Atmospheric Pollution Research xxx (2016) 1–12

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Atmospheric Pollution Research

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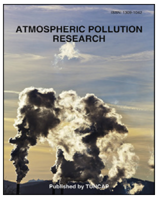

Original article

Spatial variability in ambient atmospheric fine and coarse mode aerosols over Indo-Gangetic plains, India and adjoining oceans during the onset of summer monsoons, 2014

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ARTICLE INFO

Article history:
Received 12 August 2015
Accepted 3 January 2016
Available online xxx

Keywords:
$PM_{2.5}$
PM_{10}
Cluster analysis
PSCF
CWT

ABSTRACT

Enhanced transport of dust with the prevailing mid to upper tropospheric westerly winds from arid regions in South-west Asia and North-west India into the Indo-Gangetic Plain (IGP) and the influx of marine aerosol from the Arabian Sea (AS), Tropical Indian Ocean (TIO) and Southern Bay of Bengal (SBoB) into India along with the low level south-west wind flow during the onset of the South-west (SW) monsoon, 2014 was observed in campaign mode. Ambient airborne particulates ($PM_{2.5}$ and PM_{10}) were collected at 9 sites in and around IGP, India, viz. Patiala, Delhi, Lucknow, Varanasi, Giridih, Kolkata, Darjeeling, Bhubaneswar and Nagpur; over AS, TIO and SBoB providing a glimpse into the aerosol loading and its transport mechanisms. The highest average $PM_{2.5}$ (61.8 ± 18.6 μg m^{-3}) and PM_{10} (182.2 ± 58.0 μg m^{-3}) mass concentrations were recorded at Delhi (upper IGP) and Lucknow (middle IGP) respectively. Average $PM_{2.5}$ (18.1 ± 10.1 μg m^{-3}) and PM_{10} (39.6 ± 15.8 μg m^{-3}) levels recorded over the open oceanic regions in AS, TIO and SBoB were much lower than those observed over the land stations and the average $PM_{2.5}$ recorded over coastal AS and SBoB (49.1 ± 28.7 μg m^{-3}).

Cluster analysis, Potential Source Contribution Function (PSCF) and Concentration Weighted Trajectory (CWT) analysis portray that $PM_{2.5}$ and PM_{10} levels at the land stations were influenced by weak to moderate contributions from AS, BoB, the arid South-west Asia and North-west India, peninsular India and from the polluted IGP region.

1. Introduction

India along with its adjoining oceans, i.e. Bay of Bengal (BoB), Arabian Sea (AS) and tropical Indian Ocean (TIO), have a unique weather pattern, wherein they are subjected to the seasonal reversal of winds associated with the Indian monsoon system and their accompanying distinct air

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Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.

http://dx.doi.org/10.1016/j.apr.2016.01.001

Please cite this article in press as: Sen, A., et al., Spatial variability in ambient atmospheric fine and coarse mode aerosols over Indo-Gangetic plains, India and adjoining oceans during the onset of summer monsoons, 2014, Atmospheric Pollution Research (2016), http://dx.doi.org/10.1016/j.apr.2016.01.001

masses (continental and marine). The North-east or winter monsoon occurs during the boreal winter and is associated with weak low level north-easterly winds blowing from the Asian land mass towards BoB and Northern Indian Ocean (NIO), leading to the long range outflow of continental pollutants onto both coastal and open oceanic marine regions (Nair et al., 2007; Li and Ramanathan, 2002; Ramanathan et al., 2001a). The South-west or summer monsoon occurs during the boreal summer months and is associated with the advection of strong moisture laden marine air masses from the southern hemisphere, crossing the equator and blowing from a south-westerly direction towards the subcontinent. The summer monsoon provides maximum rainfall over the Indian mainland, accounting for up to 80% of the annual mean precipitation over most regions of India, making it the most important source of water for drinking, irrigation and power generation purposes (Bollasina et al., 2011).

There have been numerous observational campaigns conducted over various locations in India and adjoining oceans, either during the winter monsoon or during pre-monsoon seasons in the last two decades, focusing on the sources, transport pathways and physico-chemical, and optical properties of atmospheric particulates. A few notable examples are: INDOEX (Ramanathan et al., 2001a); Land Campaign-I over peninsular India in February, 2004 (LC-I: Moorthy et al., 2005), Land Campaign-II over IGP in December, 2004 (LC-II: Ramachandran et al., 2006; Tare et al., 2006; Nair et al., 2007), Integrated Campaign of Atmospheric Aerosols, gases, and Radiation Budget during March–May, 2006 (ICARB: Nair et al., 2008; Babu et al., 2008; Reddy et al., 2008; Moorthy et al., 2008), Winter ICARB during December, 2008–January, 2009 (W_ICARB: Moorthy et al., 2010; Kharol et al., 2011; Sharma et al., 2012a,b; Saxena et al., 2014a,b). However, with the exception of the Arabian Sea Monsoon Experiment (ARMEX: Vinoj and Satheesh, 2003) and some others, e.g. Madhavan et al. (2008); there is a noted scarcity of studies focusing on the aerosol properties over India and adjoining oceans during the summer monsoon, due to the unfavorable overcast sky conditions as well as the difficulty in conducting oceanic observations due to rough, choppy seas during that season. Recently, a field campaign was conducted during the 2013–14 winter monsoon by our group; involving the simultaneous collection of ambient aerosols ($PM_{2.5}$ and PM_{10}) from multiple locations across India and the BoB, with the objective of investigating the sources leading to the outflow of continental aerosols into the BoB (Sen et al., 2014). Several model studies have shown that dust transport from Sahara desert as well as Thar desert can alter distribution of particulate matter over Indo Gangetic Plain (IGP). Study of chemical characteristics of particulate matter could give an idea how transported particulate matter recombines with anthropogenic sources of IGP.

This current study involves sampling of ambient $PM_{2.5}$ and PM_{10} aerosols across multiple locations in India, the BoB, AS and TIO; carried out with the objective of picturing the transport of mineral dust with the westerly flow from the arid South-west Asian region into the IGP and the influx of marine aerosols from the AS and TIO into India along with the low level south-westerly wind flow prior to and during the onset of the summer monsoon as well as studying their interaction with the prevalent background continental aerosols over the land stations.

2. Data and methodology

2.1. Sampling locations

2.1.1. Land sites

The IGP is a vast expanse of fertile plain encompassing most of northern and eastern India, bound by the Himalayas to the north, Vidhyan Satpura ranges as well as the Chota Nagpur Plateau to the south, BoB to the east and the AS and Thar Desert to the west. It supports about ~40% of the Indian population (Nair et al., 2007), making it one of the most densely populated and heavily polluted regions in the world (Dey and Di Girolamo, 2010; Srivastava et al., 2011), subject to rapid industrialization, urbanization and a rampant increase in vehicular traffic and biomass burning to meet the needs of the growing population and resulting in a massive anthropogenic aerosol loading over it (Ram and Sarin, 2010; Rengarajan et al., 2007; Sharma et al., 2015a). The sampling sites situated in the IGP, viz. Patiala (30°21′ N, 76°22′ E), Delhi (28°38′ N, 77°10′ E), Lucknow (26°52′ N, 80°56′ E), Varanasi (25°15′ N, 82°59′ E) and Kolkata (22°34′ N, 88°21′E) are ideal for analyzing the alteration in the ambient aerosol composition due to indiscriminate biomass and fossil fuel combustion (Sharma et al., 2015a). These are located in the hearts of megacities having a high population density and are subjected to pollution from heavy roadside traffic as well as many small to large scale industries surrounding them. Bhubaneswar (20°17′ N, 85°49′ E) and Kolkata are coastal stations bordering the BoB, along the east coast of India. Giridih (24°10′ N, 86°17′ E) is located on the Chota Nagpur plateau and coal mining is one of its major industries. Nagpur (21°08′N, 79°05′E) is located in the Deccan plateau and serves as a representative station for central India. Ambient atmospheric conditions at these locations are also observed to vary greatly; from semi polluted Himalayan air at Darjeeling (27°01′ N, 88°15′ E), to highly polluted conditions prevailing in large metropolitan cities like Kolkata, Delhi, Varanasi and Lucknow. The co-ordinates of the sampling sites and duration of sampling carried out at each of the sites are summarized in Table 1.

2.1.2. Cruise details

Two scientific cruises were conducted during campaign, with the objective of covering both remote and coastal marine regions. ORV Sagar Kanya (SK-313) set sail from Mormugao Port, Goa on 3rd June, 2014; proceeded in coastal waters close to the west coast of India, sailed around Sri Lanka and carried out a few transects in Southern BoB (SBOB) near the Indian coastline, before finally arriving at Chennai Port, Chennai on 21st June 2014. ORV Sagar Nidhi (SN-85) set sail from Mormugao Port, Goa (15°24′ N, 73°48′ E) on 30th May, 2014 and proceeded to South West Tropical Indian Ocean (SWTIO) covering mostly remote marine regions in AS and SWTIO, the southernmost extent of the track being 8° S, 69° E (SWTIOTS), where the ship remained stationary for 10 days to facilitate time series observations, following which it proceeded to sail around Sri Lanka, cover a small extent of SBOB and finally call at Chennai Port, Chennai (13°4′ N, 80°17′ E) on 27th June, 2014.

Locations of the sampling sites along with the cruise tracks of SK-313 and SN-85, shown as solid lines over oceanic regions, have been plotted in Fig. 1.

2.2. Sample collection

Ambient particulates with aerodynamic diameters ≤ 2.5 μm ($PM_{2.5}$) and ≤ 10 μm (PM_{10}) respectively were collected simultaneously on pre-combusted and pre-weighed QM-A quartz fiber filters (Whatman, UK) using mass-flow controlled High Volume Samplers (HVS) installed on elevated surfaces free from obstacles. The QM-A filters used in the study were pre-combusted in a muffle furnace for 6 h at 550 °C and desiccated for a period of 48 h prior to sample collection to eradicate all traces of impurities and moisture. Simultaneously, $PM_{2.5}$ samples were collected from the marine atmospheric boundary layer (MABL) of coastal AS and SBoB utilizing a HVS installed onboard ORV Sagar Kanya (SK-313), while $PM_{2.5}$ and PM_{10} samples were collected from the MABL of AS, SWTIO and SBOB via two HVS installed onboard ORV Sagar Nidhi (SN-85). The samplers utilized onboard both vessels were installed on the upper

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Table 1
Description of sampling sites, period and instruments operated.

Sites	Coordinates		Instruments operated		Sampling duration
	Latitude (N)	Longitude (E)	PM$_{2.5}$	PM$_{10}$	
Patiala	30°21′	76°22′	IPM-FDS-2.5μ/10μ	n.a	3rd June–27th June
Delhi	28°38′	77°10′	APM550	APM550	2nd June–27th June
Lucknow	26°52′	80°56′	APM550	APM-460NL	1st June–27th June
Varanasi	25°15′	82°59′	IPM-FDS-2.5μ/10μ	APM-460BL	2nd June–26th June
Giridih	24°10′	86°17′	APM550	n.a	1st June–30th June
Kolkata	22°34′	88°21′	APM550	APM-460BL	24th May–30th June
Darjeeling	27°01′	88°15′	APM550	APM-460BL	26th May–29th June
Bhubaneswar	20°17′	85°49′	n.a	APM-460NL	1st June–25th June
Nagpur	21°08′	79°05′	APM550	APM-460BL	5th June–19th June
SK-313			APM550	n.a	7th June–21st June
SN-85			APM550	APM-460NL	30th May–26th June

n.a: not available.

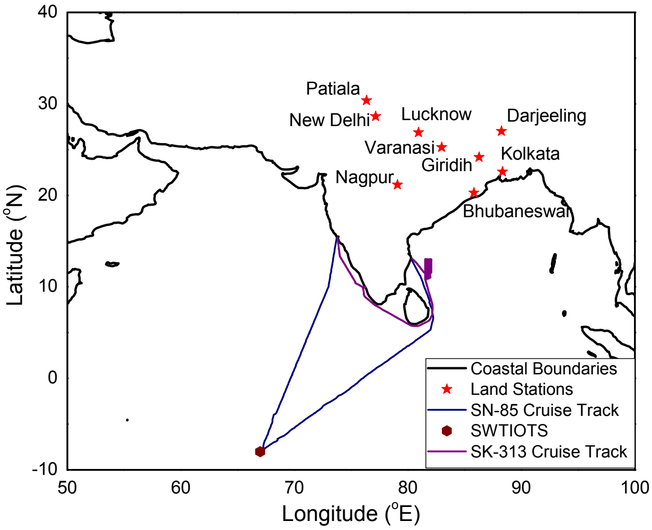

Fig. 1. Map of sampling sites and cruise tracks.

front deck at a height of ~12 m from sea surface. During ship stoppage, care was taken to position the ship in such a way that the wind did not arrive at the samplers from the aft of the ship, thereby avoiding any contamination from the ship's exhaust. The details of the make and model of the instruments operated at the land sampling sites and along the cruise tracks as well as their respective periods of sampling are available in Table 1.

Ambient fine particulate matter (PM$_{2.5}$) was collected at the sampling stations using fine dust samplers (APM 550, IPM-FDS-2.5μ/10μ). Similarly, respirable dust samplers (Models: APM 550, APM-460BL, APM-460NL and PEM-HVS-8NL) were utilized for the collection of ambient respirable suspended particulate matter (PM$_{10}$). The impactors used in all the samplers were designed according to the United States Environmental Protection Agency (USEPA) regulations. The fine dust samplers collected air-borne PM by drawing in ambient air at a constant flow rate of 1 m^3 h^{-1}. PM$_{10}$ aerosols were separated by an impactor, following which PM$_{2.5}$ aerosols were separated by a WINS-Anderson Impactor and deposited on to a 47 mm QM-A filter placed below the impaction surface. The respirable dust samplers drew in ambient air at flow rates ranging from 0.9 to 1.4 m^3 h^{-1}, following which PM$_{10}$ aerosols were separated out by cyclones and deposited on to 20 × 25 cm^2 QM-A filters. A detailed description of the HVS samplers operated during the campaign is reported by Sen et al. (2014).

The filters were kept in desiccators for at least a period of 48 h following sample collection and prior to final weight measurement to eradicate any absorbed moisture during sampling. PM mass loading was ascertained by gravimetric analysis. PM mass was measured by determining the difference between the final and initial weights of the filter and its concentration was determined by dividing it by the volume of air sampled for the duration of the experiment.

2.3. *Prevailing meteorology*

Mean synoptic wind patterns at 1000 hPa and 700 hPa over the Indian subcontinent and adjoining oceans for the month of June, 2014, using ERA-Interim reanalysis data, are presented in Fig. 2a and b. The near-surface wind pattern (1000 hPa) is similar to that known to prevail during the summer monsoon; i.e. strong winds from the southern hemisphere cross the equator and blow from a south-westerly direction across AS and BoB towards the subcontinent. On the contrary, the wind pattern at elevated altitudes (700 hPa) reveals the advection of air masses from bordering arid regions in South-west Asia, Arabian Peninsula and north western India (Thar Desert) into the IGP, peninsular India and into the BoB. The prevalent wind regimes during the study period point towards increased aerosol loading over the subcontinent through the transport of desert dust from Arabia and North Africa at elevated altitudes by westerly winds and advection of sea salt aerosols by strong south-westerly near surface winds (Li and Ramanathan, 2002).

The meteorological conditions, viz. Temperature (°C), Pressure (mb), Relative Humidity (%), Rainfall (mm), Wind Speed (m s^{-1}) and Wind Direction (°N); prevailing at the sampling sites and along the cruise tracks during the study period are depicted in Table 2. Meteorological data was assimilated from automatic weather stations (AWS) in constant operation at each of the sampling sites and on the bow of the upper decks (height ~10 m.a.s.l) on both ORVs. They were clear of any obstructions to ensure legitimacy of the meteorological data recorded. Amongst the land stations, Patiala (38.1 ± 4.9 °C) and Darjeeling (22.7 ± 0.9 °C) recorded the highest and lowest average ambient temperatures respectively. Kolkata (997.6 ± 1.4 mb) and Bhubaneswar (993.7 ± 1.5 mb) recorded the highest pressure while Darjeeling registered the highest RH % (96.5 ± 3.0%). The strongest and weakest average winds were noted at Bhubaneswar (3.3 ± 0.8 m s^{-1}) and Patiala (0.1 ± 0.3 m s^{-1}) respectively. Both apparent wind speed and true wind speed encountered on the ORVs were recorded by their AWS, after taking into account the ship speed and direction. Winds encountered by ORV Sagar Kanya

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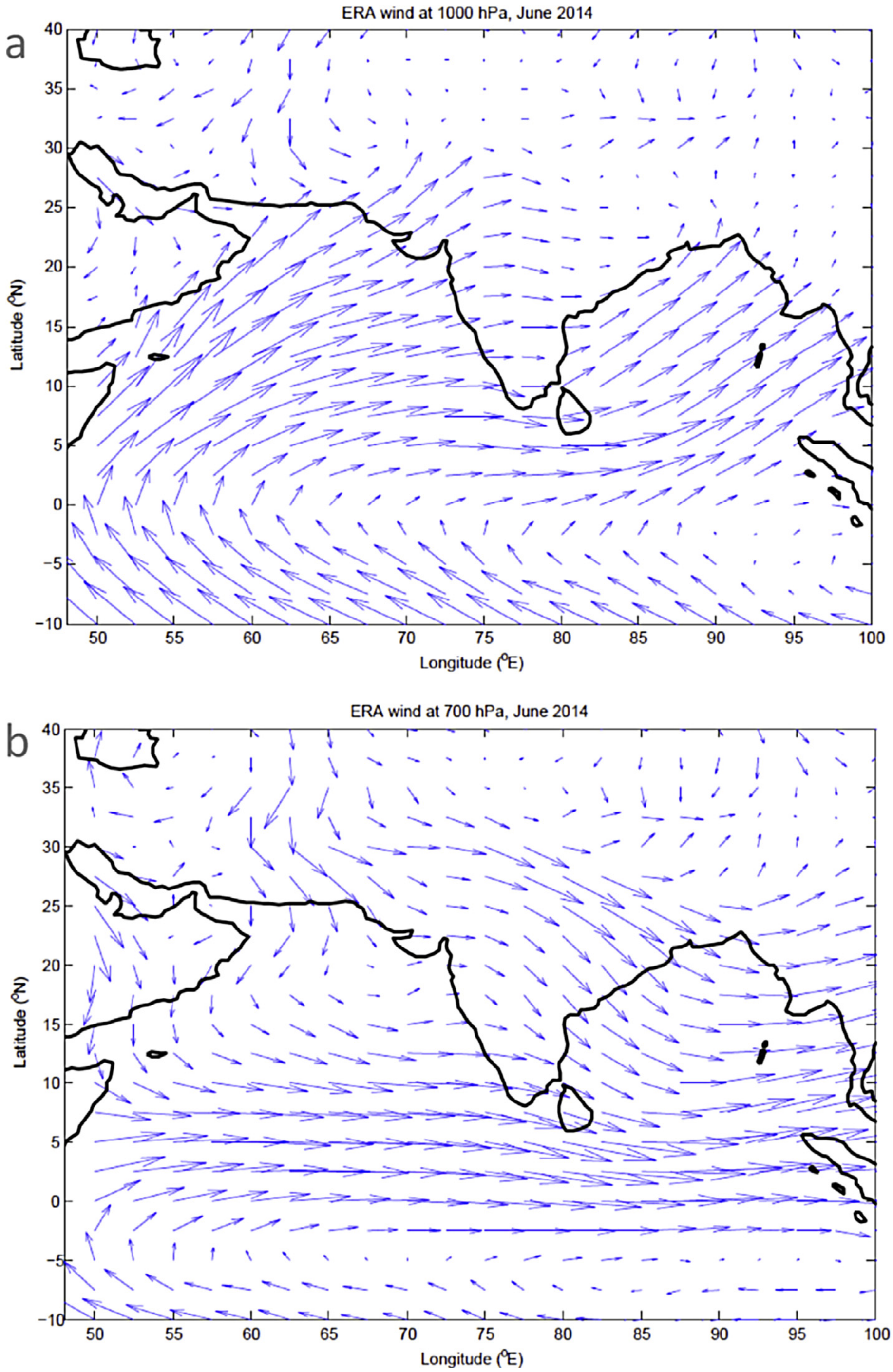

Fig. 2. Synoptic wind patterns over India and its surrounding oceans during June, 2014 (a) at 1000 hpa, (b) at 700 hpa.

(SK-313) and ORV Sagar Nidhi (SN-85) were found to average 4.6 ± 1.2 m s^{-1}, 233.9 ± 19.0 °N and 9.3 ± 3.3 m s^{-1}, 224.4 ± 64.6 °N, respectively. The diurnal average air temperature and RH (%) were seen to range between ~ 25 and 31 °C and ~75–78% respectively. Diurnal mean atmospheric pressure at the surface was noted to range between 1001.3 mb and 1005.7 mb along the cruise track of SK-313; while for SN-85, it was seen to increase from 1003.8 mb to 1010.7 mb and then dip again towards the end of the cruise. Overcast atmospheric conditions were prevalent, with light drizzle and occasionally torrential rain encountered during the course of SN-85.

3. Methods

3.1. Air mass back trajectory and cluster analysis

National Oceanic and Atmospheric Administration's (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT; Draxler and Rolph, 2003) model (http://www.arl.noaa.gov/ready/hysplit4.html) was employed for the computation of 120 h back trajectories culminating at 0700 UTC at a height of 500 m above ground level above the receptor sites and the cruise tracks. The back trajectory computation was carried out for the entirety of the

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Table 2
Meteorological conditions at the sampling sites during the period of study.

Site	Temperature (°C)		Pressure (mb)		Relative humidity (%)		Rainfall (mm)		Wind speed (m s^{-1})		Wind direction (°N)	
	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD
Patiala	23.1–44.1	38.1 ± 4.9	964.8–976.3	970.1 ± 2.7	5.9–40.4	17.7 ± 10.9	0.0–20.0	1.7 ± 4.8	0.0–0.9	0.1 ± 0.3	107.0–108.0	107.8 ± 0.4
Delhi	28.9–37.1	33.1 ± 2.3	964.8–1002.2	990.3 ± 8.8	33.7–64.3	45.3 ± 7.7	0.0–0.3	0.0 ± 0.1	0.9–3.0	1.9 ± 0.6	333.0–355.3	352.3 ± 5.9
Lucknow	29.0–37.0	33.3 ± 2.1	993.0–1003.0	996.6 ± 2.7	32.0–77.0	50.1 ± 11.9	0.0–5.1	0.5 ± 1.3	0.8–4.4	2.3 ± 1.0	114.3–292.9	195.2 ± 61.6
Varanasi	22.3–46.0	33.6 ± 1.7	982.9–987.3	984.9 ± 1.4	31.0–71.0	49.7 ± 11.1	0.0–12.0	2.1 ± 3.8	0.3–5.0	2.6 ± 1.3	81.5–276.5	213.7 ± 54.6
Giridih	27.3–36.8	32.2 ± 2.7	960.3–968.0	962.8 ± 2.4	38.1–95.4	69.8 ± 15.5	0.0–13.4	2.5 ± 4.6	0.5–1.6	1.1 ± 0.3	100.4–276.5	175.7 ± 54.3
Kolkata	27.5–32.6	30.1 ± 2.1	995.4–999.4	997.6 ± 1.4	76.4–100.0	89.7 ± 8.8	0.0–28.8	7.7 ± 10.3	0.8–2.3	1.5 ± 0.7	123.8–194.2	158.9 ± 25.2
Darjeeling	21.4–24.1	22.7 ± 0.9	880.9–886.0	883.6 ± 1.7	90.2–98.8	96.5 ± 3.0	0.0–33.2	5.4 ± 10.8	0.3–0.9	0.7 ± 0.2	131.3–204.2	168.4 ± 24.9
Bhubaneswar	24.9–31.5	28.7 ± 2.0	991.0–996.8	993.7 ± 1.5	54.3–99.4	86.7 ± 16.8	0.0–27.4	6.1 ± 9.0	2.0–5.2	3.3 ± 0.8	92.9–286.7	218.5 ± 66.3
Nagpur	28.8–41.9	37.2 ± 3.9	962.3–971.7	966.8 ± 2.7	16.2–82.1	34.6 ± 19.4	0.0–0.1	0.0 ± 0.0	0.8–3.9	2.6 ± 1.0	162.1–300.0	263.3 ± 38.8
SK-313	27.9–31.1	30.1 ± 0.8	1001.3–1005.7	1002.9 ± 1.3	74.1–86.3	77.8 ± 3.2	0.4–110.5	29.3 ± 36.8	2.6–6.3	4.6 ± 1.2	208.5–275.5	233.9 ± 19.0
SN-85	25.6–30.1	28.0 ± 1.2	1003.8–1010.7	1007.2 ± 2.0	67.9–86.0	74.8 ± 4.5	7.0–60.0	43.1 ± 16.8	3.2–14.5	9.3 ± 3.3	85.9–334.7	224.4 ± 64.6

SD: standard deviation; n.a: not available.

campaign period to get a more reliable depiction of the synoptic airflow pattern from the potential source regions to the receptor sites and is represented in Fig. 3.

Cluster analysis is a multivariate statistical technique which splits the trajectory data set into a number of clusters with maximum homogeneity within themselves and maximum heterogeneity between them (Brankov et al., 1998). It was used to classify trajectories with similar pathways, demarcating the mean transport pathways of the trajectories from the several existent trajectory pathways to the receptor land sites during the study period. The results of the cluster analyses for the $PM_{2.5}$ and PM_{10} sampling periods are represented in Fig. S1a and b respectively.

It must be noted that one cannot rely solely on cluster analysis for deciphering the sources of pollutants in a geographical region since it merely provides the directions in which possible sources may lie, but fails at pinpointing the exact locations of the sources.

3.2. Potential source contribution function (PSCF) analysis

The Potential Source Contribution Function (PSCF) was used in the present study to identify the potential source locations contributing to the $PM_{2.5}$ and PM_{10} mass over the land stations. It is a hybrid receptor model used to link the residence times in upwind areas of sampling stations with high concentrations (over an arbitrarily set criterion) of a species through a conditional probability field (Ashbaugh et al., 1985; Malm et al., 1986). The possible source region is divided into a gridded *i* by *j* array and PSCF value of the *ij*th cell is defined as:

$$\mathrm{PSCF}_{ij} = \frac{m_{ij}}{n_{ij}}$$

where PSCF_{ij} is the probability that air mass originates in the *ij*th cell on days with high species concentrations, m_{ij} is the number of trajectories arriving at the receptor site with species concentration greater then a predetermined criterion value (75th percentile of $PM_{2.5}/PM_{10}$ concentrations in our study) and n_{ij} is the total number of trajectories originating in the *ij*th grid during the study period. The PSCF value for grid cells with small number of trajectory end points (n_{ij}) compared to the average number of trajectory end points per cell ($\tilde{n}_{ij}$), will have very little confidence associated with them. In order to remove this large uncertainty, an arbitrary weight function ($W(n_{ij})$) is multiplied to the PSCF values to down-weigh the values for those cells with total number of end points less than about three times the average number of end points per grid cell (Han et al., 2005). The arbitrary weight function ($W(n_{ij})$) is defined as:

1 when $n_{ij} \geq 3\tilde{n}_{ij}$;
0.7 when $1.5\tilde{n}_{ij} \geq n_{ij} \geq 3\tilde{n}_{ij}$;
0.42 when $0.75\tilde{n}_{ij} \geq n_{ij} \geq 1.5\tilde{n}_{ij}$; and
0.17 when $n_{ij} < 0.75\tilde{n}_{ij}$

The entire geographical domain traversed by the air mass back trajectories reaching the land stations (15° S to 45° N latitude and 30° E to 100° E longitude) were divided into a geospatial grid having an array of 8400 grid cells of 0.5° × 0.5° latitude and longitude.

Fig. S2a and b depicts the PSCF results, informing us about the potential source regions adding to the $PM_{2.5}$ and PM_{10} mass respectively at the receptor sites. The results of the PSCF analysis are represented in the form of maps of the investigated domain upon which the PSCF values (ranging from 0 to 1) are displayed in the form of a color scale. Grids with dark red hues represent potential source areas in that region while those with light red to

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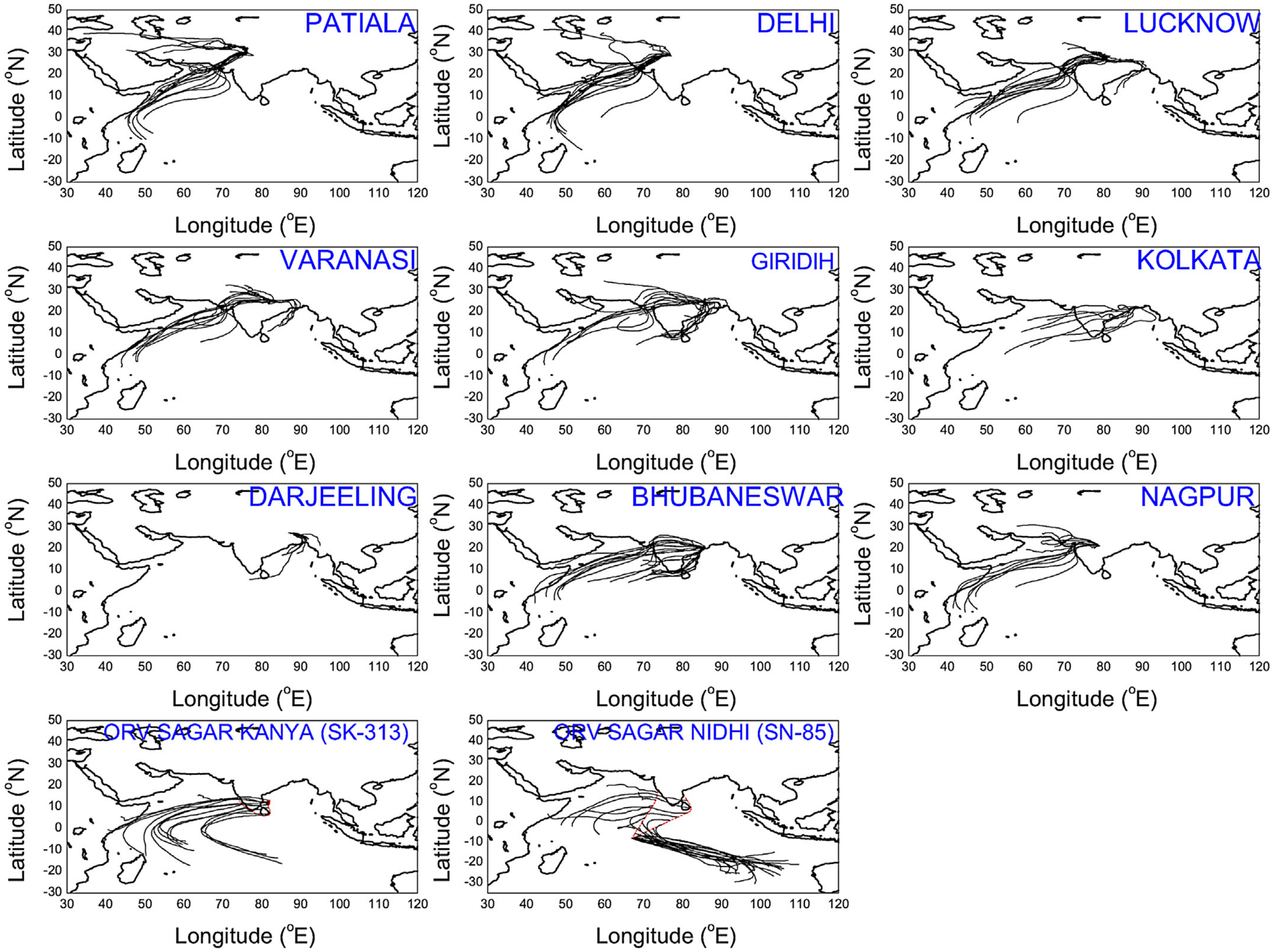

Fig. 3. Air mass HYSPLIT back trajectories plotted for the land stations and along the cruise tracks for the duration of the sampling period.

yellowish hues probably weren't significant sources for PM to the land receptor stations. However a cell with low PSCF value does not necessarily imply low emissions from that cell since the emissions may not be undergoing transport to the receptor site (Polissar et al., 2001).

3.3. *Concentration weighted trajectory (CWT) analysis*

The PSCF model output has difficulty in delineating strong sources from weak ones. In order to determine the relative significance of potential sources to the measured species concentrations at the receptor sites, we have used the Concentration Weighted Trajectory (CWT) model (Hsu et al., 2003). It is a method of assigning the species concentration values at the receptor site to the relevant back trajectories. The possible source region is divided into a gridded *i* by *j* array and CWT is defined for the *ij*th grid cell as:

$$C_{ij} = \frac{1}{\sum_{l=1}^{M} \tau_{ijl}} \sum_{l=1}^{M} C_l \tau_{ijl}$$

where C_{ij} is the average weighted concentration in the *ij*th grid cell, C_l is the measured species concentration at the receptor site on arrival of trajectory *l*, *M* is the total number of trajectories, and τ_{ijl} is the total number of trajectory end points in the *ij*th grid cell associated with the C_l sample. CWT method also employs an arbitrary weight function, similar to PSCF, in order to eliminate the grid cells containing few trajectory end points.

Fig. 4a and b depicts the distributions of weighted trajectory concentrations which inform us about the relative contributions of the potential source regions affecting the $PM_{2.5}$ and PM_{10} concentrations at the receptor sites respectively. CWT values are represented in the form of a color scale on the map of the investigated domain with reddish or orangish grids signifying strong source regions while violet, blue and green grids indicate weak to moderate sources of particulates to the receptor sites. The CWT is a function of the PM concentrations at the receptor sites as well as the residence times of the trajectories in each grid cells.

4. Results and discussion

The average and range in mass concentrations of ambient $PM_{2.5}$ and PM_{10} observed at the sampling stations and along the cruise tracks are depicted in Table 3, along with the $PM_{2.5}/PM_{10}$ ratios computed to determine locations with a relative domination in the loading of fine particulates to the coarser fractions. Previously reported mass concentrations of $PM_{2.5}$ and PM_{10}, as well as the $PM_{2.5}/PM_{10}$ ratios reported during the summer/monsoon season in and around the study region are summarized in Table 4.

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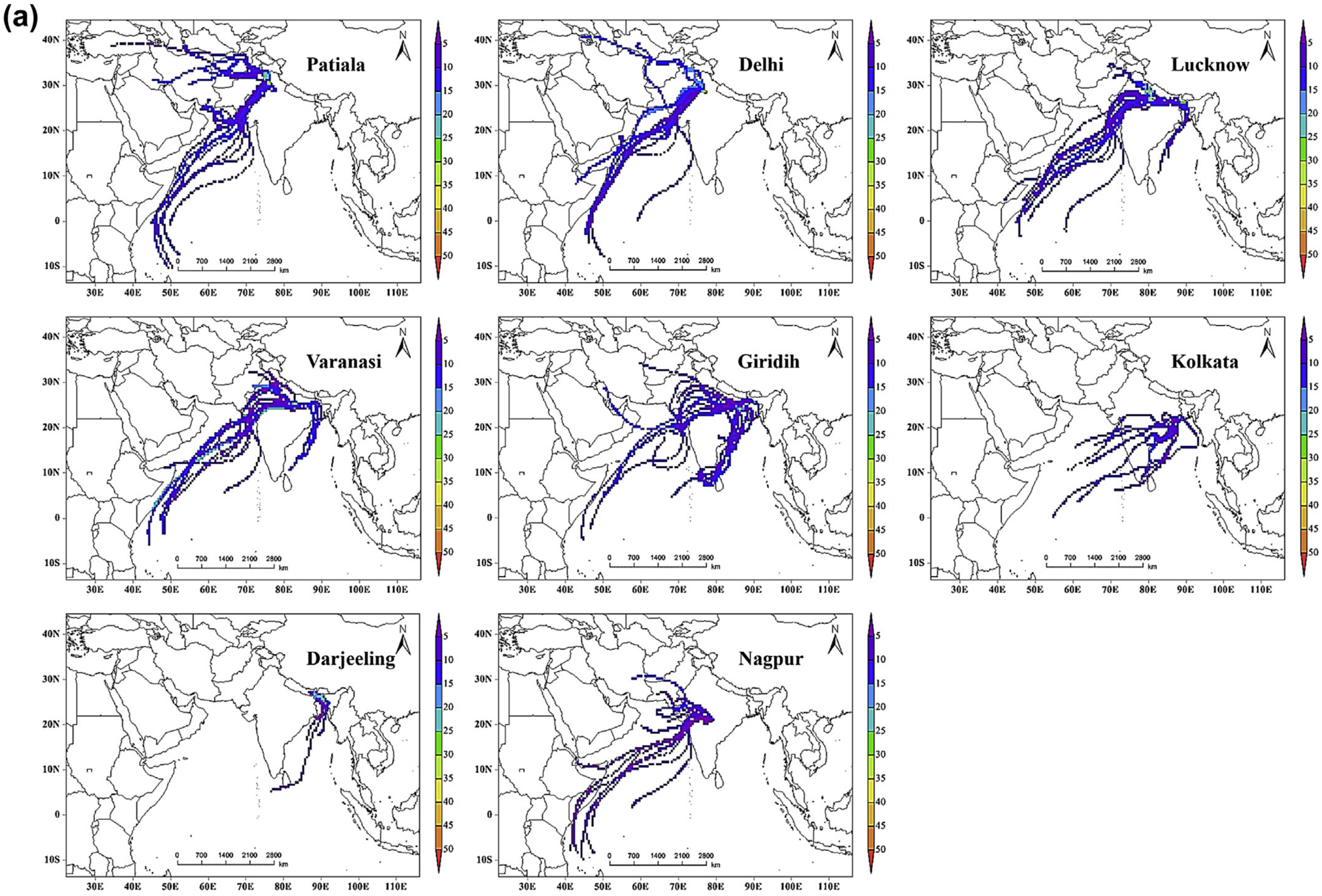

Fig. 4. Maps of concentration weighted trajectories (CWTs) of the land stations for (a) $PM_{2.5}$, (b) PM_{10}.

4.1. $PM_{2.5}$ mass concentration

Sampling sites in the upper IGP, viz. Delhi (61.8 ± 18.6 $\mu g\ m^{-3}$; average ± SD) and Patiala (55.4 ± 13.5 $\mu g\ m^{-3}$), were observed to have the highest daily average $PM_{2.5}$ concentrations, followed closely by those in the middle IGP, viz. Varanasi (52.5 ± 28.6 $\mu g\ m^{-3}$) and Lucknow (51.5 ± 17.7 $\mu g\ m^{-3}$). Kolkata (47.6 ± 9.3 $\mu g\ m^{-3}$) was noted to have an even lower concentration of fine mode $PM_{2.5}$ aerosols probably due to the onset of monsoon over Kolkata leading to particulate washout through wet deposition. Average $PM_{2.5}$ levels reported by Mandal et al. (2014) (142.50 ± 23.57 $\mu g\ m^{-3}$) for an industrial area of Delhi during June, 2011 are more than double the $PM_{2.5}$ levels observed during this campaign. $PM_{2.5}$ concentrations over Lucknow reported by Pandey et al. (2012) during summer (45.65 ± 1.4 $\mu g\ m^{-3}$) and monsoon (39.83 ± 4.6 $\mu g\ m^{-3}$) of 2007–09, were only slightly lower than the levels observed during this campaign. It is to be noted that all the aforementioned sites are major metropolitan cities spread across the IGP. Average $PM_{2.5}$ mass concentration over Giridih (48.6 ± 15.7 $\mu g\ m^{-3}$) was substantially lower than the values reported for the summer season over central Indian sites neighboring it, such as 128.3 ± 13.8 $\mu g\ m^{-3}$ (Raipur; Deshmukh et al., 2013), 83.5 $\mu g\ m^{-3}$ (Durg; Deshmukh et al., 2011) and 150.9 ± 78.6 $\mu g\ m^{-3}$ (Raipur; Deshmukh et al., 2010). Darjeeling (31.3 ± 20.0 $\mu g\ m^{-3}$), a hill station in the Himalayas; and Nagpur (35.2 ± 18.4 $\mu g\ m^{-3}$), a site in the Deccan Plateau, were noted to have the lowest average $PM_{2.5}$ mass concentrations amongst all the land stations. Chatterjee et al. (2010) had reported a much lower ambient $PM_{2.5}$ mass concentration (~18.0 $\mu g\ m^{-3}$) over Darjeeling during June, 2008. The daily mean $PM_{2.5}$ mass concentration at all the sampling sites in India with the exception of Delhi, were observed to fall within the 24 h average $PM_{2.5}$ value prescribed by the National Ambient Air Quality Annual Standards (60 $\mu g\ m^{-3}$) (NAAQS: http://www.scorecard.org/env-releases/def/cap_naaqs.html; set up by Central Pollution Control Board (CPCB), India).

The average $PM_{2.5}$ mass concentration observed in the MABL of coastal AS and SBoB by ORV Sagar Kanya (SK-313) (49.1 ± 28.7 $\mu g\ m^{-3}$; range: 20.6–100.8 $\mu g\ m^{-3}$) were in the vicinity of those observed over Giridih, Kolkata, Lucknow and Varanasi. In contrast, average $PM_{2.5}$ levels observed in the MABL of open oceanic regions of AS, SWTIO and SBoB by ORV Sagar Nidhi (SN-85) were the lowest (18.1 ± 10.1 $\mu g\ m^{-3}$, range: 5.3–50.9 $\mu g\ m^{-3}$) observed in this study.

4.2. PM_{10} mass concentration

The highest daily average PM_{10} mass concentrations were observed at sites in the middle IGP, viz. Lucknow (182.2 ± 58.0 $\mu g\ m^{-3}$) and Varanasi (139.6 ± 68.0 $\mu g\ m^{-3}$); followed closely by Delhi (127.4 ± 62.2 $\mu g\ m^{-3}$) located in the upper IGP. PM_{10} levels over Lucknow were found to be in the range of the values reported for summer (187.2 ± 17.1 $\mu g\ m^{-3}$) and monsoon (155.72 ± 22.7 $\mu g\ m^{-3}$) 2007–2009 (Pandey et al., 2012); and average PM_{10} levels over Varanasi were similar to the levels reported by Murari et al. (2014) for June, 2013 (150.2 $\mu g\ m^{-3}$). Similar, concentrations of ambient PM_{10} mass over Delhi were reported

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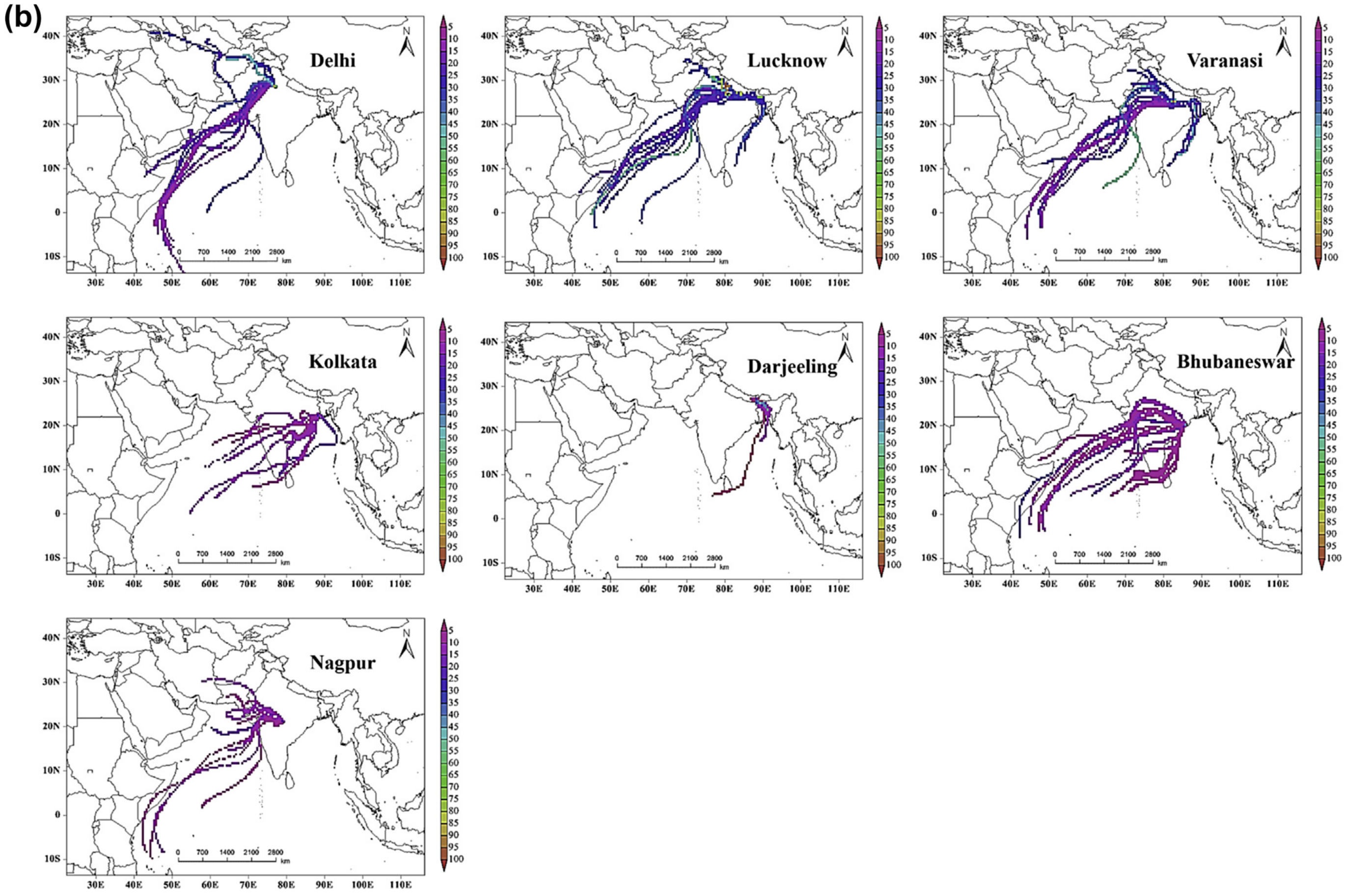

Fig. 4. (*continued*).

Table 3
Average and range of mass concentrations of $PM_{2.5}$ and PM_{10} and $PM_{2.5}/PM_{10}$ ratios at the sampling sites.

Site	$PM_{2.5}$ ($\mu g\ m^{-3}$)		PM_{10} ($\mu g\ m^{-3}$)		$PM_{2.5}/PM_{10}$
	Range	Mean ± SD	Range	Mean ± SD	
Patiala	11.6–74.9	55.4 ± 13.5	n.a	n.a	n.a
New Delhi	29.2–94.0	61.8 ± 18.6	45.3–293.3	127.4 ± 62.2	0.48
Lucknow	27.4–89.5	51.5 ± 17.7	93.5–360.6	182.2 ± 58.0	0.28
Varanasi	14.4–126.4	52.5 ± 28.6	64.2–301.9	139.6 ± 68.0	0.40
Giridih	19.6–78.7	48.6 ± 15.7	n.a	n.a	n.a
Kolkata	26.2–59.3	47.6 ± 9.3	48.8–104.2	66.7 ± 17.0	0.71
Darjeeling	9.2–62.2	31.3 ± 20.0	12.7–89.7	51.2 ± 29.2	0.61
Bhubaneswar	n.a	n.a	34.8–124.4	57.8 ± 22.4	n.a
Nagpur	10.9–78.5	35.2 ± 18.4	23.4–94.2	53.9 ± 23.7	0.65
SK-313	20.6–100.8	49.1 ± 28.7	n.a	n.a	n.a
SN-85	5.3–50.9	18.1 ± 10.1	21.0–75.8	40.7 ± 15.5	0.45

n.a: not available.

during monsoon 2010–11 (140.1 ± 43.9 $\mu g\ m^{-3}$) (Sharma et al., 2014) and for monsoon 2013–14 (143.86 ± 36.27 $\mu g\ m^{-3}$) (Sharma et al., 2015b). Kolkata, a representative sampling site in the lower IGP, recorded the lowest PM_{10} concentration (66.7 ± 17.0 $\mu g\ m^{-3}$) across the IGP. Stations located outside the confines of the IGP such as Bhubaneswar (57.8 ± 22.4 $\mu g\ m^{-3}$), Nagpur (53.9 ± 23.7 $\mu g\ m^{-3}$) and Darjeeling (51.2 ± 29.2 $\mu g\ m^{-3}$) had the lowest PM_{10} concentrations among the land stations, falling well within the National Ambient Air Quality Annual Standard (100 $\mu g\ m^{-3}$). However, PM_{10} levels over Darjeeling during this study are noted to be quite higher than that reported for June, 2008 (~31.0 $\mu g\ m^{-3}$) (Chatterjee et al., 2010), while those over Bhubaneswar are quite lower than the values reported for summer, 2010 (64.0–152.0 $\mu g\ m^{-3}$) (Bhol et al., 2013).

The daily average PM_{10} mass concentration recorded onboard ORV Sagar Nidhi (SN-85) from the MABL in open oceanic regions of AS, SWTIO and SBoB; were the lowest (40.7 ± 15.5 $\mu g\ m^{-3}$, range: 21.0–75.8 $\mu g\ m^{-3}$) throughout the campaign.

4.3. $PM_{2.5}/PM_{10}$ ratio

Kolkata (0.71) recorded the highest $PM_{2.5}/PM_{10}$ ratio followed by Nagpur (0.65) and Darjeeling (0.61), while Lucknow (0.28) recorded the lowest (Table 3). Stations located in the lower IGP

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Table 4
Previously reported $PM_{2.5}$, PM_{10} mass concentrations, and $PM_{2.5}/PM_{10}$ ratios in the Indian subcontinent during the summer/monsoon season.

Site	Location type	Sampling period	$PM_{2.5}$ (μg m^{-3})	PM_{10} (μg m^{-3})	$PM_{2.5}/PM_{10}$	Reference
Delhi	Urban	Monsoon, 2013–14		143.86 ± 36.27		Sharma et al. (2015b)
Delhi	Urban	June, 2011	142.50 ± 23.57			Mandal et al. (2014)
Delhi	Urban	Summer, 2010–11		192.7 ± 28.4		Sharma et al. (2014)
Delhi	Urban	Monsoon, 2010–11		140.1 ± 43.9		Sharma et al. (2014)
Lucknow	Urban	Summer, 2007–09	45.65 ± 1.4	187.2 ± 17.1	0.24	Pandey et al. (2012)
Lucknow	Urban	Monsoon, 2007–09	39.83 ± 4.6	155.72 ± 22.7	0.25	Pandey et al. (2012)
Varanasi	Urban	June, 2013		150.2		Murari et al. (2014)
Raipur	Urban/Industrial	Summer, 2009	128.3 ± 13.8	292.0 ± 47.1		Deshmukh et al. (2013)
Durg	Urban/Industrial	Summer, 2009–10	83.5			Deshmukh et al. (2011)
Raipur	Urban/Industrial	Summer, 2010	150.9 ± 78.6			Deshmukh et al. (2010)
Darjeeling	Remote Forested	Monsoon, 2005	~18.0	~31.0		Chatterjee et al. (2010)
Bhubaneswar	Urban	Summer, 2010	(16.0–60.0)[a]	(64.0–152.0)[a]		Bhol et al. (2013)

[a] Range.

recorded high $PM_{2.5}/PM_{10}$ ratios, showing a dominance of fine mode aerosols over coarse mode aerosols. It is also noted that stations in the upper and middle IGP were subjected to both the highest coarse mode and fine mode aerosol concentrations. This could be due to the mixing of the locally produced carbonaceous soot with the mineral dust aerosols advected into the region with the pre-monsoon westerlies, undergoing long range transport from desert regions in north-west India as well as from Saharan desert passing through South-west Asian arid regions; in accordance with the Elevated Heat Pump (EHP) hypothesis (Lau and Kim, 2006). The influx of coarse mode sea salt aerosols into the subcontinent with the onset of the summer monsoon could be contributing to the elevated PM_{10} mass concentrations (Table 3). It is noteworthy that SN-85 sampled only a slightly higher proportion of PM_{10} compared to $PM_{2.5}$ aerosols ($PM_{2.5}/PM_{10} = 0.45$).

4.4. Air mass back trajectory and cluster analysis

Fig. S1a and b depicts that a majority of the back trajectory clusters arriving at Patiala, Delhi, Lucknow and Varanasi originated in the AS or TIO traveling across the AS and over North-west India, implying possible transport of coarse mode marine and dust aerosols to the sampling sites. A couple of trajectory clusters arriving at Patiala and Delhi were noted to originate in South-west Asia and travel solely over continental land mass, indicating likely advection of desert dust aerosols to the study areas. Lucknow and Varanasi had trajectory clusters originating in the Northern India, Eastern India and over the BoB, possibly resulting in the inflow of a mixture of coarse marine and fine continental pollutant aerosols to the sites. The results were in line with the observations of Kumar et al. (2015) for Varanasi, where the fine particulate fraction was found to originate from north-eastern dry regions while the coarser particulate fraction was seen to accumulate there due to strong intercontinental westerlies. The airflow patterns observed for Kolkata, Giridih and Bhubaneswar are different from the rest of the aforementioned sites in the IGP. Though, a bulk of the clusters reaching these sites were south-westerly, originating in AS, they had to traverse across the peninsular region in central and southern India in order to reach Kolkata. The marine influence is slightly higher for Bhubaneswar, since its trajectory clusters were seen to traverse a greater distance over oceanic regions. Cluster analysis for Darjeeling revealed that most of the air masses reaching it originated in the BoB and had to travel only a small distance over lower IGP. Similarly, one can expect a significant marine influence in the PM collected over Nagpur, since a majority of the clusters reaching Nagpur were from the AS, depicting possible induction of a significant proportion of coarse mode marine aerosols into the receptor site. As expected, predominantly oceanic air masses originating in the Southern TIO and were encountered in coastal AS and BoB along the cruise path of ORV Sagar Kanya (SK-313) and in open oceanic regions by ORV Sagar Nidhi (SN-85) (Fig. 3).

4.5. Potential source contribution function (PSCF) analysis

AS and BoB serve as major potential source regions for accentuating $PM_{2.5}$ and PM_{10} levels at all the land receptor sites (Fig. S2a and b). Also, arid regions in South-west Asia and Northern India are major potential contributors to the measured $PM_{2.5}$ mass concentrations at Patiala, Delhi and Nagpur. Similarly, fine particulate concentrations at Varanasi and Lucknow were possibly enhanced by sources in Northern and North western India. Regions in AS, peninsular India and BoB were major probable sources for increasing $PM_{2.5}$ levels at Kolkata, while the $PM_{2.5}$ levels in Darjeeling were influenced mainly by potential sources in the BoB. Possible source areas contributing to the PM_{10} mass concentrations measured over Lucknow, Varanasi, Nagpur and Bhubaneswar were mainly in the AS, whereas they were observed to be mostly in the BoB for Kolkata and Darjeeling. Regions in Pakistan and in Northern and North western India were probable contributing sources to PM_{10} in Delhi, Lucknow and Varanasi respectively. The Deccan plateau is a potential PM source region for both Bhubaneswar and Nagpur, and the PM recorded at the former site could also have sources in the Horn of Africa.

4.6. Concentration weighted trajectory (CWT) analysis

A bulk of the air masses arriving at the land receptor sites were mainly from a south-westerly direction. Moderate contributions to both $PM_{2.5}$ and PM_{10} levels from source regions in the AS are observed at almost all sampling sites (Fig. 4a and b). Lucknow, Varanasi, Darjeeling, Kolkata and Bhubaneswar are also observed to have moderate $PM_{2.5}$ and PM_{10} contributions from sources in the BoB. Patiala, Delhi, Lucknow, Varanasi and Giridih have weak to moderate contributions of both $PM_{2.5}$ and PM_{10} from arid source regions in South-west Asia, Northern Pakistan and North-west India, and from sources within the IGP region. Also, source regions in peninsular India contribute fairly to enhancing the $PM_{2.5}$ and PM_{10} concentrations at Bhubaneswar, Kolkata and Nagpur. Nagpur is the only land station to receive contributions, albeit small, to both its $PM_{2.5}$ and PM_{10} levels from the Horn of Africa in addition to that received from sources in Pakistan, West India and AS.

4.7. SW monsoon, 2014 vs NE monsoon, 2014

During the course of the present campaign, it was observed that all the sampling sites in India recorded very low concentrations of

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both fine and coarse particulates compared to that observed for the previous campaign conducted during the winter monsoon 2013–14 (Sen et al., 2014). This can be attributed to the greater scavenging of these particulates on account of higher precipitation during summer monsoon compared to the winter monsoon period. The solar heating of the land during summer results in increased convective activity leading to increased planetary boundary layer (PBL) height compared to the winter season, and this rise in ventilation coefficient leads to a faster dispersal of aerosols. Thus summer across the Indian mainland is characterized by a high PBL height and relatively high low-level winds favoring elevated dispersal of aerosols, which could have manifested as the sharp decrease in the ambient particulate levels at the sites during this campaign as against the winter campaign.

During the winter, 2013–14 campaign, elevated particulate levels in the IGP were attributed to the increase in usage of biomass fuels and also due to the fact that winter across IGP is characterized by low and stable PBL and relatively low low-level winds, resulting in increased stagnation of particulates. Furthermore, the low fine mode aerosol concentrations over the land stations can also be due to the fact that a majority of the air masses arriving at these sites during the campaign period were observed to originate in Southwest Asia or AS and travel mostly across AS, leading to an influx of more coarse mode mineral dust and sea salt aerosols as compared to the finer fractions. On the other hand, the results of the back trajectory analysis during the NE monsoon period revealed that a majority of the air masses were originating and flowing through the IGP resulting in an influx of mostly fine mode continental pollutants to the sampling sites. A major portion of the cruise track of ORV Sagar Nidhi (SN-85) covered remote marine regions of AS, SWTIO and BoB; and back trajectory analysis shows that the air-masses encountered were predominantly oceanic in nature, originating in the Southern Ocean or in the AS, possibly accounting for the exceedingly low particulate levels encountered along cruise track. The elevated $PM_{2.5}$ levels in the MABL of the cruise track of ORV Sagar Kanya (SK-313) compared to that of ORV Sagar Nidhi (SN-85) can be explained by the vicinity of the cruise track of to the Indian coast. Back trajectory analysis indicates that almost all of the air masses reaching the track in SBoB had to traverse through peninsular India, which would result in an influx of continental fine mode pollutants in addition to the coarse mode sea salt aerosols.

5. Conclusions

This study involved collection of ambient $PM_{2.5}$ and PM_{10} aerosols across multiple locations in India, as well as from coastal and open oceanic regions; and was carried out with the objective of accurately estimating and comparing the fine ($PM_{2.5}$) aerosol and coarse (PM_{10}) aerosol levels observed over those regions during the onset of summer monsoon, 2014. It was conducted as a follow up to the campaign study conducted by Sen et al. (2014) over different regions of India and BoB during winter monsoon 2013–14; in order to visualize the inter-seasonal variation in $PM_{2.5}$ and PM_{10} mass concentrations across the study region as well as to catch the effect of pre-monsoonal dust deposition from Sahara desert as well as Thar desert. Salient features of the study are summarized below:

- Sites located in the upper and middle IGP were noted to have higher daily average $PM_{2.5}$ and PM_{10} levels compared to the other land stations. The average $PM_{2.5}$ and PM_{10} mass concentrations during this campaign at all the land stations were far lower than the measurements at the same sites during the winter monsoon season possibly due to the higher PBL height and faster low level winds observed during the summer monsoons.
- The proximity of the cruise track of ORV Sagar Kanya (SK-313) to the Indian mainland could allow sampling of continental fine mode pollutants and resulting in $PM_{2.5}$ levels almost at par with most of the land stations. However, average $PM_{2.5}$ and PM_{10} levels measured onboard ORV Sagar Nidhi (SN-85) were significantly lower than that measured at any of the land stations since the cruise track mainly covered remote marine regions, far away from any source of terrestrial pollutants.
- Sites in upper and middle IGP, recorded a lower $PM_{2.5}/PM_{10}$ ratio compared to the other sites, indicating a lower proportion of fine particulate loading compared to coarser fractions at the former sites.
- A majority of the back-trajectory clusters arriving at the land stations were originating in the marine regions of TIO or AS, and in arid landscapes of South-west Asia or North-west India, possibly resulting in the increased advection of coarse marine aerosols and coarse dust aerosols to the sites. As expected, predominantly oceanic air masses were encountered onboard ORV Sagar Kanya (SK-313) and ORV Sagar Nidhi (SN-85).
- PSCF and CWT analysis indicate that a majority of the potential marine source regions that could have led to a moderate enhancement in $PM_{2.5}$ and PM_{10} levels at the land stations are located across AS or BoB. Probable sources with weak to moderate contributions to $PM_{2.5}$ levels at a majority of the land sites were also located in arid regions of South-west Asia and North-west India and in the polluted IGP region. Similarly, potential source regions with moderate contributions to increment in PM_{10} levels at some of the land sites are noted to be spread across Pakistan, North and North-west India and in peninsular India.

A complete chemical characterization of the organic and elemental carbon species, trace metal species and WSIS in both $PM_{2.5}$ and PM_{10} over the different regions is ongoing and will be reported in the near future.

Conflict of interest

There is no conflict of interest.

Acknowledgments

The authors are grateful to the Director, CSIR-NPL, India and Head RASD, CSIR-NPL, India for their encouragement and support during the course of the work. Avirup Sen is indebted to CSIR, New Delhi for providing SPMF-SRF fellowship to carry out this research. Two of the authors, S. Goel and Padma S. Rao, would like to thank Director, CSIR-NEERI for his continuous support and guidance. We acknowledge the Indian Meteorological Department for allowing the usage of the meteorological data at respective sites. Authors duly acknowledge the financial support provided by University Grants Commission, New Delhi for sampling at BHU, Varanasi. The authors are grateful to Director, IITR, Lucknow for his encouragement and support and permission to participate in this study. Authors are thankful to ISRO - GBP for their financial assistance in the form of Research project under ARFI Network stations for sampling at Kadapa. Authors acknowledge Prof Sibaji Raha for all support for carrying out the observation at Kolkata and Darjeeling. Authors acknowledge the technical support of Mr Sabyasachi Majee, Darjeeling and Mr Bhaskar Ray and Miss Debolina Seal, Kolkata. The authors are thankful to Director, Indian Statistical Institute and Head, Human Resources Development Group, CSIR, Extramural Research division, New Delhi for their encouragement and support for this study at Giridih. Authors are thankful to The National Centre for Antarctic and Ocean Research (NCAOR) for providing the

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opportunity in ORV Sagar Nidhi (SN-85) for participating in South West Tropical Indian Ocean (SWTIO) and ORV Sagar Kanya (SK-313) for carrying out scientific work. Authors are grateful to National Oceanic and Atmospheric Administration's (NOAA) for allowing the trajectory calculation using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model.

Appendix A. Supplementary material

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2016.01.001.

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