SHORT COMMUNICATION

Atmospheric Fine and Coarse Mode Aerosols at Different Environments of India and the Bay of Bengal During Winter-2014: Implications of a Coordinated Campaign

A. Sen¹, Y. N. Ahammed², B. C. Arya¹, T. Banerjee³, G. Reshma Begam², B. P. Baruah⁴,
A. Chatterjee⁵, A. K. Choudhuri⁶, A. Dhir⁷, T. Das⁸, P. P. Dhyani⁹, N. C. Deb¹⁰, R. Gadi¹¹,
M. Gauns¹², S. K. Ghosh⁵, A. Gupta⁷, K. C. Sharma¹², A. H. Khan¹³, K. M. Kumar¹⁴,
M. Kumar³, A. Kumar¹, J. C. Kuniyal¹⁵, A. Lakhani¹⁴, R. K. Meena¹⁴, P. S. Mahapatra⁸,
S. W. A. Naqvi¹⁶, D. P. Singh¹², S. Pal¹⁰, S. Panda⁸, Rohtash¹, J. Saikia⁴, P. Saikia⁴,
A. Sharma¹, P. Sharma¹⁵, M. Saxena¹, D. M. Shenoy¹⁶, C. Viswanatha Vachaspati²,
S. K. Sharma¹ and T. K. Mandal¹*

¹Radio and Atmospheric Sciences Division, CSIR-National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110012, India

²Department of Physics, Yogi Vemana University, Kadapa 516003, India

³Institute of Environment and Sustainable Development, Banaras Hindu University, Banaras, UP, India

⁴CSIR-North East Institute of Science and Technology, Jorhat, Assam, India

⁵Centre for Astroparticle Physics and Space Science, Bose Institute, Kolkata and Darjeeling, West Bengal, India

⁶Indian Statistical Institute, Giridih, Jharkhand, India

⁷School of Energy and Environment, Thapar University, Patiala, Punjab, India

⁸CSIR-Institute of Minerals and Materials Technology, Bhubaneswar, Odisha, India

⁹G.B. Pant Institute of Himalayan Environment and Development, Kosi-Katarmal, Almora, Himachal Pradesh, India

¹⁰Indian Statistical Institute, B.T. Road, Kolkata, West Bengal, India

¹¹Indira Gandhi Delhi Technical University for Women, Kashmiri Gate, Delhi, India

¹²Central University of Rajasthan, Ajmer, Rajasthan, India

¹³CSIR-Indian Institute of Toxicology Research, Lucknow, Uttar Pradesh, India

¹⁴Dayabagh Educational Institute, Dayalbagh, Agra, Uttar Pradesh, India

¹⁵G.B. Pant Institute of Himalayan Environment and Development, Himachal Unit, Mohal, Kullu, Himachal Pradesh, India

¹⁶CSIR-National Institute of Oceanography, Dona Paula, Goa 403004, India

Received: 11 March 2014 / Accepted: 12 June 2014

© Metrology Society of India 2014

Abstract: In this paper, we present mass concentrations of particulate matter $[PM_{2.5}, PM_{10}]$ size fractions and total suspended particulates (TSP)] measured simultaneously over land stations (Kullu, Patiala, Delhi, Ajmer, Agra, Lucknow, Varanasi, Giridih, Kolkata, Darjeeling, Jorhat, Itanagar, Imphal, Bhubaneswar, and Kadapa), mostly distributed across the

^{*}Corresponding author, E-mail: tuhin_m@yahoo.com; tuhin@nplindia.org

Indo-Gangetic plain (IGP) of India as well as in the marine atmosphere over Bay of Bengal (BoB) in the period from 20 January to 3 February, 2014. The main objective of this study was to quantify the continental outflow of particulates (PM_{2.5}, PM₁₀ and TSP) from IGP and associated regions into the BoB along with low level north-east wind flow during winter monsoon period. The present study provides a glimpse of the aerosol loading over the IGP region. During this campaign, the highest average PM_{2.5} (187.8 ± 36.5 µg m⁻³, range 125.6–256.2 µg m⁻³), PM₁₀ (272.6 ± 102.9 µg m⁻³, range 147.6–520.1 µg m⁻³) and TSP (325.0 ± 71.5 µg m⁻³, range 220.4–536.6 µg m⁻³) mass concentrations were recorded at Varanasi, Kolkata and Lucknow over middle and lower IGP regions. The PM_{2.5} (average 41.3 ± 11.9 µg m⁻³; range 15.0–54.4 µg m⁻³), PM₁₀ (average 53.9 ± 18.9 µg m⁻³; range 30.1–82.1 µg m⁻³) and TSP (average 78.8 ± 29.7 µg m⁻³; range 49.1–184.5 µg m⁻³) loading over BoB were found to be comparable to land stations and suggests possible continental outflow. Over the continental region, the highest PM_{2.5}/PM₁₀ ratio was recorded at Delhi (0.87). The PM_{2.5}/PM₁₀ ratio over BoB (0.77) was found to be quite high and comparable to Varanasi (0.80) and Agra (0.79).

Keywords: PM_{2.5}; PM₁₀; TSP and Bay of Bengal

1. Introduction

Atmospheric aerosols comprise only a minuscule fraction of the Earth's atmosphere and have short atmospheric residence times; however they are a highly important constituent, being capable of significantly altering the radiation budget of the atmosphere regionally, either directly by scattering or absorbing the incoming solar radiation or indirectly by cloud formation [1, 2]. Long term exposure to fine particulates could lead to acute health effects owing to the fact that they are able to penetrate into the lungs and are more likely to increase chances of cardiopulmonary and carcinogenic diseases [3]. Varotsos [4] carried out airborne measurements of the vertical distribution of aerosol number density and aerosol size (aerosol mean diameter), erythemal ultraviolet irradiance (EUVI), ozone mixing ratio, relative humidity (RH) and temperature using research aircrafts and weather balloons, reporting aerosol mean diameter and ozone mixing ratio decrease with altitude thereby influencing a general increase in EUVI with altitude. Moreover, studies conducted in Greece have successfully documented the corrosive and soiling effects of aerosols on materials [5, 6].

Polluted air masses originating from the heavily populated south-Asian region could travel long distances maintaining their identity whilst being transported by the winds [7]. These air masses contain large concentrations of carbonaceous aerosols, emitted from bio-fuel and fossil fuel combustion [8]. Rapid urbanization, industrialization and a rampant increase in vehicular traffic across the megacities of India, particularly over the IGP region also contribute heavily to the ever increasing load of atmospheric particulates [9–12]. Pollutants from the IGP (being a source region) not only affect the local air quality but also undergo long range transport due to the high convective activity prevalent in the tropics [13]. The Bay of Bengal is landlocked from three sides and bordered by densely populated areas. It is considered to be one of the most suitable locations to study the influx of continental aerosols into the marine atmosphere and their interaction with natural marine aerosols, due to their long range transport during winter on account of the near surface flow being primarily north easterly [14].

Numerous studies have been conducted in the Indian region as well as the BoB over the course of the last few years, with several authors reporting the chemical composition of the ambient $PM_{2.5}$ and PM_{10} [15–22] and documenting the transport of natural (mineral dust) as well as anthropogenic aerosols from Indian subcontinent and south-Asian region [14, 21, 23–26]. However, these studies have been of an intermittent nature. This is probably the first time that ambient aerosols have been collected simultaneously at multiple locations across the Indian subcontinent and the BoB with the objective of examining the sources leading to the outflow of continental aerosols over the BoB during the winter monsoon period.

2. Experimental

2.1. Site Description

Sampling of ambient particulates falling in the PM2.5 and PM₁₀ size fractions as well as TSP was carried out simultaneously at 15 locations across India. A majority of these stations are spread across the IGP-India. Three of the sampling sites, viz. Jorhat, Imphal and Itanagar are in North-East India. Kullu, Ajmer and Kadapa serve as representative stations for North, North-West and South India respectively. Ajmer is a semi-arid site which is cut off from the Thar Desert since it is surrounded by the Aravalli Mountains. Kadapa is located in the rain shadow region and is a tropical semi-arid continental site in Andhra Pradesh. Bhubaneswar and Kolkata were chosen with the objective of serving as coastal stations bordering the BoB. Giridih is positioned on a plateau slightly to the south of the IGP and coal mining is a major industrial activity there. The atmospheric conditions at these sites also vary greatly;

Table 1 Description of sampling locations, period and instruments operated

Sites	Location	State	Coordinates		Instruments operated			Sampling	
			Latitude (N)	Longitude (E)	PM _{2.5}	PM ₁₀	TSP	Duration	
Site 1	Kullu	Himachal Pradesh	31°37′	77°20′	APM 550	APM 460 NL		20 January–5 February	
Site 2	Patiala	Punjab	30°21′	76°22′	IPM-FDS- 2.5 μm/10 μm		IPM-115- BL-NL	20 January–5 February	
Site 3	New Delhi	Delhi	28°38′	77°10′	APM 550	APM 460 NL		20 January–6 February	
Site 4	Ajmer	Rajasthan	26°34′	74°51′		APM 460 NL		20 January–5 February	
Site 5	Agra	Uttar Pradesh	27°13′	78°00′	APM 550	APM 460 NL	APM 430	20 January–5 February	
Site 6	Lucknow	Uttar Pradesh	26°52′	80°56′	APM 550	APM 460 NL	APM 430	20 January–8 February	
Site 7	Varanasi	Uttar Pradesh	25°15′	82°59′	IPM-FDS- 2.5 μm/10 μm	IPM-FDS- 2.5 μm/10 μm		20 January–5 February	
Site 8	Giridih	Jharkhand	24°10′	86°17′		PEM-HVS-8NL		20 January–5 February	
Site 9	Kolkata	West Bengal	22°34′	88°21′	APM 550	APM 460 NL		20 January–5 February	
Site 10	Darjeeling	West Bengal	27°01′	88°15′	APM 550	APM 460 NL		20 January–4 February	
Site 11	Jorhat	Assam	26°44′	94°09′	APM 550	APM 460 NL	APM 430	20 January–4 February	
Site 12	Itanagar	Arunachal Pradesh	27°07′	93°44′	APM 550	APM 460 NL	APM 430	20 January–4 February	
Site 13	Imphal	Manipur	24°49′	93°55′	APM 550	APM 460 NL	APM 430	20 January–4 February	
Site 14	Bhubaneswar	Odisha	20°17′	85°49′	PES-LFAS-10	APM 460 NL		20 January–6 February	
Site 15	Kadapa,	Andhra Pradesh	14°28′	78°55′	Anderson Sampler	Anderson Sampler	F&J HV 1E	20 January–4 February	
Site 16	Bay of Bengal				APM 550	APM 550	Tisch TE- 2000P	23 January–3 February	

from pristine Himalayan air at Kullu and Darjeeling to highly polluted conditions prevailing in metropolitan cities like Kolkata and Delhi. The co-ordinates of the sampling sites are available in Table 1.

Ambient aerosol samples ($PM_{2.5}$, PM_{10} and TSP) were collected from the marine atmospheric boundary layer (MABL) of BoB during the cruise onboard ocean research vessel Sagar Kanya (SK-308). The cruise set sail from Paradip, Odisha (20.26°N, 86.67°E) on 23rd January (DOY-023), 2014 and culminated at Chennai (13.08°N, 80.29°E) on 3rd February, 2014 (DOY: 034) with the track (Fig. 1) covering 13–20°N latitudes and 83–89°E longitudes.

2.2. Sample Collection and Analysis

The APM 550, APM 460 NL and APM 430 samplers used during the campaign are manufactured by M/s. Envirotech

Instruments Pvt. Ltd., New Delhi. The APM 550 is utilized for sampling aerosols falling in the $PM_{2.5}$ and PM_{10} size fractions (http://www.envirotechindia.com). The impactors are designed as per United States Environmental Protection Agency (USEPA) standards. Ambient air enters the sampler through an omni-directional air inlet. PM₁₀ aerosols in the air stream are separated by an impactor following which the PM_{2.5} aerosols can be separated if a WINS impactor is used. Finally, the particulate fraction remaining in the air stream is deposited on to a 47 mm QM-A quartz micro fibre filter. A constant flow rate of 1 m³ h⁻¹ (accuracy ± 2 % of F.S) is maintained irrespective of voltage fluctuations. The APM 460 NL sampler uses a blower with an inbuilt thermal cut-off eliminating the requirement of a voltage stabilizer (http://www.envirotechindia.com). PM₁₀ aerosols are deposited onto 20 \times 25 cm² QM-A filters. It has a flow rate ranging from 0.9 to 1.4 m³ h⁻¹ (accuracy ± 2 % of F.S). The



Fig. 1 Site-map and cruise track

APM 430 is a High Volume Sampler (HVS) utilized for collecting the TSP in the ambient air. The IPM-FDS-2.5 μ m/ 10 µm and IPM-115-BL-NL samplers are produced by Instrumex (http://www.instrumex.net) and designed as per USEPA guidelines. IPM-FDS-2.5 µm/10 µm is an ambient fine dust sampler whose mode of operation is similar to that of the APM 550 utilizing the same mechanisms for PM₁₀ and PM_{2.5} aerosol separation. The flow rate meter has a resolution of 0.01 LPM under actual operating conditions and the flow control maintains a flow of 1 m³ h⁻¹. The IPM-115-BL-NL is a Respirable Dust Sampler and used for the measurement of TSP in the ambient atmosphere. Particles with aerodynamic diameter below 10 microns ($D_p < 10 \ \mu m$) are collected onto the filter paper whereas the larger particles with 10 μ m < D_p < 100 μ m are separated by a cyclone separator and collected in a separate bottle. It is a HVS, drawing in air at a flow rate of $\sim 1.4 \text{ m}^3 \text{ h}^{-1}$. PEM-HVS-8NL is a HVS manufactured by M/s. Polltech Instruments Pvt. Ltd. (http://polltechinstruments.tradeindia.com), drawing in ambient air at a constant sampling rate of 1.13 m³ h⁻¹ (flow range: $0.6-1.8 \text{ m}^3 \text{ h}^{-1}$). The particulates in the air stream are accelerated through numerous circular impactor nozzles and those with $D_p > 10 \ \mu m$ impact on a greased collection plate while the aerosols with $D_p < 10 \ \mu m$ are deposited onto 20×24 cm² OMA filter. The TE 2000P is a HVS manufactured by Tisch Environmental, Inc. used for estimating the TSP concentration in the ambient air (http:// tisch-env.com). The airborne TSP is trapped onto a 110 mm diameter QM-A filter paper. The HV 1E is a HVS manufactured by F&J Specialty Products, Inc. featuring a single rotameter (http://www.fjspecialty.com) and working similar to the TE 2000P. The Andersen sampler has nine aluminum stages (D $_{\rm p} < 0.43~\mu{\rm m}, 0.43~\mu{\rm m} < D_{\rm p} < 0.65~\mu{\rm m}, 0.65~\mu{\rm m} <$ $D_p < 1.1 \ \mu m, \ \ 1.1 \ \mu m < D_p < 2.1 \ \mu m, \ \ 2.1 \ \mu m < D_p <$ 3!.3 μ m, 3.3 μ m < D_p < 4.7 μ m, 4.7 μ m < D_p < 5.8 μ m,

5.8 μ m < D_p < 9 μ m, 9 μ m < D_p < 10 μ m) to measure the size distribution and concentration of aerosols in ambient air (http://www.newstarenvironmental.com). It is operated at a flow rate of 28 LPM.

PM_{2.5} and PM₁₀ Low Volume Samplers (LVS) were operated for a period of 24 h each along with a High Volume Sampler (HVS) having a run time of 24 h to collect the ambient particulate matter at the respective sampling sites. Samples were collected on prebaked Pallflex QM-A micro fibre filters. The QM-A filters were prebaked in a muffle furnace for 6 h at 550 °C to remove any traces of impurities and were desiccated for a period of 48 h prior to sample collection. The instruments onboard oceanic research vessel (ORV) Sagar Kanya was installed on the upper deck at a height of 15 m from sea surface. While sampling, care was taken to avoid any contamination from the ship's exhaust. To understand and remove the contamination of the chimney, free-fall particulates were simultaneously collected on a separate filter paper along with the present observations. Description of sampling locations, period of sampling and details of the make and model of the instruments operated at each of the sites are available in Table 1.

The QMA filters were also desiccated for a period of 48 h prior to measurement of their final weights. PM mass was calculated by gravimetric analysis based on the difference between the final and initial weights of the QM-A filters measured using a microbalance (Make: M/s Sartorius, resolution $\pm 1 \mu g$) and its concentration was determined by dividing it by the volume of air sampled for the duration of the experiment.

2.3. Meteorological Conditions

The meteorological conditions existing at each of the stations during the period of sampling has been depicted in Table 2. The data has been assimilated from the automatic weather stations (AWS) operating at the sampling sites and has been supplemented by data acquired from India Meteorological Department. Kadapa recorded the highest average ambient temperature $(23.1 \pm 2.4 \text{ °C})$ among the land stations while Delhi had the highest RH $(82.2 \pm 9.5 \%)$. Sporadic rainfall was received at Patiala, Delhi, Ajmer and Lucknow while the other stations didn't receive any. The highest average wind speed was recorded at Lucknow $(7.4 \pm 1.9 \text{ km h}^{-1})$. Winds speeds encountered in the BoB were ranging between 0.6 and 21.0 km h^{-1} (mean 10.0 \pm 5.0 km h^{-1}) and the average wind direction was 76.0 \pm 83.8°. True wind is defined as a vector wind with a speed referenced to the fixed earth and a direction referenced to the true north. The corrected true wind speeds and directions have been computed taking into account the ship speed and direction according to the

approach described in detail by Smith et al. [27]. A typical root mean square (RMS) error for a wind speed observation is averaging around 2.5 m s⁻¹ (9 km h⁻¹) if the anemometer observation is not corrected for the height of the instrument above the sea surface, with the RMS wind speed errors smaller than average in the tropics [28]. A major cause of the error lies in the calculation of true wind speed and direction, with only 50 % of the reported wind speeds lying within 1 m s⁻¹ (3.6 km h⁻¹) of the correct value and about 30 % of them being more than 2.5 m s⁻¹ (9 km h⁻¹) inaccurate, while about 70 % of the winds were reported to blow within $\pm 10^{\circ}$ of the correct direction and 13 % of them blowing from outside $\pm 50^{\circ}$ [28]. The average temperature, pressure and RH were recorded as 24.4 ± 2.0 °C, $1.013.4 \pm 1.7$ mb and 69.5 ± 8.4 % respectively over BoB. Clear sky conditions prevailed and there was absence of any rainfall during the duration of this cruise.

2.4. Air Mass Back-Trajectory Analysis

Five day air mass back trajectories were computed using the NOAA (National Oceanic and Atmospheric Administration) Air Resource Laboratory (ARL) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model with the Global Data Assimilation System data (http://www.arl.noaa.gov) as an input. The 5 day back trajectories are calculated every day at 12:30 h (IST) at 500 m above the ground level (AGL) for the sampling duration and have been represented in Fig. 2. They have been plotted in order to picture the origin of the air masses and their transport pathways to the sampling sites. All the air mass trajectories for the sites located within the IGP are seen to originate as well as undergo transportation within the IGP, thereby signifying the dominance of anthropogenic emission transport to the sampling sites. Lucknow is noted to have just one trajectory during the entire study period originating in the Arabian Peninsula and traversing Northern Arabian Sea, while Darjeeling is seen to receive air masses originating over a large area with three of the trajectories crossing the Arabian Sea. Kadapa is a noted exception among the land stations since all the air masses received by the site had to traverse across northern BoB. In fact, nearly all the air masses reaching at most of the land stations excluding Kadapa and Darjeeling, were seen to originate and traverse across continental land masses. Though Kolkata is a coastal station bordering the BoB, all the air mass trajectories culminating at the two sites originated and underwent transport across the IGP. Air masses originating in North-East India itself were seen to be transported to Jorhat and Itanagar. However Imphal is noted to receive a couple of trajectories depicting long range transport from the western coast of India. Most of the air masses sampled along the cruise had originated in the IGP and could thus lead to the influx of continental aerosols into the marine atmosphere.

3. Results and Discussion

The mean and ranges of mass concentrations of ambient $PM_{2.5}$, PM_{10} and TSP observed at the sampling stations and in BoB for the duration of the study period as well as the $PM_{2.5}/PM_{10}$ and $PM_{2.5}/TSP$ ratios were computed to determine the sites where the fine particulate loading in the ambient atmosphere overwhelms the coarser fractions and is outlined in Table 3.

In the present study, Varanasi (187.8 \pm 36.5 µg m⁻³) was observed to have the highest concentration of fine mode PM_{2.5} aerosols followed by two other major metropolitan cities, New Delhi (178.2 \pm 56.5 µg m⁻³) and Kolkata $(168.7 \pm 77.9 \ \mu g \ m^{-3})$. Patiala averaged a slightly lower PM_{2.5} concentration (138.7 \pm 27.0 µg m⁻³) while Agra and Lucknow had almost similar concentrations of 120.6 \pm 36.2 μ g m⁻³ and 120.4 \pm 47.8 μ g m⁻³ respectively. Kullu $(20.4 \pm 3.1 \ \mu g \ m^{-3}),$ Kadapa $(PM_{2.1} = 34.1 \pm$ 4.6 μ g m⁻³), Darjeeling (43.3 ± 19.7 μ g m⁻³), Bhubaneswar (65.2 \pm 11.8 µg m⁻³) as well as all the sampling locations in North-East India, viz. Jorhat (43.6 \pm 12.3 μ g m⁻³), Itanagar (77.4 ± 27.1 μ g m⁻³) and Imphal $(87.4 \pm 31.9 \ \mu g \ m^{-3})$ had fine particulate mass concentrations lower than 100 μ g m⁻³. The daily average PM_{2.5} at all the stations, except Kullu, Darjeeling, Jorhat and Kadapa, exceed the value set by the National Ambient Air Quality Annual Standard of India (40 μ g m⁻³). The PM_{2.5} mass concentration observed in the BoB was ranging from 15.0 to 54.4 μ g m⁻³ with a mean concentration of 41.3 \pm 11.9 $\mu g m^{-3}$.

If we compare our observation with sporadically measurement over different sites, one could find that the average PM25 mass concentration prevailing over Himachal Pradesh was observed to be $41.78 \pm 7.92 \ \mu g \ m^{-3}$ that over Kullu was estimated to while be $34.32 \pm 1.32 \ \mu g \ m^{-3}$ by Sharma et al. [12] during a campaign conducted in the period from 12 to 22 March, 2013. Delhi was reported to have an annual mean PM_{2.5} concentration of 97 \pm 56 µg m⁻³ in 2007 [16]. In another study, PM_{2.5} mass concentration during winter over Delhi was observed to be $151.2 \pm 26.1 \ \mu g \ m^{-3}$ [29]. Barman et al. [30] carried out a study to discern the mass concentrations of fine particulates in two residential areas of Lucknow in November, 2008 and reported the 24 h average PM_{2.5} to be 131.74 μ g m⁻³ and 115.83 μ g m⁻³ respectively. The fine particulate concentration in Lucknow was estimated to be $101.05 \pm 22.55 \ \mu g \ m^{-3}$ (range 75.87–129.85 μ g m⁻³) while the winter season average was $212.35 \pm 55.0 \ \mu g \ m^{-3}$ (99.3–299.3 $\ \mu g \ m^{-3}$) [17]. In

of
period
the
during
sites
sampling
the
at
conditions
Meteorological
2
ble

Table 2	Meteorologic	conditions a	t the sampling sites	s during the period	of study							
Sites	Temperature	(C) (C)	Pressure (mb)		RH (%)		Rainfall (1	mm)	Wind Spee	ed (km h ⁻¹)	Wind Directio	(N°) n
	Range	$\text{Mean}\pm\text{SD}$	Range	$Mean\pm SD$	Range	$Mean\pm SD$	Range	$Mean\pm SD$	Range	$\text{Mean}\pm\text{SD}$	Range	$Mean\pm SD$
Site 2	12.7–21.3	16.9 ± 2.7	978.7–988.8	985.4 ± 3.4	40.4-89.0	62.2 ± 14.2	0.0 - 0.4	0.0 ± 0.1	0.0-8-0	2.6 ± 2.7	0.0-360.0	229.7 ± 96.4
Site 3	11.3-18.3	14.5 ± 1.8	na	na	67.5-97.0	82.2 ± 9.5	0.0 - 10.0	0.7 ± 2.4	2.0-7.0	4.0 ± 1.6	na	na
Site 4	12.0-21.0	16.5 ± 2.3	955.3–967.5	962.6 ± 2.5	50.0-99.0	72.4 ± 15.4	0.0 - 7.0	0.6 ± 1.8	2.0 - 13.0	5.3 ± 2.9	105-281	177.8 ± 43.1
Site 5	12.0-25.0	18.0 ± 3.9	988.3-998.2	994.7 ± 3.1	47.2–94.5	78.7 ± 16.7	0.0 - 0.0	0.0 ± 0.0	0.5-5.0	2.2 ± 1.5	0.0 - 360.0	231.7 ± 80.0
Site 6	12.0-21.0	15.2 ± 3.0	1,010.0-1,012.0	$1,016.6 \pm 4.2$	55.0-93.0	77.8 ± 11.2	0.0 - 2.0	0.1 ± 0.5	5.0 - 11.0	7.4 ± 1.9	0.0 - 350.0	246.5 ± 84.8
Site 7	11.6-19.2	14.8 ± 2.0	998.8-1,010.7	$1,005.8 \pm 3.1$	75.4-99.3	93.9 ± 6.4	0.0 - 0.0	0.0 ± 0.0	2.6-12.0	5.4 ± 2.3	106.8–290.9	220.2 ± 61.8
Site 8	6.2 - 28.0	17.3 ± 1.2	na	na	49.0-100.0	85.4 ± 7.6	0.0 - 0.0	0.0 ± 0.0	5.6-42.2	21.7 ± 9.3	135–315	272 ± 21.7
Site 9	14.6-20.2	16.8 ± 2.1	1,006.7 - 1,017.4	$1,012.6 \pm 3.7$	68.2-86.7	75.6 ± 5.1	0.0 - 0.0	0.0 ± 0.0	0.6 - 0.0	2.4 ± 1.9	0.0 - 360.0	282.7 ± 77.7
Site 10	9.7 - 20.0	13.9 ± 2.6	889.2-897.5	894.2 ± 2.3	53.4-83.4	68.1 ± 9.3	0.0 - 0.0	0.0 ± 0.0	0.0 - 6.0	2.4 ± 1.4	0.0 - 350.0	232.8 ± 94.2
Site 11	4.3-29.5	14.7 ± 3.6	991.4-1,012.6	$1,004.2 \pm 3.1$	45.6–99.2	77.8 ± 12.6	0.0 - 1.1	0.4 ± 0.62	0.0 - 4.1	0.69 ± 1.26	na	na
Site 12	6.8–27.6	11.0 ± 2.6	1,002.6 - 1,010.6	$1,006.3 \pm 2.42$	38.0-99.0	87.6 ± 14.3	0.0 - 0.8	0.02 ± 0.14	0.0-1.4	0.36 ± 0.54	na	na
Site 13	5.6-30.5	10.4 ± 5.5	913.5-1,004.8	998.7 ± 6.4	40.2–98.5	67.8 ± 10.2	0.0 - 0.0	0.0 ± 0.0	0.0–3.7	0.41 ± 1.3	na	na
Site 14	20.7-24.8	22.0 ± 1.1	997.1-1,006.5	$1,002.7 \pm 2.8$	59.2-86.8	73.3 ± 6.0	0.0 - 0.0	0.0 ± 0.0	2.1–3.7	2.8 ± 0.4	162.7–254.3	200.3 ± 33.2
Site 15	18.6-28.1	23.1 ± 2.4	989.5–997.5	993.8 ± 1.5	47.0–90.5	68.6 ± 11.9	0.0 - 0.0	0.0 ± 0.0	0.0–9.8	3.1 ± 1.6	0.0 - 360.0	92.6 ± 80.2
Site 16	15.7-28.7	24.4 ± 2.0	1,010.1-1,018.9	$1,013.4\pm1.7$	42.6–94.3	69.5 ± 8.4	0.0 - 0.0	0.0 ± 0.0	0.7–21.0	10.0 ± 5.0	5.0-359.0	76.0 ± 83.8
na not a	vailable											

Table 3 Average and range of mass concentrations of $PM_{2.5}$, PM_{10} and TSP at the sampling sites

Sites	Location	$PM_{2.5} \ (\mu g \ m^{-3})$		$PM_{10} \ (\mu g \ m^{-3})$		TSP ($\mu g m^{-3}$)		PM _{2.5} /	PM _{2.5} /
		Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	PM_{10}	TSP
Site 1	Kullu	9.0–22.0	20.4 ± 3.1	16.5–96.5	46.9 ± 19.1	29.7-112.5	66.2 ± 27.7	0.44	0.31
Site 2	Patiala	93.4-201.9	138.7 ± 27.0	na	na	68.6–348.7	250.6 ± 68.2		0.55
Site 3	Delhi	114.6–253.9	178.2 ± 56.5	89.7-358.2	205.7 ± 71.3	99.2-451.3	259.0 ± 92.9	0.87	0.69
Site 4	Ajmer	na	na	28.8-89.9	57.6 ± 18.7	na	na		
Site 5	Agra	35.5-180.6	120.6 ± 36.2	43.6-201.1	152.7 ± 43.7	50.1-251.4	174.2 ± 49.6	0.79	0.69
Site 6	Lucknow	51.8-233.9	120.4 ± 47.8	101.1-348.8	186.9 ± 67.3	220.4-536.6	325.0 ± 71.5	0.64	0.37
Site 7	Varanasi	125.6-256.2	187.8 ± 36.5	131.8-335.3	235.1 ± 57.6	na	na	0.80	
Site 8	Giridih	na	na	126.3-328.3	223.9 ± 53.5	na	na		
Site 9	Kolkata	71.7–396.4	168.7 ± 77.9	147.6-520.1	272.6 ± 102.9	na	na	0.62	
Site 10	Darjeeling	16.8-92.2	43.3 ± 19.7	59.8-78.9	67.4 ± 5.8	na	na	0.64	
Site 11	Itanagar	37.3-86.9	77.4 ± 27.1	156.6-306.7	267.8 ± 56.7	108.3-521.1	294.6 ± 78.5	0.29	0.26
Site 12	Jorhat	16.8-51.6	43.6 ± 12.3	76.0-237.5	193.5 ± 46.1	75.4–572.0	287.9 ± 92.3	0.23	0.15
Site 13	Imphal	41.5-116.7	87.4 ± 31.9	124.6-326.3	277.6 ± 61.2	121.9-567.4	306.4 ± 86.4	0.32	0.29
Site 14	Bhubaneswar	49.6-87.7	65.2 ± 11.8	92.0-191.7	133.9 ± 30.6	na	na	0.49	
Site 15	Kadapa	30.4-39.8*	$34.1 \pm 4.6^{*}$	48.5-70.7	60.1 ± 8.2	30.2-101.2	62.9 ± 17.0	0.57#	$0.54^{\$}$
Site 16	Bay of Bengal	15.0–54.4	41.3 ± 11.9	30.1-82.1	53.9 ± 18.9	49.1–184.5	78.8 ± 29.7	0.77	0.52

na not available

[#] PM_{2.1}/PM₁₀

^{\$} PM_{2.1}/TSP

another study, the average mass concentration of PM_{2.5} over Lucknow in the period from 2007 to 2009 was $87.3 \pm 64.2 \ \mu g \ m^{-3}$ (range be quantified to 28.0–196.9 μ g m⁻³) by Pandey et al. [31]. Annual average PM_{2.5} mass concentrations for Agra was estimated to be $104.9 \pm 47.1 \ \mu g \ m^{-3}$ and $91.1 \pm 50.3 \ \mu g \ m^{-3}$ at urban and rural sites respectively [18]. Pachauri et al. [15] also carried out sampling for ambient particulates across different sites in Agra and reported average mass concentrations of $308.3 \pm 51.8 \,\mu\text{g/m}^3$ (high traffic area). $140.8 \pm 22.3 \ \mu g \ m^{-3}$ (campus area) and $91.2 \pm 17.3 \ \mu g \ m^{-3}$ (rural area). Das et al. [32] reported 24 h average PM_{2.5} mass concentration of $178.57 \pm 31.53 \ \mu g \ m^{-3}$ (range 112.42–201.96 $\mu g \ m^{-3}$) in Kolkata while Gupta et al. [33] reported the same to be $181.19 \pm 90.86 \ \mu g \ m^{-3}$ (range $49.47-537.77 \ \mu g \ m^{-3}$). The PM_{2.5} concentration during winter in Jorhat was reported to be 143.5 \pm 23 µg m³ [34]. PM_{2.5} mass concentrations over the BoB were found to range from 2.0 to 76.7 μ g m⁻³ during 27 December, 2008 to 28 January, 2009 in the study conducted by Kumar et al. [25].

In this present study the sites in the upper and middle IGP region (Patiala, New Delhi, Agra, Lucknow and Varanasi) recorded elevated concentrations of fine particulates as expected and could be attributed to the fact that the winter in these regions are characterized by low and stable planetary boundary layer and relatively low wind speeds leading to stagnation of pollutants due to little dispersion in the lower atmosphere [35]. In addition, during winter the increase in the usage of biomass for heating purposes might have caused an increase in fine particle loading into the atmosphere [15, 16]. Kolkata, being located in the lower IGP recorded very high PM2.5 mass concentrations. The high concentrations of fine particulates at all of these sites can be attributed to the fact that these cities are highly populated metropolitan cities with a very high associated fine mode aerosol contribution from fossil fuel burning including vehicular exhaust. The average PM_{2.5} mass concentration over Kolkata was found to be similar to that reported by Das et al. [32] (179 μ g m⁻³) for the winter season. Moreover, elevated PM_{2.5} concentrations at these sites could be attributed to the fact that the air masses received at these sites were observed to originate and undergo transport in the IGP itself and could thus be carrying fine mode continental pollutants to these sites. The gradient between the continental and marine (BoB) PM_{2.5} concentrations suggest the possibility of continental outflow of pollutants to the clean pristine marine environment. All the sites located outside the IGP and thereby outside the fringes of the north-easterly flow had relatively lower PM_{2.5} concentrations than the sites in the IGP. Low fine particulate mass concentrations observed at Kullu and Darjeeling could be because they are located in the Himalayas and have pristine Himalayan air. In Fig. 2, all

^{*} PM_{2.1}

the air mass trajectories received at Kadapa during the period of this study had to traverse long distances across the BoB and this could possibly have led to the dilution of fine particulate concentrations at the site. Balakrishnaiah et al. [36] reported the similar order of concentration of fine particulate over Anantapur (21.29 \pm 2.31 µg m⁻³), a tropical semi-arid site located nearby, during the winter season. Though the air masses received at Bhubaneswar, a coastal station, originated from IGP, traversed a short distance across northern BoB encountering marine environment, this could be the prime cause for the relatively low PM_{2.5}. North-East India has dense forest cover and lower anthropogenic pollutant emissions compared to the sites in the IGP, as was observed in the low PM_{2.5} mass concentrations of Jorhat, Itanagar and Imphal. Imphal was seen to receive a few air masses which had to traverse across India picking up continental pollutants along the way, thereby resulting in it having the highest PM_{2.5} concentrations among the three stations.

The highest PM_{10} mass concentrations recorded over Kolkata (272.6 ± 102.9 µg m⁻³), Varanasi (235.1 ± 57.6 µg m⁻³), Giridih (223.9 ± 53.5 µg m⁻³), Delhi

 $(205.7 \pm 71.3 \ \mu g \ m^{-3})$ and all the sampling locations in North-East India, viz. Jorhat (193.5 \pm 46.1 µg m⁻³), Itanagar (267.8 \pm 56.7 μ g m⁻³) and Imphal (277.6 \pm 61.2 μ g m⁻³). The sampling sites which are located outside the IGP such as Kullu (46.9 \pm 19.1 µg m⁻³), Darjeeling $(67.4 \pm 5.8 \ \mu g \ m^{-3})$, Kadapa $(60.1 \pm 8.2 \ \mu g \ m^{-3})$ and Ajmer (57.6 \pm 18.7 µg m⁻³) were mostly observed to have the lowest PM₁₀ concentration during campaign period within the National Ambient Air Quality Annual Standard (60 μ g m⁻³). These values were comparable to that of BoB $(53.9 \pm 18.9 \ \mu g \ m^{-3}$, range 30.1 to 82.1 $\mu g \ m^{-3}$). Delhi (0.87) exhibited the highest ratio of PM_{2.5}/PM₁₀, followed by Varanasi (0.80) and Agra (0.79) while Jorhat (0.23), Itanagar (0.29) and Imphal (0.32) have the lowest PM_{2.5}/PM₁₀ ratio. The two coastal stations, i.e. Kolkata (0.62) and Bhubaneswar (0.49) were noted to have intermediate PM_2 5/PM₁₀ ratios.

The present observation of PM_{10} concentration over Delhi is very much similar to earlier observations (Tiwari et al. [16]: 219 ± 84 µg m⁻³ in 2007; Sharma et al. [22]: 213.1 ± 15.0 µg m⁻³; Sharma et al. [37]: 191.4 ± 45.5 µg m⁻³ in 2010–2011). Sharma et al. [37] reported higher concentration of PM_{10} (241.4 ± 50.5 µg m⁻³) during the winter



Fig. 2 Air mass HYSPLIT back trajectories plotted for the land stations and along the cruise track for the duration of the sampling period

season of 2010-2011. Over Lucknow, the PM₁₀ mass concentration was quantified to be $204.07 \pm 26.75 \ \mu g \ m^{-3}$ (range 163.96–218.43 μ g m⁻³) with the winter season average of the order of $269.32 \pm 42.9 \ \mu g \ m^{-3}$ (99.3– 299.3 μ g m⁻³) and the annual PM₂ ₅/PM₁₀ mass fraction ratio being 0.42 ± 0.26 [17]. In another study, Pandey et al. [31] reported the PM₁₀ mass concentrations over Lucknow during 2007 to 2009 as $168.1 \pm 29.1 \ \mu g \ m^{-3}$ (range 102.3– 240.5 μ g m⁻³). Sharma et al. [38] reported PM₁₀ mass concentrations in residential, commercial and industrial sectors of Lucknow to be 146.9, 178.4 and 107.6 μ g m⁻³ respectively. Annual average PM₁₀ mass concentrations at urban and rural sites in Agra was reported to be 154.2 ± 68.0 and $148.4 \pm 64.4 \ \mu g \ m^{-3}$ respectively [18]. The computed 24 h average PM₁₀ mass concentration in Kolkata was quantified to be $303.85 \pm 49.48 \ \mu g \ m^{-3}$ (range 201.33–347.18 $\mu g \ m^{-3}$) [32] while the PM_{2.5}/PM₁₀ ratio was 0.59 ± 0.032 . Meanwhile in a similar study conducted by Gupta et al. [33] in Kolkata the PM₁₀ mass concentration was 288.93 \pm 123.52 μ g m⁻³ (range 66.66–637.70 μ g m⁻³). The PM₁₀ concentration during winter in Jorhat was measured to be $222.9 \pm 47 \ \mu g \ m^{-3}$ [34]. PM₁₀ mass concentrations over the BoB were found to range from 6.5 to 108 μ g m⁻³ in the study conducted by Kumar et al. [25]. In another study conducted in BoB during the same study period average PM₁₀ mass concentrations of 55.9 \pm 26.3 µg m⁻³ (range 22.5–107.2 µg m⁻³) was reported [21].

Kolkata recorded the highest PM₁₀ mass concentrations $(272.6 \pm 102.9 \ \mu g \ m^{-3})$ during the study period; but the PM_{2.5}/PM₁₀ ratio (0.64) in Kolkata was much lower than most other sites in the IGP. This could be due to the fact that it is a coastal station leading to the loading of coarse mode marine aerosols in addition to the influx of finer continental particulates from the IGP and the local pollutant emissions. The other coastal station, viz. Bhubaneswar was noted to have quite a low $PM_{2.5}/PM_{10}$ ratio (0.49), representing almost equal loading of fine and coarse mode particulates. Both Varanasi and Delhi were observed to have high PM₁₀ concentrations (>205 μ g m⁻³). They also have the highest PM_{2.5}/PM₁₀ ratios (Delhi: 0.87, Varanasi: 0.80), signifying that almost all of the PM₁₀ mass at these sites is made up of fine mode particulates. Both these sites are located in the IGP and have very high fine particulate emissions locally as well as input from the surrounding IGP region, as suggested by trajectory analysis. Giridih is another site with a relatively high PM₁₀ mass concentration which could be due to coal mining and associated activities near the site. Moreover all the air masses received by Giridih were seen to originate and traverse across the IGP, thereby leading to an influx of continental pollutants from the IGP into that region. It is observed among the stations in the IGP that the PM₁₀ mass concentrations are the highest in the lower IGP region compared to the middle and upper IGP regions, suggesting the possibility of outflow to the clean marine region. Low particulate concentrations at Kullu and Darjeeling could be attributed to the fact that these stations are located at elevated altitudes and have pristine Himalavan air with little local generation of aerosols. Ajmer was noted to have a low PM₁₀ concentration which can be accounted for by the fact that air mass sampling was carried out in a university campus (Central University of Rajasthan) with few local sources of aerosols. Moreover, Ajmer is surrounded by the Aravalli Range, which could result in lower windblown dust from the adjoining Thar Desert. Kadapa was observed to have a $PM_{2.5}/PM_{10}$ ratio of 0.57 with fine particulates accounting for a little more than half of the PM₁₀ mass. Absence of rainfall, low RH and cloud free skies prevailing at the site during the sampling period were conducive to the enhanced loading of fine continental aerosols. Trajectory analysis suggests that the influx of marine coarse mode aerosols from the BoB to Kadapa could be adding to the relatively low local input of coarse particulates into the atmosphere, resulting in an intermediate PM2.5/PM10 ratio. PM10 concentrations observed for the North-Eastern sites broadly concur ($\pm 50 \text{ }\mu\text{g m}^{-3}$) with the concentration reported for Jorhat during winter by Khare et al. [34]. The PM_{2.5}/PM₁₀ (0.77) ratio for BoB was observed to be quite high compared to a majority of the land stations. This could be due to the fact that the proximity of the cruise track to India allowed sampling of continental pollutants which had undergone long range transport and outflowed into the BoB during the sampling period due to favorable wind patterns. The trajectory analysis performed along the cruise track also suggests that the air masses encountered in the BoB were originating in and travelling across the IGP, and could thus be capable of picking up fine mode pollutants along its path from those regions.

Among the few selected sites where TSP could be monitored, Lucknow (325.0 \pm 71.5 µg m⁻³) had the highest, $(62.9 \pm 17.0 \ \mu g \ m^{-3})$ while Kadapa and Kullu $(66.2 \pm 27.7 \ \mu g \ m^{-3})$ had the lowest TSP mass concentrations in India. TSP mass concentration was found to average $78.8 \pm 29.7 \ \mu g \ m^{-3}$ (range $49.1 - 184.5 \ \mu g \ m^{-3}$) over the BoB. In an earlier study, TSP mass concentrations in BoB was found to average $22 \ \mu g \ m^{-3}$ (range 5–47 μ g m⁻³) with the mean TSP concentration in northern BoB being 41 μ g m⁻³ and the average concentration for the rest of BoB being only 16 μ g m⁻³ [39]. It is also interesting to note that the PM_{2 5}/TSP ratio in Lucknow was only 0.37 was much lower than Delhi (0.69) and Agra (0.69) which had the highest contribution of fine particulates to the total TSP loading. The PM_{2 5}/TSP (0.52) ratio was observed to be quite high for BoB, indicating that more than half of the total suspended particulates in the bay during the study period were made up of fine particulates and providing further evidence of the influx of continental particulates into BoB.

This study was aimed at estimating the mass concentrations of $PM_{2.5}$ and PM_{10} size fractions as well as TSP in the ambient atmosphere at different sites across India as well as the BoB. The salient features of this study are as follows:

- The IGP region (Patiala, Varanasi, Lucknow, Kolkata etc.) had the highest concentration of $PM_{2.5}$ suggesting that anthropogenic activity e.g., biomass burning occurring there could outflow to the clean marine region over Bay of Bengal. However two sites (Darjeeling and Kadapa) which are outside the fringes of the north-east flow reported low $PM_{2.5}$ (43.3 ± 19.7 µg m⁻³) and $PM_{2.1}$ (34.1 ± 4.6 µg m⁻³) concentrations.
- As expected, the lower IGP region was observed to have the highest PM_{10} concentration compared to the upper and middle IGP region. Kolkata recorded the highest average PM_{10} (272.6 ± 102.9 µg m⁻³) concentration across India while Kullu (46.9 ± 19.1 µg m⁻³) had the lowest mass concentration. Darjeeling (67.4 ± 5.8 µg m⁻³), Ajmer (57.6 ± 18.7 µg m⁻³) and Kadapa (60.1 ± 8.2 µg m⁻³) had very low PM₁₀ concentrations and were found to be comparable to Bay of Bengal (53.9 ± 18.9 µg m⁻³)
- In the north eastern region of India, highest concentration for $PM_{2.5}$, PM_{10} and TSP was observed at Imphal (87.4 ± 31.9, 277.6 ± 61 and 306.4 ± 86.4 µg m⁻³ respectively) while the lowest concentration of the same were noted at Jorhat (43.6 ± 12.3, 193.5 ± 46.1 and 287.9 ± 92.3 µg m⁻³ respectively).
- Lucknow recorded the highest average TSP $(325.0 \pm 71.5 \ \mu g \ m^{-3})$ concentration across India while Kadapa had the lowest (62.9 \pm 17.0 $\mu g \ m^{-3}$). TSP mass concentration in BoB was (78.8 \pm 29.7 $\mu g \ m^{-3}$).
- Delhi and Agra recorded very high PM_{2.5}/PM₁₀ and PM_{2.5}/TSP ratios while Varanasi displayed a high PM_{2.5}/PM₁₀ ratio. This is indicative of a higher loading of fine ambient pollutant particulates compared to the natural coarser aerosols.
- Similarly both the $PM_{2.5}/PM_{10}$ (0.77) and $PM_{2.5}/TSP$ (0.52) ratios in the BoB were observed to be on the higher side signifying outflow of fine pollutants from the landmasses.

Chemical properties (inorganic ionic composition, carbonaceous fraction and metal fraction of $PM_{2.5}$, PM_{10} and TSP) may lead to more evidence of the outflow of continental pollutants to the clean marine region. This will be reported in future.

Acknowledgments We acknowledge the Indian Meteorological Department for allowing the usage of the meteorological data at

respective sites. The authors (TKM, SKS, MS) are thankful to Director, NPL, New Delhi and Head, Radio and Atmospheric Sciences Division, NPL, New Delhi for the encouragement and support rendered during the course of this study. The authors (JCK, PS) are heartily thankful to Director, G.B. Pant Institute of Himalayan Environment and Development, Kosi-Katarmal, Almora, Uttarakhand, India for providing facilities at Mohal-Kullu, Himachal Unit of the Institute. Authors (TB, MK) duly acknowledge the financial support provided by University Grants Commission, New Delhi for sampling at BHU, Varanasi. One of the authors (AKK) is grateful to Director, IITR, Lucknow for his encouragement and support and permission to participate in this study. Author (JCK) is thankful to ISRO-GBP, ARFI and DBT. Author (YNA) is thankful to ISRO-GBP for their financial assistance in the form of Research project under ARFI Network stations for sampling at Mohal-Kullu and Kadapa. One of the Authors (G RB) is also thankful to DST for awarding JRF under INSPIRE Program. The authors (BPB, JS, PS) are also grateful to Director, CSIR-NEIST, Jorhat for his encouragement and support. Authors are also thankful for the efforts made by the members of two branch labs of CSIR-NEIST, viz., Itanagar and Imphal. Authors (AC, SKG) acknowledge Prof Sibaji Raha for all support for carrying out the observation at Kolkata and Darjeeling and the technical support of Mr. Sabyasachi Majee, Darjeeling and Mr. Bhaskar Ray and Miss. Debolina Seal, Kolkata. The authors (NCD and SP) are thankful to Director, Indian Statistical Institute and HRDG, CSIR for their encouragement and financial support for this study at Giridih. A. Sen is grateful to HRDG, CSIR for providing SPM fellowship. We acknowledge the help and support rendered by crew members and Caption (SK308) in allowing us to collect samples over the BoB as a piggy back of the SIBER-India program. The authors (TD, PSM, SP) are thankful to Director, CSIR-IMMT and Head, Environment and Sustainability Department of CSIR-IMMT for their encouragement and financial support. One of the authors (AS) is thankful to ISRO/ DOS for fellowship under ATCTM project.

References

- R.J. Charlson, S.E. Schwartz, J.M. Hales, R.D. Cess, J.J. Coakley, J.E. Hansen, D.J. Hofmann, Climate forcing by anthropogenic aerosols, Science, 255(5043) (1992) 423–430.
- [2] J. Hansen, M. Sato, R. Ruedy, Radiative forcing and climate response, J. Geophys. Res. Atmos. (1–10), **102**(D6) (1997) 6831–6864.
- [3] C.A. Pope III, R.T. Burnett, M.J. Thun, E.E. Calle, D. Krewski, K. Ito, G.D. Thurston, Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, Jama, 287(9) (2002) 1132–1141.
- [4] C. Varotsos, Airborne measurements of aerosol, ozone, and solar ultraviolet irradiance in the troposphere, J. Geophys. Res. Atmos., **110** (2005) D09202, doi:10.1029/2004JD005397.
- [5] C. Tzanis, C. Varotsos, M. Ferm, J. Christodoulakis, M.N. Assimakopoulos, C. Efthymiou, Nitric acid and particulate matter measurements at Athens, Greece, in connection with corrosion studies, Atmos. Chem. Phys., 9(21) (2009) 8309–8316.
- [6] C. Tzanis, C. Varotsos, J. Christodoulakis, J. Tidblad, M. Ferm, A. Ionescu, R.A. Lefevre, K. Theodorakopoulou, K. Kreislova, On the corrosion and soiling effects on materials by air pollution in Athens, Greece, Atmos. Chem. Phys., 11(23) (2011) 12039–12048.
- [7] M.G. Lawrence, J. Lelieveld, Atmospheric pollutant outflow from southern Asia: a review, Atmos. Chem. Phys. Discuss., 10(4) (2010) 11017–11096.
- [8] T. Saud, M. Saxena, D.P. Singh, Saraswati, Manisha Dahiya, S.K. Sharma, A. Datta, Ranu Gadi, T.K. Mandal, Chemical

composition of aerosol particles from direct emissions of biomass fuel used in Indo Gangetic Plain, Atmos. Environ., **71** (2013) 158–169.

- [9] S. Menon, J. Hansen, L. Nazarenko, Y. Luo, Climate effects of black carbon aerosols in China and India, Science, 297(5590) (2002) 2250–2253.
- [10] M.V.S.N. Prasad, C. Sharma, B.C. Arya, T.K. Mandal, S. Singh, M.J. Kulshrestha, R. Agnihotri, S.K. Mishra, S.K. Sharma, Experimental facilities to monitor various types of atmospheric parameters in the Radio and Atmospheric Sciences Division (RASD) of CSIR-National Physical Laboratory, Mapan-Journal of Metrology Society of India, 28(3) (2013) 193–203.
- [11] S.K. Sharma, T.K. Mandal, Rohtash, M. Kumar, N.C. Gupta, H. Pathak, R.C. Harit, M. Saxena, Measurement of ambient ammonia over the National Capital Region of Delhi, India, Mapan-Journal of Metrology Society of India, (2014a). doi: 10.1007/s12647-014-0098-9.
- [12] S.K. Sharma, T.K. Mandal, C. Sharma, J.C. Kuniyal, R. Joshi, P.P. Dhayani, Rohtash., A. Sen, H. Ghayas, N.C. Gupta, B.C. Arya, A. Kumar, P. Sharma, M. Saxena, and A. Sharma, Measurements of particulate (PM_{2.5}), BC and trace gases over the northwestern Himalayan region of India, Mapan-Journal of Metrology Society of India, (2014b). doi:10.1007/s12647-014-0104-2.
- [13] P.S. Monks, C. Granier, S. Fuzzi, A. Stohl, M. L. Williams, H. Akimoto, M. Amann, A. Baklanov, U. Ballensperger, I. Bey, N. Blake, R.S. Blake, K. Carslaw, O.R. Cooper, F. Denfener, D. Fowler, E. Fragkou, G.J. Frost, S. Generoso, F. Ginoux, V. Grewe, A. Guenther, H.C. Hansson, S. Henne, S. Hjorth, A. Hofzumahaus, H. Huntrieser, I.H.A. Isaksen, M.E. Jenkin, J. Kaiser, M. Kanakidou, Z. Klimont, M. Kulmala, P. Laj, M.G. Lawrence, J.D. Lee, C. Liousse, M. Maione, G. McFiggans, A. Metzger, A. Mieville, N. Moussiopoulos, J.J. Orlando, C.D. O'Dowd, P.J. Palmer, D.D. Parrish, A. Petzold, U. Platt, U. Poschl, A.S.H. Prevot, Atmospheric composition change–global and regional air quality, Atmos. Environ., **43**(33) (2009) 5268–5350.
- [14] K.K. Moorthy, S.K. Satheesh, S.S. Babu, C.B.S. Dutt, Integrated campaign for aerosols, gases and radiation budget (ICARB): an overview, J. Earth Syst. Sci., 117(1) (2008) 243–262.
- [15] T. Pachauri, A. Satsangi, V. Singla, A. Lakhani, K.M. Kumari, Characteristics and sources of carbonaceous aerosols in PM_{2.5} during wintertime in Agra, India, Aerosol Air Qual. Res., **13**(3) (2013) 977–991.
- [16] S. Tiwari, D.M. Chate, P. Pragya, K. Ali, D.S. Bisht, Variations in Mass of the PM_{10} , $PM_{2.5}$ and PM_1 during the Monsoon and the Winter at New Delhi, Aerosol Air Qual. Res., **12**(1) (2012) 20–29.
- [17] P. Pandey, A.H. Khan, A.K. Verma, K.A. Singh, N. Mathur, G.C. Kisku, S.C. Barman, Seasonal trends of PM2.5 and PM10 in ambient air and their correlation in ambient air of Lucknow city, India, Bull. Environ. Contam. Toxicol., 88(2) (2012) 265–270.
- [18] Kulshrestha, P.G. Satsangi, J. Masih, A. Taneja, Metal concentration of $PM_{2.5}$ and PM_{10} particles and seasonal variations in urban and rural environment of Agra, India, Sci. Total Environ., **407**(24) (2009) 6196–6204.
- [19] L.A.K. Reddy, U.C. Kulshrestha, J. Satyanarayana, M.J. Kulshrestha, K. Krishna Moorthy, Chemical characteristics of PM10 aerosols and airmass trajectories over Bay of Bengal and Arabian Sea during ICARB, J. Earth Syst. Sci., **117**(1) (2008) 345–352.
- [20] K.M. Latha, K.V.S. Badarinath, Seasonal variations of black carbon aerosols and total aerosol mass concentrations over urban environment in India, Atmos. Environ., **39**(22) (2005) 4129–4141.

- [21] S.K. Sharma, A.K. Singh, T. Saud, T.K. Mandal, M. Saxena, S. Singh, S.K. Ghosh, S. Raha, Study on water-soluble ionic composition of PM10 and related trace gases over Bay of Bengal during W_ICARB campaign, Meteorol. Atmos. Phys., **118**(1–2) (2012a) 37–51.
- [22] S.K. Sharma, M. Saxena, T. Saud, S. Korpole, T.K. Mandal, Measurement of NH3, NO, NO2 and related particulates at urban sites of Indo Gangetic Plain (IGP) of India, J. Sci. Ind. Res., 71(5) (2012b) 360–362.
- [23] D.G. Kaskaoutis, M.C.R. Kalapureddy, K. Krishnamoorthy, P.C.S. Devara, P.T. Nastos, P.G. Kosmopoulos, H.D. Kambezidis, Heterogeneity in pre-monsoon aerosol types over the Arabian Sea deduced from ship-borne measurements of spectral AODs, Atmos. Chem. Phys., **10**(10) (2010) 4893–4908.
- [24] V. Ramanathan, P.J. Crutzen, J. Lelieveld, A.P. Mitra, D. Althausen, J. Anderson, M.O. Andreae, W. Cantrell, G.R. Cass, C.E. Chung, A.D. Clarke, J.A. Coakley, W.D. Collins, W.C. Conant, F. Dulac, J. Heintzenberg, A.J. Heymsfield, B. Holben, S. Howell, J. Hudson, A. Jayaraman, J.T. Kiehl, T.N. Krishnamurti, D. Lubin, G. McFarquhar, T. Novakov, J.A. Ogren, I.A. Podgorny, K. Prather, K. Priestley, J.M. Prospero, P.K. Quinn, K. Rajeev, P. Rasch, S. Rupert, R. Sadourny, S. K. Satheesh, G. E. Shaw, P. Sheridan, and F.P.J. Valero, Indian Ocean Experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, J. Geophys. Res. Atmos. (1984–2012), **106**(D22) (2001) 28371–28398.
- [25] Kumar, M.M. Sarin, B. Srinivas, Aerosol iron solubility over Bay of Bengal: Role of anthropogenic sources and chemical processing, Mar. Chem., **121**(1) (2010) 167–175.
- [26] K.K. Moorthy, S.N. Beegum, S.S. Babu, A. Smirnov, S.R. John, K.R. Kumar, K. Narasimhulu, C.B.S. Dutt, V.S. Nair, Optical and physical characteristics of Bay of Bengal aerosols during W-ICARB: spatial and vertical heterogeneities in the marine atmospheric boundary layer and in the vertical column, J. Geophys. Res. Atmos. (1984–2012), **115**(D24) (2010).
- [27] S.R. Smith, M.A. Bourassa, R.J. Sharp, Establishing more truth in true winds, J. Atmos. Ocean. Technol., 16(7) (1999) 939–952.
- [28] P.K. Taylor, E.C. Kent, The accuracy of meteorological observations from Voluntary Observing ships: Present status and future requirements. First Session of the CMM Subgroup on Voluntary Observing Ships, Athens, 8–12 March, (1999).
- [29] S.K. Sharma, M. Kumar, N.C. Gupta, M. Saxena, T.K. Mandal, Characteristics of ambient ammonia over Delhi, India, Meteorol. Atmos. Phys., 124(1–2) (2014c) 67–82.
- [30] S.C. Barman, R. Singh, M.P.S. Negi, S.K. Bhargava, Fine particles (PM2. 5) in residential areas of Lucknow city and factors influencing the concentration, Clean–Soil Air Water, 36(1) (2008) 111–117.
- [31] P. Pandey, D.K. Patel, A.H. Khan, S.C. Barman, R.C. Murthy, G.C. Kisku, Temporal distribution of fine particulates (PM_{2.5}, PM₁₀), potentially toxic metals, PAHs and Metal-bound carcinogenic risk in the population of Lucknow City, India, J. Environ. Sci. Health Part A, **48**(7) (2013) 730–745.
- [32] M. Das, S.K. Maiti, U. Mukhopadhyay, (2006). Distribution of PM2.5 and PM10-2.5 in PM10 fraction in ambient air due to vehicular pollution in Kolkata megacity, Environ. Monit. Assess., **122**(1–3) (2006) 111–123.
- [33] A.K. Gupta, S. Nag, U.K. Mukhopadhyay, Characterisation of PM10, PM2.5 and benzene soluble organic fraction of particulate matter in an urban area of Kolkata, India, Environ. Monit. Assess., 115(1–3) (2006) 205–222.
- [34] P. Khare, B.P. Baruah, P.G. Rao, Water-soluble organic compounds (WSOCs) in PM_{2.5} and PM₁₀ at a subtropical site of India, Tellus B, **63**(5) (2011) 990–1000.
- [35] P. Murugavel, D.M. Chate, Volatile properties of atmospheric aerosols during nucleation events at Pune, India, J. Earth Syst. Sci., **120**(3) (2011) 347–357.

- [36] G. Balakrishnaiah, K.R. Kumar, B.S.K. Reddy, K.R. Gopal, R.R. Reddy, L.S. S. Reddy, K. Narasimhulu, Y. N. Ahammed, C. Balanarayana, K.K. Moorthy, S.S. Babu, Characterization of PM, PM10, PM2.5 mass concentrations at a tropical semi-arid station in Anantpur, India. Indian J. Radio Space Phys., 40 (2011) 95–104.
- [37] S.K. Sharma, T.K. Mandal, M. Saxena, Rashmi, Rohtash, A. Sharma, R. Gautam, Source apportionment of PM10 by using positive matrix factorization at an urban site of Delhi, India. Urban Climate, (2013) doi:10.1016/j.uclim.2013.11.002.
- [38] K. Sharma, R. Singh, S.C. Barman, D. Mishra, R. Kumar, M.P.S. Negi, S.K. Mandal, G.C. Kisku, A.H. Khan, M.M. Kidwai, S.K. Bhargava, Comparison of trace metals concentration in PM10 of different locations of Lucknow City, India, Bull. Environ. Contam. Toxicol., 77(3) (2006) 419–426.
- [39] A.K. Sudheer, M.M. Sarin, Carbonaceous aerosols in MABL of Bay of Bengal: Influence of continental outflow, Atmos. Environ., 42(18) (2008) 4089–4100.