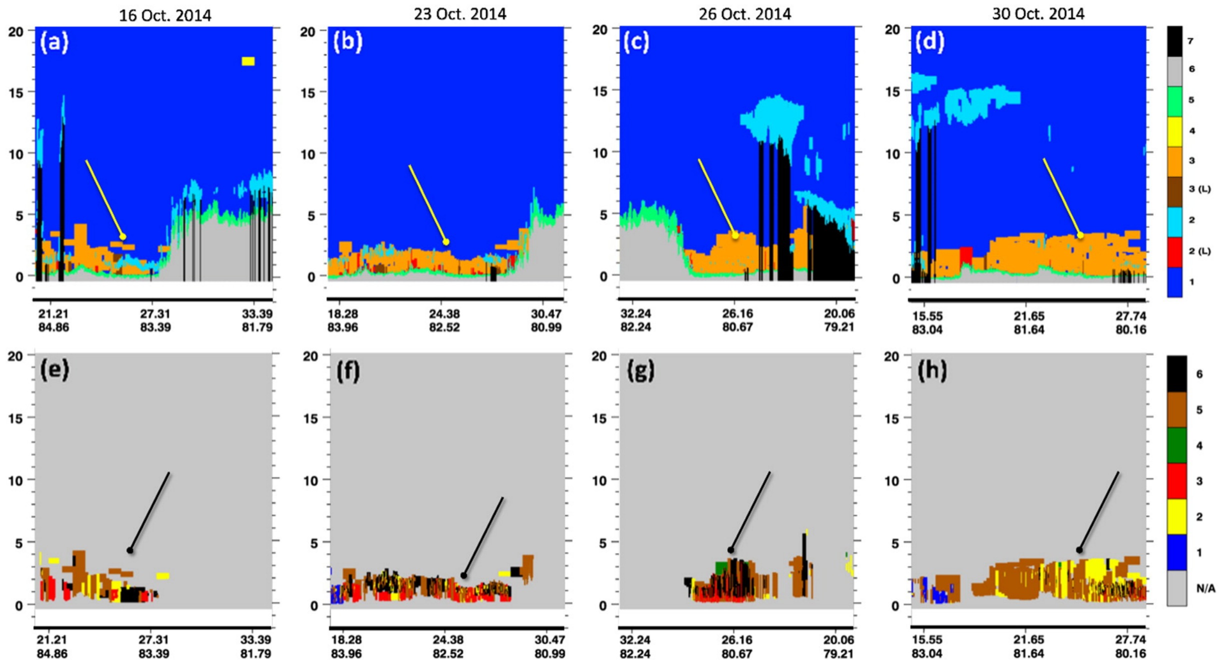

Fig. 15. (a–h) CALIPSO profile for the entire Diwali episode. (a–d) Vertical feature mask, (e–h) aerosol subtypes. Note: Black line represents selected coordinate for Varanasi. VFM: 1 — clear air, 2 — cloud, 3 — aerosols, 4 — stratospheric layer, 5 — surface, 6 — subsurface, 7 — totally attenuated, L — low/no confidence. Aerosol subtypes: 1 — clean marine, 2 — dust, 3 — polluted continental, 4 — clean continental, 5 — polluted dust, 6 — smoke.

air quality model forecasting. The principal outcomes uncovered from the present research may be summarized as follows:

(a) Sudden upsurge of aerosol concentrations were observed throughout India during festive days. Diwali-day specific particulate mass loading reached as high as 2220 μg m^{-3} (Ahmedabad) and 412 μg m^{-3} (Delhi), much higher than the national standards.
(b) Concurrent employment of space-borne passive sensor recognized IGP and few parts of Deccan province with moderate to high AOD (0.3–0.8) during normal days which successively modified to very high (0.41–1.80) during pre-Diwali to Diwali-episode.
(c) Aerosol surface mass loading at middle IGP revealed a sharp increase in particulate loading (PM_{10}: 62.8%; $PM_{2.5}$: 121.2% and TSPM: 56.0%) during festival days compared to their background concentrations.
(d) Accumulation of finer aerosols attributed to nucleation and gas to particle conversion on subsequent days which lead to additional increase in coarser particulates.
(e) On the contrary, on both pre- and post-Diwali day's BC profiles showed a declining pattern from their background concentrations (−22.8% and −36.0% respectively) possibly associated to reduced vehicular emission.
(f) An increase in firework specific tracers like P (385%), S (341%), K (313%) in PM_{10} mass along with Ba (24.7 times), Sr (2.98 times) and Cu (3.18 times) expressed potential harmful effects of firework. $PM_{2.5}$ mass exhibited similar pattern of elemental characteristics with substantial increase in firework specific tracers (Mg: 322%, Cu: 533% and S: 405%) during festive days.
(g) Uniform pattern of highest aerosol mass loadings, BC and firework tracer elements was observed during 20:00–00:00 h on Diwali-day which coincide with maximum fireworks intensity.

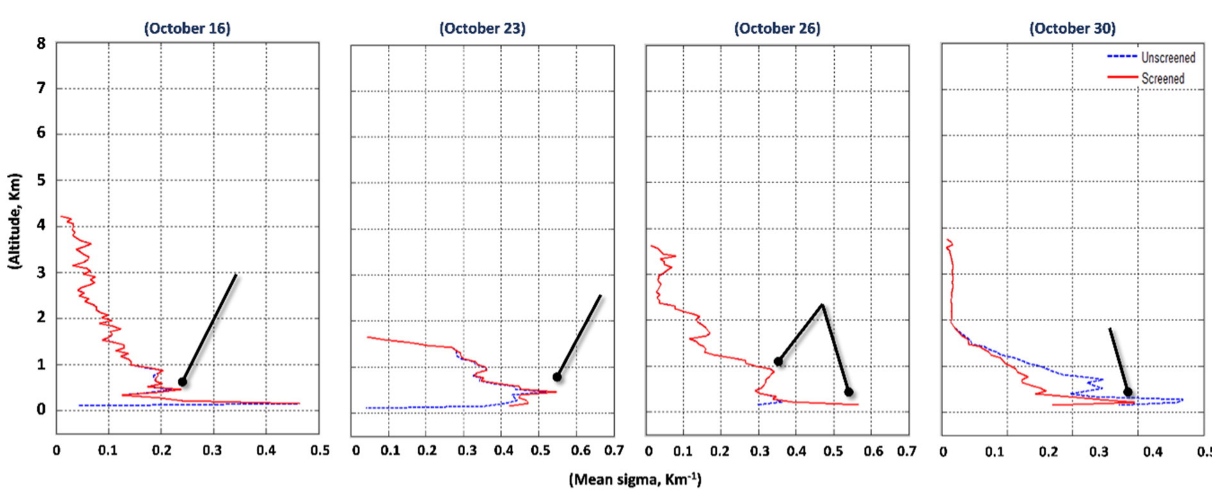

Fig. 16. Vertical profiles of aerosol extinction coefficient (red lines) for entire Diwali-specific firework episode (black line shows point of interest). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(h) OMI-aerosol index found dominance of absorbing aerosol (OMI-AI: 0.60–1.20) over entire Indian subcontinent which further enhanced (OMI-AI: 0.80–1.40) during post-festive days, more prominently in upper and middle-IGP and parts of Gujarat and central India.
(i) CALIPSO altitude-orbit-cross-section profiles recognized the presence of high altitude (3.0–3.8 km) dust, polluted dust and polluted continental over middle IGP which gradually modified during festive days with association of smoke and polluted continental aerosol.
(j) Aerosol extinction coefficient and depolarization ratio profiles revealed existence of multiple and elevated non-spherical aerosol layers consisting mixtures of polluted continental, polluted dust and smoke.

Acknowledgements

Airborne particulate monitoring was financially supported by the University Grants Commission, New Delhi (F. No. 41-1111/2012, SR) and SERB, Department of Science and Technology, New Delhi (F. No. SR/FTP/ES-52/2014) while BC monitoring was funded by the Indian Space Research Organization, Thiruvananthapuram (ARFI: P-32-13). Except Varanasi, all particulate loading information is the courtesy of CPCB, Govt. of India. The MODIS-AOD and OMI-AI are courtesy of NASA's Earth–Sun System Division, distributed by LAADS Web and GES DISC. The CALIPSO data were obtained from the Atmospheric Science Data Center at NASA Langley Research Center. The authors also acknowledge the NCEP/NCAR reanalysis team for providing synoptic meteorological data. The guidance and cooperation provided by the Director, IESD-BHU is also acknowledged. The authors also appreciate the reviewer's comment to improve the article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.atmosres.2016.05.014.

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