



Particulate Matter Concentrations in the Southern Tip of India: Temporal Variation, Meteorological Influences, and Source Identification

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Abstract

Objective This study investigates the temporal variability of particulate matter (PM₁₀ and PM_{2.5}) concentrations at an inland tropical coastal site in Thiruvananthapuram (8.5°N, 76.9°E), which is in the province of Kerala in the southern part of India.

Method Continuous measurements of PM₁₀ and PM_{2.5} were carried out with high resolution datasets from March 2014 to February 2016. Variations of PM₁₀ and PM_{2.5} concentrations were statistically analyzed. The conditional bivariate probability function analysis, coherence wavelet transform and HYSPLIT back trajectory model are used for this study.

Results The annual mean mass concentrations of PM₁₀ and PM_{2.5} were 44.5 ± 14.6 and $27.7 \pm 10 \mu\text{g m}^{-3}$, respectively, for March 2014 to February 2015. Similarly, in March 2015 to February 2016, the concentrations were 40.3 ± 11.9 and $24.4 \pm 7.8 \mu\text{g m}^{-3}$, respectively, for PM₁₀ and PM_{2.5}. Both concentrations exhibited higher values during winter and lower values during monsoon. The coarse particle concentration (PM₁₀–PM_{2.5}) during the study period was $17.38 \pm 3.64 \mu\text{g m}^{-3}$. The mean PM_{2.5}/PM₁₀ ratio was found to be $0.56 \pm 0.13 \mu\text{g m}^{-3}$, which varied from 0.05 to 0.99 in March 2014–February 2015, and in March 2015–February 2016 it varied from 0.03 to 0.99 with a mean of $0.54 \pm 0.08 \mu\text{g m}^{-3}$. Diurnal analysis revealed that the concentrations were higher during morning (~08:00 IST) and evening (~20:00 IST) and low during 12:00 IST–17:00 IST. The anthropogenic activities, the diurnal variation of

boundary-layer height and the effects of meteorological parameters are the major elements for these variations. The effects of meteorological parameters on particulate matter were studied, and a negative correlation was observed with wind speed, rainfall, and relative humidity. The relationship between particulate matter and rainfall was investigated by applying coherence wavelet transform. The bivariate analysis showed that high PM_{2.5} concentrations were associated with low wind speeds indicating the presence of local pollutants. The highest concentrations were found for PM₁₀ with moderate to strong winds, which represent the presence of local emissions, as well as long-range transport. The conditional bivariate probability function analysis was performed to identify the probability of source contributions to PM concentrations at different wind speed and direction. The results indicated that the highest probability to get high concentrations of PM from south-southwest direction with wind speed $<5 \text{ ms}^{-1}$. HYSPLIT model back-trajectories of high PM₁₀ episodes show two dominant streamlines, one originating from the Bay of Bengal region and another from Middle East Asia.

Keywords PM₁₀ · PM_{2.5} · Meteorological parameters · Diurnal variation · Source identification

1 Introduction

The urban atmosphere, in particular, is very often affected by pollution from various anthropogenic sources and negatively affects the health of inhabitants (Yu et al. 2011). One of the most frequently encountered pollutants in urban areas is particulate matter (PM), which is introduced into the atmosphere from a variety of anthropogenic and natural sources. Environmental regulators have chosen to divide airborne

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particles into different size fractions as measured by their aerodynamic diameter. Airborne particles of aerodynamic diameters less than 10 μm (PM₁₀) and less than 2.5 μm (PM_{2.5}) adversely affect human health (Hall 1966) because the finer aerosol particles can penetrate deeper into the respiratory system. Hence, an understanding of the finer aerosol concentration in the atmosphere is important. Even though PM₁₀ is of major concern in worldwide environmental issues, such as visibility degradation and air quality assessment (Engler et al. 2012), PM_{2.5} is mostly responsible for health hazards (Jin et al. 2006). Fine (PM_{2.5}) particles have longer residence times in the atmosphere and are composed of higher fractions of organic compounds compared to PM₁₀ (Jaenicke 1984). The lifetime of atmospheric aerosol particles depends on their properties (size, chemical composition, etc.) and altitude range.

In the atmospheric boundary layer (lower troposphere), the residence time of aerosol particles is usually less than a week, often on the order of a day, depending on aerosol properties and meteorological conditions. Typically, in the free troposphere the particle lifetime is 3–10 days and the particles are easily transported a long distance. Therefore, large variability in the particle concentration reflects the geographical distribution of sources and sinks. Particles in the PM₁₀ category have a residence time of minutes to hours with travel distance of <1 to 10s of km. The residence time varies from days to weeks for PM_{2.5}, and particulates of this size can probably travel 100–1000s of km (Wilson and Suh 1997). The major anthropogenic sectors contributing to the coarser fraction (PM₁₀) of aerosols are traffic, industry, agriculture and forestry, households, construction, quarrying and mining, cement plants and ceramic industries, and fossil fuel power plants. On the other hand, the major natural sources include sea spray, soil re-suspension, volcanic eruption, biological particles, and debris. PM_{2.5} in the atmosphere consists of primary and secondary pollutants including volatile, non-volatile, and semi-volatile components, which originate from various sources (Eatough et al. 2003). According to the World Health Organization (WHO), particulate matter concentrations exceeding beyond permissible limits (50 $\mu\text{g m}^{-3}$ of PM₁₀ and 10 $\mu\text{g m}^{-3}$ of PM_{2.5} on annual mean basis) can cause respiratory and cardiovascular disease and can have significant impact on mortality and morbidity (Schwartz et al. 1996). WHO states that an increase in total PM by 10 $\mu\text{g m}^{-3}$ per year results 6% increase in mortality. A short-term increase in levels of PM₁₀, PM_{2.5}, and PM_{1.0} concentration for several days can cause coughing, respiratory problems, and mortality (WHO 2000, 2003). Hence, the assessment of PM_{2.5} and PM₁₀ concentrations, as well as their yearly variation and trend are significantly important from environmental and health perspectives. The meteorological conditions influence the trends and variability of all aerosol species in the atmosphere (Elminir 2005). In addition, emission

sources and their transportation to the sampling location are also important factors. Therefore, to assess the trends and variability of particulate matter, the emission sources at the sampling location and the meteorological conditions need to be studied.

The concentration of aerosols in the atmosphere, as well as their size distribution is affected by various source processes, removal processes like washout due to rain (Flossmann et al. 1985; Byrne and Jennings 1993), by aging, and by particle growth due to humidity changes (Hanel 1976; Exton et al. 1985). Owing to the effect of gravity, coarse particles are rapidly removed from the air by sedimentation; also, fine particles are rapidly transformed into coarser particles by coagulation processes. Removing particles from the atmosphere to the surface of the earth is a combination of processes including five steps: (1) sedimentation, (2) diffusion, (3) turbulence, (4) wash-out, and (5) wet deposition (Beckett et al. 1998).

In recent years, rapid urbanization and industrialization have resulted in an increased vehicle emissions, as well as industrial emissions in the city of Thiruvananthapuram (8.5°N, 76.9°E; southwest coast of Kerala, India) and its surrounding areas. As per the Central Pollution Control Board (CPCB, Government of India) reports, the urban areas in Thiruvananthapuram are now falling under moderate to high pollution levels. Over the city of Thiruvananthapuram, limited studies have been conducted to characterize the particulate matter concentrations on a short time scale. Pillai et al. (2002), assessed PM₁₀ and PM_{2.5} concentrations over a tropical coastal station, Thumba, Thiruvananthapuram. Even though their study reported the temporal variation of particulate matter concentrations, it did not investigate the relationship to synoptic meteorology and source identification. Ragi et al. (2013) assessed the particulate matter concentrations (PM₁₀, PM_{2.5}, and PM_{1.0}) during May–June 2012 at 12 selected areas in Thiruvananthapuram city using Grimm Aerosol Spectrometer. Until recently, no continuous high temporal resolution air quality observations are available for this location, and hence PM₁₀ and PM_{2.5} assessments could not be made for long periods. However, under the Modelling Atmospheric Pollution and Networking (MAPAN) project of the Indian Institute of Tropical Meteorology, a continuous ambient air quality monitoring station was established in Thiruvananthapuram, which made possible uninterrupted measurements of PM₁₀ and PM_{2.5} for a 2-year period (March 2014–February 2016). This paper reports the analysis of PM₁₀ and PM_{2.5} mass concentrations on a monthly scale, seasonal scale, and their diurnal variability over Thiruvananthapuram during March 2014–February 2016. Furthermore, it gives an explanation about the air quality of the study area in terms of fine (PM_{2.5}) and coarse particles, which is the subset of PM₁₀ that is larger than 2.5 μm and smaller than 10 μm (PM_{10–2.5}) to PM₁₀ particles. The study was

extended to examine the relationships between PM and prevailing meteorological conditions over the study area. An attempt was made for the source emission identification of the particles via bivariate plots and conditional bivariate probability function (CBPF) methods.

2 Observation site and measurement techniques

Thiruvananthapuram, the capital city of Kerala state (8.5°N , 76.9°E) is situated in the southwestern tip of mainland India. It is geographically located on the southwest coast of India, bounded by the Arabian Sea in the west and Western Ghats in the east. The study area is a semi-urban site at Akkulam, which is about 5 km away from the city center of Thiruvananthapuram and 3 km away from the coast of Arabian Sea. Vehicle emission and construction activities in connection with different offices and residence buildings are major sources of various particulate and gaseous pollutants near to the observation site. In addition, windblown dust, unpaved roads, dust from open fields, etc., contributes PM₁₀ and PM_{2.5} concentrations at the sampling site.

The air pollution monitoring facility was installed at the National Centre for Earth Science Studies (NCESS), Akkulam (Fig. 1) in February 2014, and data recording was started in March 2014 onwards. Therefore, the data collection and analyses explained in this paper was from March 2014 to February 2016. The entire study period was divided into 2 years: year 1 (Mar 2014–Feb 2015) and year 2 (Mar 2015–Feb 2016). During this study period, the mass concentrations

of both PM_{2.5} and PM₁₀ were measured continuously using a Beta Attenuation Monitor (BAM-1020; Met One Instruments, USA). The instrument was placed on the rooftop of a building (~ 15 m above the ground level) on the campus of NCESS. The BAM-1020 automatically measures and records airborne particulate concentration levels (in milligrams or micrograms per cubic meter) using the principle of beta ray attenuation. The mass concentration was observed at 5-min intervals and then stored in a data logger as 1-h average, and further analyses were done by taking these data on a daily basis. In addition, meteorological parameters such as temperature, relative humidity, solar radiation, rainfall, and wind speed and direction were measured at 998 hPa using an automatic weather station at the same site. To study the diurnal behavior of particulate matter with boundary layer height, hourly planetary boundary layer (PBL) height was used from the Modern Era Retrospective Analysis for Research and Applications (MERRA) reanalysis available on NASA's website, for the 2-year period (<http://gmao.gsfc.nasa.gov/research/merra/intro.php>). HYSPLIT model back-trajectories were used using GDAS meteorological data for the trajectory analysis (<http://ready.arl.noaa.gov/HYSPLIT.php>).

3 General meteorological conditions over the study area

Since the paper explains the temporal variation of PM mass concentrations with the meteorological parameters, a brief description of the prevailing meteorological conditions

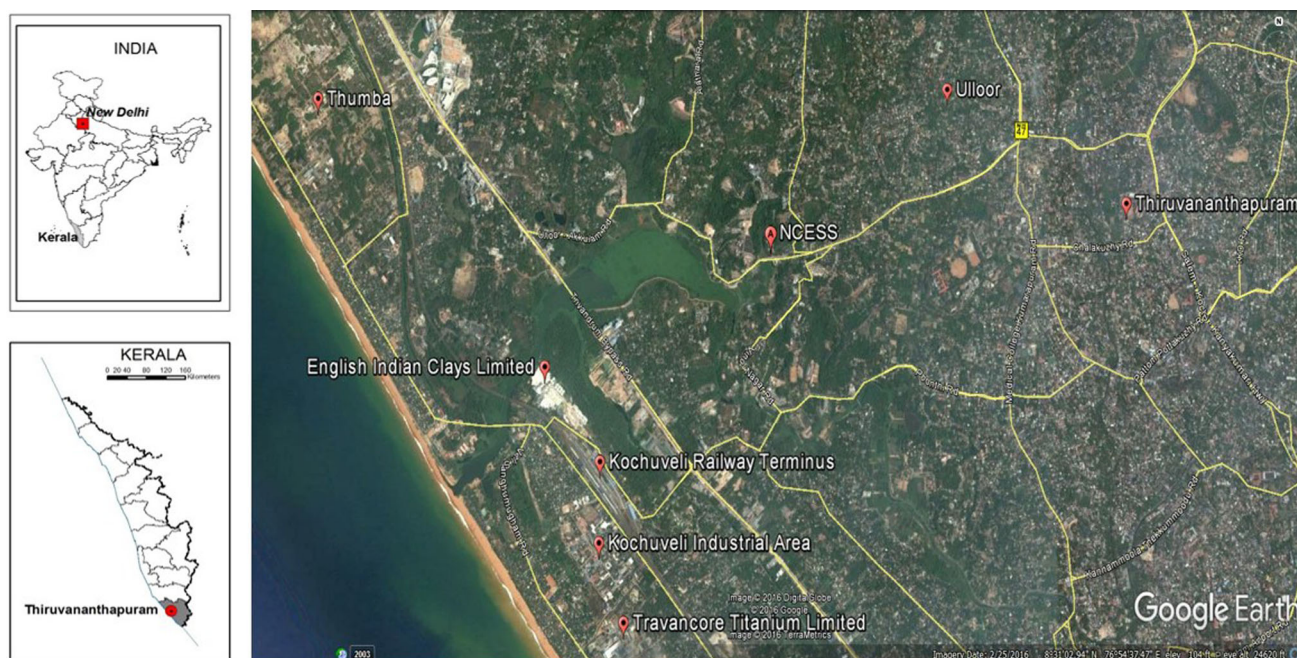


Fig. 1 Location of observational site at Thiruvananthapuram in Kerala, India

over the study area is important. The study year is divided into four major seasons, pre-monsoon (March–May), monsoon (June–September), post-monsoon (October–December), and winter (January–February), with respect to the seasons suggested by the Indian Meteorological Department (IMD), (<http://www.imd.gov.in/>). Figure 2 shows the daily and monthly average values of ambient temperature (AT), relative humidity (RH), solar radiation (SR), and wind speed (WS), while Fig. 3 presents the monthly mean rainfall (RF) during the study period. Table 1 shows the seasonal-mean concentration of PM₁₀, PM_{2.5}, and meteorological parameters. The diurnal variability of these meteorological parameters is depicted in Fig. 4.

As a tropical coastal area, the most important seasonal phenomena (from a meteorological perspective) are the changes in the wind field and rainfall associated with the Indian summer monsoon. Most of the rainfall occurs during

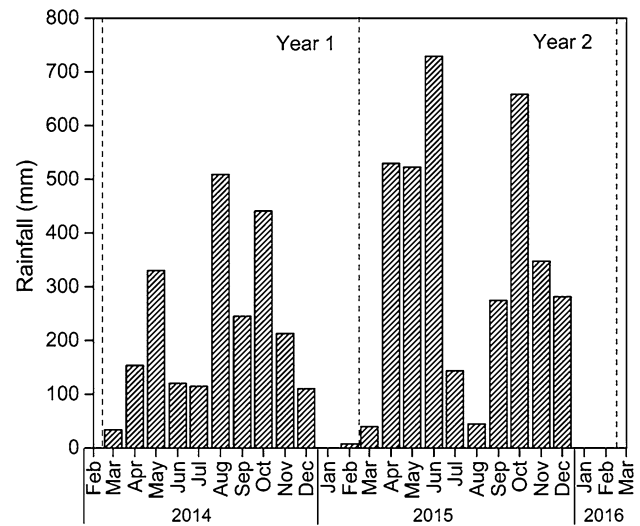


Fig. 3 Monthly total rain fall during the study period from March 2014 to February 2016

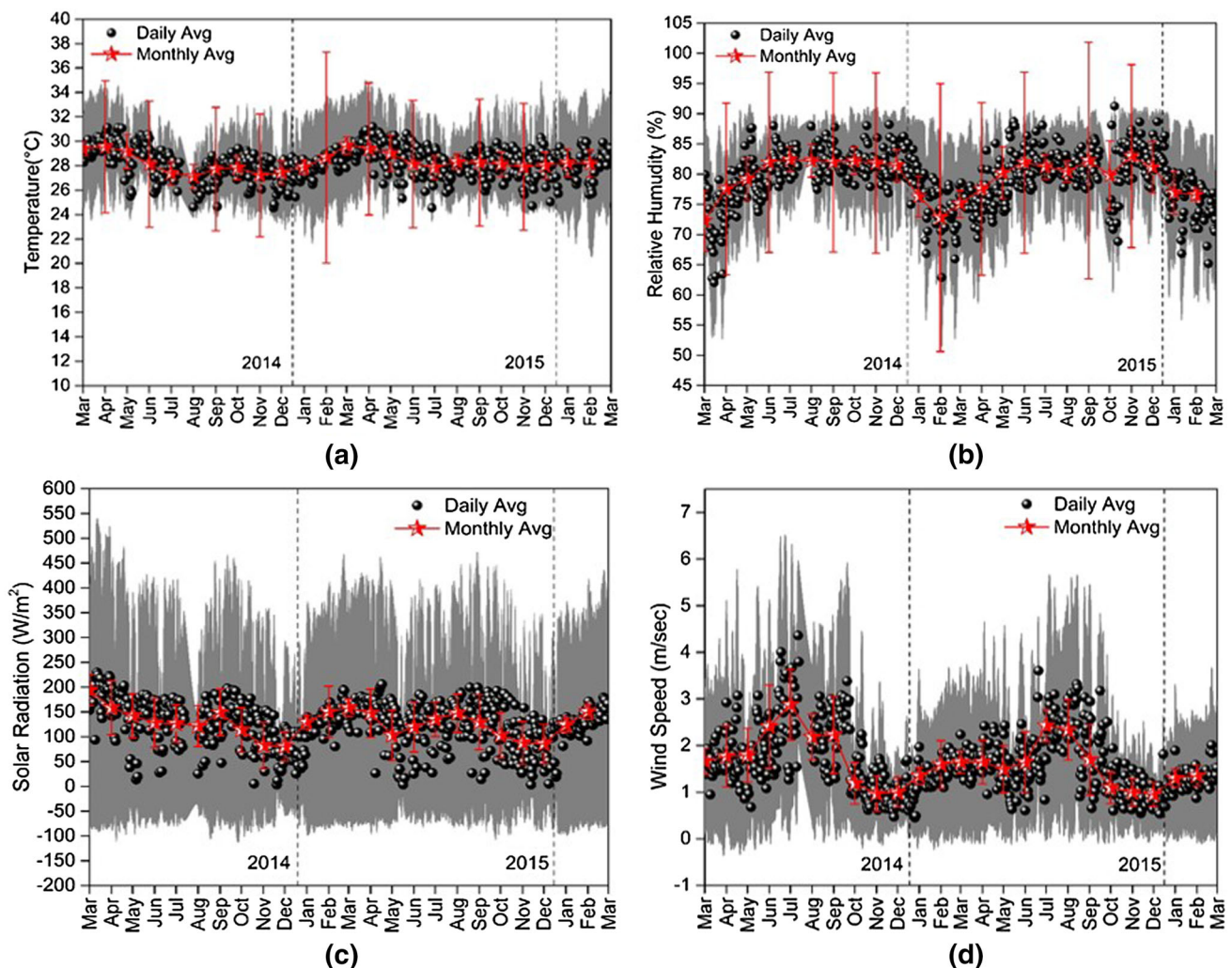
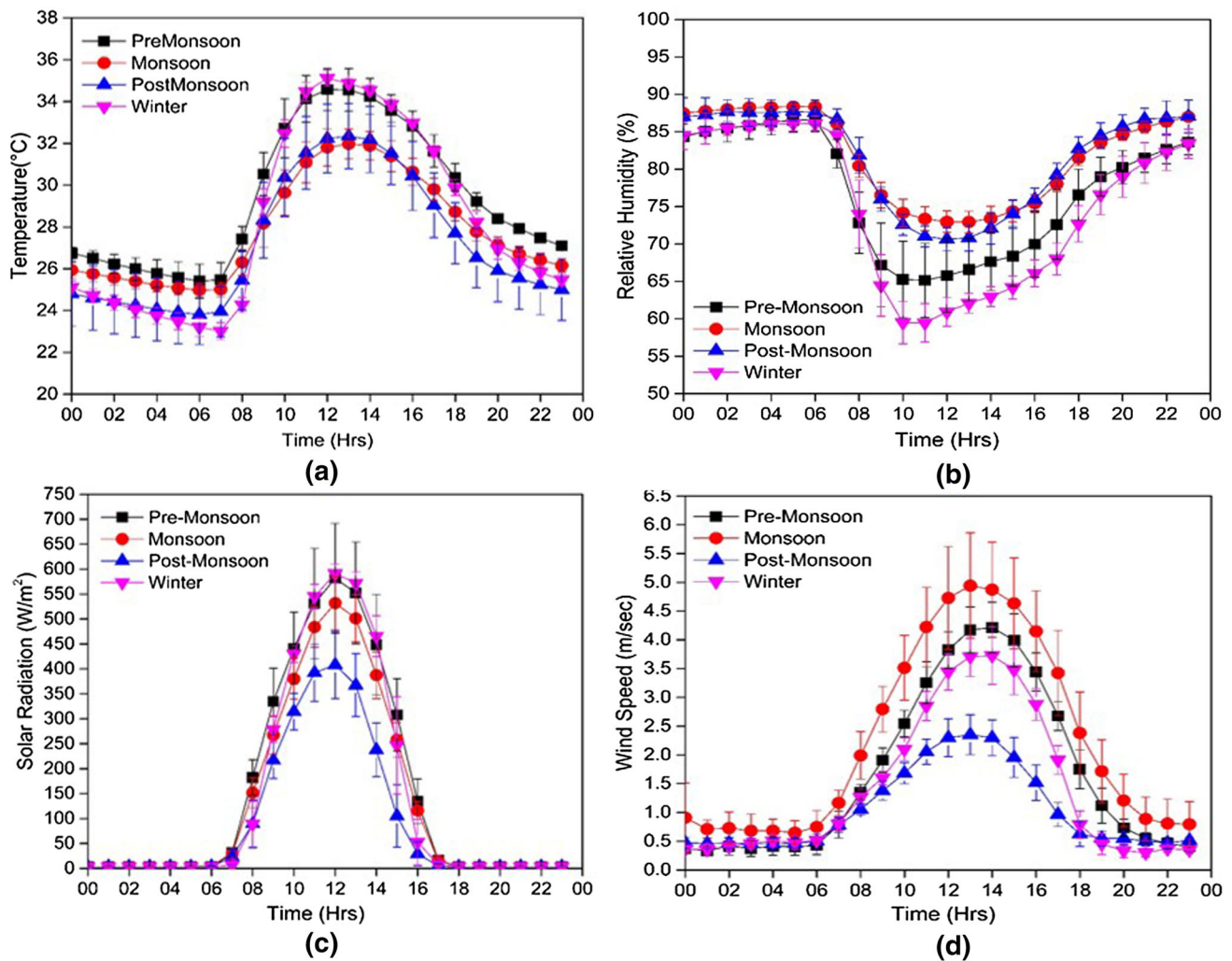


Fig. 2 Daily and monthly averages of **a** temperature, **b** relative humidity, **c** solar radiation, and **d** wind speed at the observation site from March 2014 to February 2016. The shaded area corresponds to the standard deviation from the daily means

Table 1 Seasonal statistics of mass concentrations of PM and meteorological parameters

Seasons	PM ₁₀ \pm SD ($\mu\text{g m}^{-3}$)	PM _{2.5} \pm SD ($\mu\text{g m}^{-3}$)	AT \pm SD ($^{\circ}\text{C}$)	RH \pm SD (%)	SR \pm SD (W m^{-2})	WS \pm SD (m s^{-1})
Pre-monsoon	45.6 \pm 12.8	27.4 \pm 7.6	29.3 \pm 2.5	76.9 \pm 7.3	150.6 \pm 41.2	1.7 \pm 0.5
Monsoon	26.1 \pm 8.3	11.6 \pm 3.9	27.9 \pm 3.1	81.7 \pm 9.1	132.1 \pm 44.3	2.2 \pm 0.7
Post-monsoon	42.7 \pm 17.6	32.4 \pm 14.7	27.2 \pm 2.8	81.6 \pm 7.5	92.2 \pm 41.7	1.1 \pm 0.4
Winter	70.1 \pm 17.3	49.1 \pm 12.8	28.2 \pm 2.8	75.7 \pm 7.5	138.7 \pm 23.8	1.4 \pm 0.3

**Fig. 4** Season wise average diurnal variations of **a** ambient temperature, **b** relative humidity, **c** solar radiation, **d** wind speed during March 2014–February 2016

monsoon and post-monsoon seasons. The annual rainfall in the study area was 2277 and 3570 mm, respectively, for years 1 and 2. The rainfall during monsoon period was 988 mm (in Jun–Sep 2014), about 43.4% of the total annual rainfall and 1191 mm (in Jun–Sep 2015), which was 33.3% of annual rainfall, whereas 654 mm (in Oct–Dec 2014) and 1005 mm (in Oct–Dec 2015) for the post-monsoon seasons, which formed 33.5 and 36.1% of annual rainfall. The ambient temperature varied between 21.2 and

38.2 $^{\circ}\text{C}$ with an annual mean of 28.3 ± 2.8 $^{\circ}\text{C}$. The monthly mean ambient temperature varied from 27.4 ± 0.9 $^{\circ}\text{C}$ in December 2014 to 29.7 ± 0.7 $^{\circ}\text{C}$ in March 2015 (Fig. 2a). The relative humidity at this location varied from 40.2 to 99% with an annual mean of $79.5 \pm 7.9\%$ and it has a significant seasonal variation on both diurnal and annual scale. The magnitude of wind speed varied from 0.1 to 9.4 ms^{-1} with an annual mean of 1.6 ± 0.47 ms^{-1} . On a seasonal basis the wind speed

increases from pre-monsoon to monsoon (Table 1), with a maximum of $2.8 \pm 0.8 \text{ ms}^{-1}$ in July 2014 and $2.4 \pm 0.4 \text{ ms}^{-1}$ in July 2015 (Fig. 2d). The wind speed exhibited a clear diurnal pattern, with a maximum in the late morning to afternoon (10–16 LST) hours in all seasons. It attained a high value ($4.9 \pm 0.92 \text{ ms}^{-1}$) in the monsoon season (Fig. 4d).

4 Results and discussions

4.1 Temporal variability of PM10 and PM2.5

The monthly mean mass concentrations (averages of hourly data) of PM10 and PM2.5 over the study area are plotted in stack as a function of months in Fig. 5a, b. The mass concentration of PM10 was found to be at a maximum in the months of December 2014 ($76.88 \pm 25.8 \mu\text{g m}^{-3}$) and February 2016 ($73 \pm 14.5 \mu\text{g m}^{-3}$). A minimum was observed in August 2014 ($22.5 \pm 5.9 \mu\text{g m}^{-3}$) and September 2015 ($22.4 \pm 8.0 \mu\text{g m}^{-3}$). The highest PM2.5 concentrations occurred in December 2014 ($56.9 \pm 23.9 \mu\text{g m}^{-3}$) and January 2016 ($52.1 \pm 12.4 \mu\text{g m}^{-3}$) and the lowest values in August 2014 ($9.7 \pm 3.8 \mu\text{g m}^{-3}$) and June 2015 ($10.2 \pm 3.6 \mu\text{g m}^{-3}$), respectively for year 1 and year 2.

To assess the influence of meteorological variables on both the particulate matter concentrations, correlation analyses were carried out. A positive correlation was found with atmospheric temperature (PM10; $r = 0.39$ and PM2.5; $r = 0.16$) and with solar radiation (PM10; $r = 0.33$ and PM2.5; $r = 0.11$) at 0.05 significance level. The March and April is the warmest months with average temperature of around 29°C and solar radiation in the range $160\text{--}195 \text{ W m}^{-2}$. This would have resulted in the occurrence of photochemical reactions, which supports the conversion of NO_x into nitrates and SO_2 into sulphates. These pollutants combine with already persisting suspended particulates and enhance the buildup of concentration levels (Sivaramasundaram and Muthusubramanian 2010). The rainfall was much less in the month of March in both years. In March, moderately strong southwest winds contribute to the high concentration of particulate matter, mostly PM10. The wet (rainfall) removal processes of aerosols are more effective during monsoon season. Thus, the aerosol concentration reaches a minimum value. Particularly in the month of August, rainfall was 509 mm in 2014, which decreased the level of particulate matter (PM10; $r = -0.21$ and PM2.5; $r = -0.13$) in the atmosphere. However, in the next year (2015), there was relatively less rainfall of 44.5 mm. Resultantly the PM concentrations increased by 24.2% in August 2015 due to the minimization of natural washout effect.

The less rainfall and low relative humidity conditions during winter season favors the generation of continental aerosols through naturally and from urban activities (Pillai et al. 2002). In the months of January and February, continental air mass is advected by the northeasterly winds over the study region. These air masses comprise coarse aerosols (PM10) that arise from mechanical processes associated with urban activities (mostly transportation activities around the observation site). The scanty rainfall and absence of clouds during this period renders poor scavenging processes and a high level of aerosol loading (Balakrishnaiah et al. 2011). Therefore, slightly higher PM10 mass concentration was found as compared to PM 2.5. During this period the coarser particles (PM10 and above) may accumulate over the observation site by windblown dust from southeast direction. A significant positive correlation ($r = 0.32$) was obtained with wind speed and PM10 in February 2016, and with PM2.5 a negative relationship was obtained ($r = -0.29$). The monthly variations obtained in the present study are comparable to the previous study carried out at Thumba, a coastal location in Thiruvananthapuram (Pillai et al. 2002), which is around 3.5 km away from our observation site in northwest direction.

The annual mean of PM10 and PM2.5 mass concentrations were $44.5 \pm 14.6 \mu\text{g m}^{-3}$ (varying from 1 to $316 \mu\text{g m}^{-3}$) and $27.7 \pm 10 \mu\text{g m}^{-3}$ (varying from 1 to $284 \mu\text{g m}^{-3}$) in year 1 and in year 2, mean PM10 was $40.3 \pm 11.9 \mu\text{g m}^{-3}$ (varying from 2 to $219 \mu\text{g m}^{-3}$) and mean PM2.5 was $24.4 \pm 7.8 \mu\text{g m}^{-3}$ (varying from 1 to $155 \mu\text{g m}^{-3}$), respectively. These concentrations are considerably below the annual average standards stipulated by the National Ambient Air Quality Standards (NAAQS: <http://cpcb.nic.in/National-Ambient-Air-Quality-Standard.s.php>) (PM2.5 = $40 \mu\text{g m}^{-3}$ and PM10 = $60 \mu\text{g m}^{-3}$). During the study periods, approximately 96% of PM10 and 89% of PM2.5 concentrations were below the NAAQS levels. Frequency distribution of PM2.5 and PM10 particles are represented in Fig. 5c. A positively skewed distribution with highest concentration values was in the range of $10\text{--}20 \mu\text{g m}^{-3}$ for both types of particulate matter, and these results are similar to the previous studies conducted by Pillai and Moorthy (2001) in the southwest coast of the Indian peninsula. PM10 and PM2.5 mass concentrations showed considerable daily variability (PM10; $8\text{--}153 \mu\text{g m}^{-3}$ and PM2.5; $3\text{--}135 \mu\text{g m}^{-3}$). The day to day variability of PM concentrations are also plotted in Fig. 5d. The inherent annual variations shows low PM concentration during monsoon and high values in winter followed by summer and post-monsoon seasons.

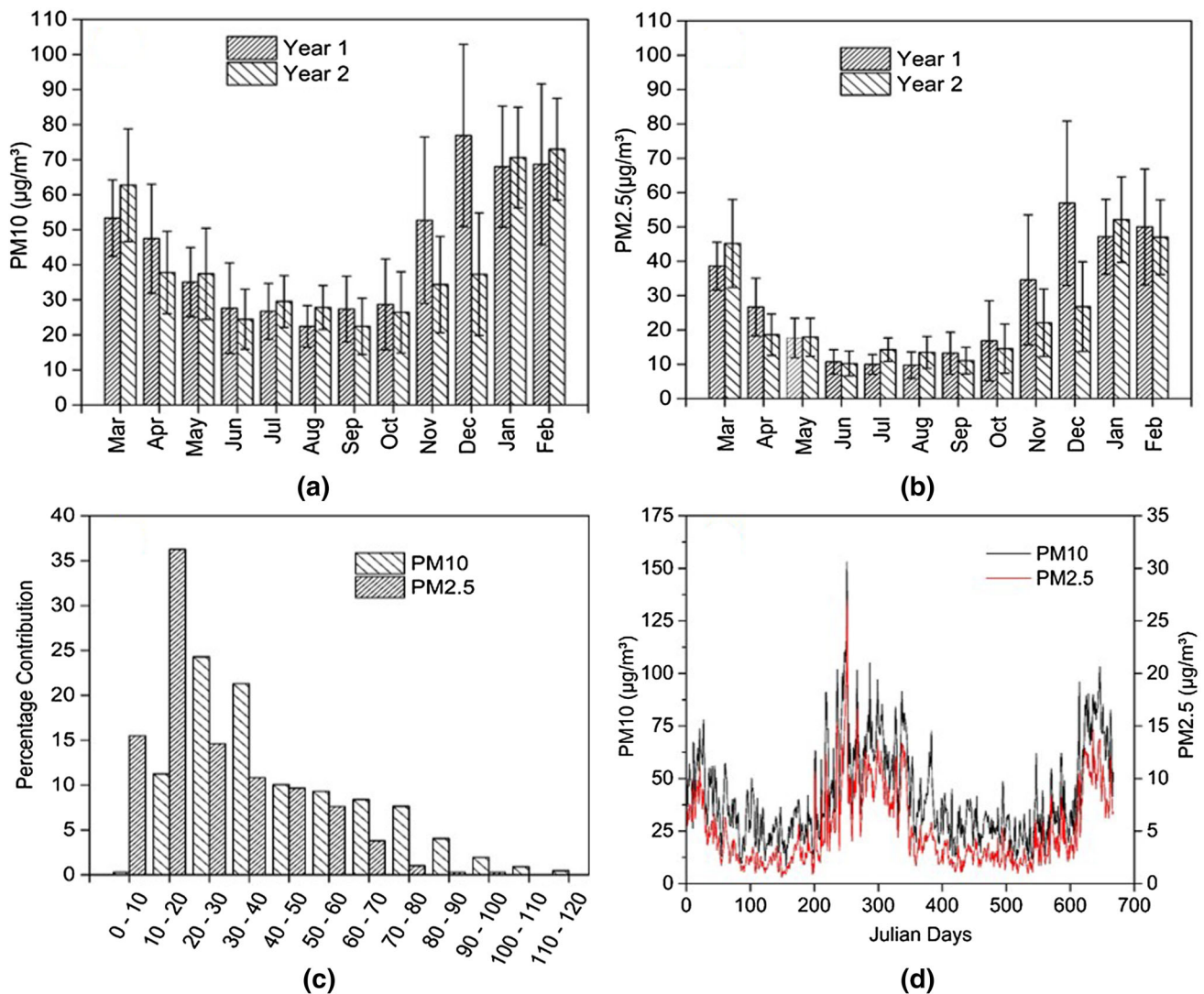


Fig. 5 Monthly mean distribution of mass concentrations of **a** PM10 and **b** PM2.5 during 2014 March–2016 February. **c** Frequency of occurrence of PM2.5 and PM10 concentration during March 2014–February 2016. **d** Daily mean variation of PM10 and PM2.5

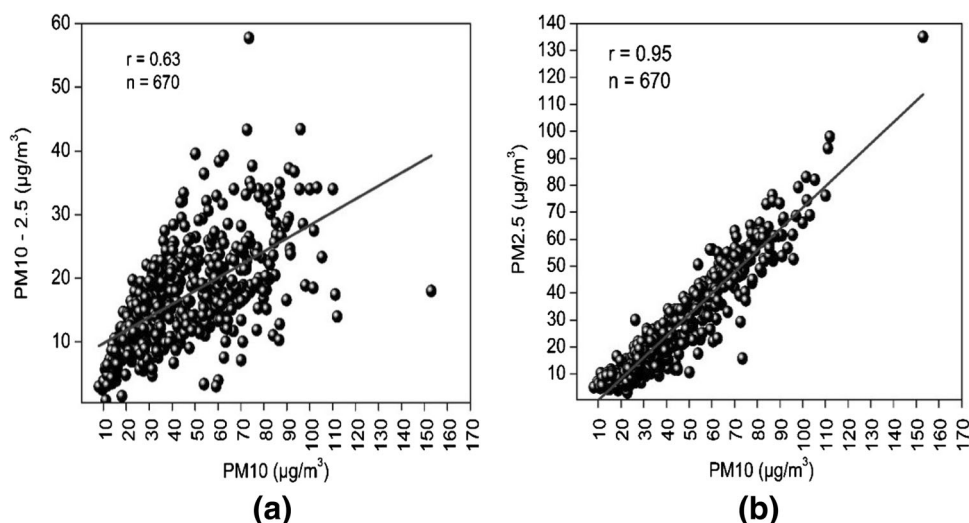
4.2 Relationships between PM2.5, PM10, and PM10–2.5

The mean coarse-mode fraction (PM10–2.5) was found to be $17.38 \pm 3.64 \mu\text{g m}^{-3}$ (varying from 12.47 to $27.7 \mu\text{g m}^{-3}$), which corresponds to 40% of the PM10 concentrations. The relationship between PM2.5 and PM10 provides understanding about the source characterization of PM (Kothai et al. 2009). The concentration of coarse particles was increased in year 2 as compared to year 1 and the variation was high in February (38% increase). On seasonal scale the coarse-mode particles show an increase from monsoon ($15.10 \mu\text{g m}^{-3}$), post-monsoon ($15.74 \mu\text{g m}^{-3}$), pre-monsoon ($19.01 \mu\text{g m}^{-3}$), and winter ($21.33 \mu\text{g m}^{-3}$) season. The ratios between PM2.5 and PM10 were calculated on a monthly basis and a large variability was found, ranging from 0.05 (June 2014) to

0.99 (February 2015) with a mean value of $0.56 \pm 0.13 \mu\text{g m}^{-3}$ in year 1. In year 2, it ranges from 0.03 (August 2015) to 0.99 (January 2016) with a mean of $0.54 \pm 0.08 \mu\text{g m}^{-3}$ over the station. However, in year 1 the ratios between PM10–2.5 and PM10 were $0.44 \pm 0.13 \mu\text{g m}^{-3}$ in the range of 0.02 (October 2014) to 1 (May 2014) and in year 2 it was in the range of 0.01–0.98 in December with a mean value of $0.43 \pm 0.11 \mu\text{g m}^{-3}$. The PM2.5/PM10–2.5 ratio has a mean value of $5.19 \pm 8.1 \mu\text{g m}^{-3}$ (February 2015) in year 1 and $5.2 \pm 7.6 \mu\text{g m}^{-3}$ (January 2016).

A scatter plot showing the contribution of fine and coarse particles in PM10 is presented in Fig. 6a, b. Season-wise PM2.5/PM10 ratios were also analyzed and found to be high during winter (0.70) followed by post-monsoon (0.58), pre-monsoon (0.56), and monsoon (0.43), which clearly indicated that the maximum contribution of fine

Fig. 6 Scatter plots showing the relationship between
a PM₁₀–2.5 with PM₁₀ and
b PM_{2.5} with PM₁₀



particles (PM_{2.5}) was during winter. The ratio of 0.43 (PM_{2.5}/PM₁₀) evolved during monsoon season is due to the large fraction of coarse particles present in mass concentration of PM₁₀. The results obtained in the seasonal analysis of PM_{2.5}/PM₁₀ ratio was similar to that of previous studies conducted at coastal station in Thiruvananthapuram (Pillai et al. 2002).

In monsoon season, the steady onshore direction of wind favors the generation and advection of coarser sea spray aerosols, which would be added to particles already resident in the atmosphere and cause a change in the size distribution (Pillai and Moorthy 2001). However, the fine particles are not replenished as the continental sources are weak and the marine source is less efficient in generating fine particles. Thus, the PM_{2.5} concentration depleted and this may be the reason for obtaining a lower PM_{2.5}/PM₁₀ ratio during this season.

During winter, the stable atmospheric conditions may play a crucial role in the trapping of fine particles near the surface, and thus fine particles will get a longer residence time that results in a higher PM_{2.5} share to PM in winter season. Season-wise PM_{2.5}/PM₁₀–2.5 ratios were 2.61 (pre-monsoon), 1.24 (monsoon), 2.81 (post-monsoon), 4.68 (winter). These ratios also indicate that contribution of fine particle is less in monsoon. The seasonal ratios between coarse mode (PM₁₀–2.5) and PM₁₀ were also calculated and found to be different from PM_{2.5}/PM₁₀ behavior. It was observed to be higher during monsoon (0.56) followed by pre-monsoon (0.43), post-monsoon (0.42), and winter (0.30). These results show that, in the observation site, a significant portion of PM₁₀ may comprise locally generated and wind-blown dust in the coarse fraction (PM₁₀–2.5) mode. On comparing the ratio (PM_{2.5}/PM₁₀) with other studies conducted in India, the result obtained in

the present study was similar to those reported from other studies in urban areas (Tiwari et al. 2014; Sharma and Maloo 2005; Satsangi et al. 2011).

4.3 Diurnal variations of PM

Studies conducted on different parts of India (Kunhikrishnan et al. 1993; Tiwari et al. 2014; Dumka et al. 2015), has reported that the diurnal variability of PM₁₀ and PM_{2.5} mass concentrations are highly related to the combined effects of local emissions, meteorology, and boundary-layer dynamics. Figure 7 shows the diurnal variation of PM₁₀ and PM_{2.5} with PBL height in four seasons. On annual basis the observed morning peak values were PM₁₀ = $53.3 \pm 28.2 \mu\text{g m}^{-3}$, PM_{2.5} = $37.8 \pm 23.4 \mu\text{g m}^{-3}$, and the evening peaks were PM₁₀ = $47.8 \pm 21.7 \mu\text{g m}^{-3}$, PM_{2.5} = $32.8 \pm 20.6 \mu\text{g m}^{-3}$, respectively. Diurnal mean lowest and highest of PM₁₀ and PM_{2.5} varied from $19 \mu\text{g m}^{-3}$ (13:00 IST) to $112 \mu\text{g m}^{-3}$ (20:00 IST) and $8 \mu\text{g m}^{-3}$ (14:00 IST) to $87 \mu\text{g m}^{-3}$ (22:00 IST).

The hourly data of PM₁₀ and PM_{2.5} peak concentrations were examined separately for daytime (06:00 IST–17:00 IST) and nighttime (18:00 IST–05:00 IST). The results revealed that the percentage of larger PM₁₀ concentrations were almost equal ($\sim 50.4\%$) in day and night and for PM_{2.5} the higher ($\sim 55\%$) concentrations were obtained in daytime. The source of PM₁₀ are evolved from re-suspension of dust particles due to traffic movement through the unpaved roads near the observation site. In addition, the low boundary layer height during the morning peak hours (07:00 IST–09:00 IST) not favors the dispersion processes of these suspended particulate matter (correlation details are discussed in Sect. 4.4). These

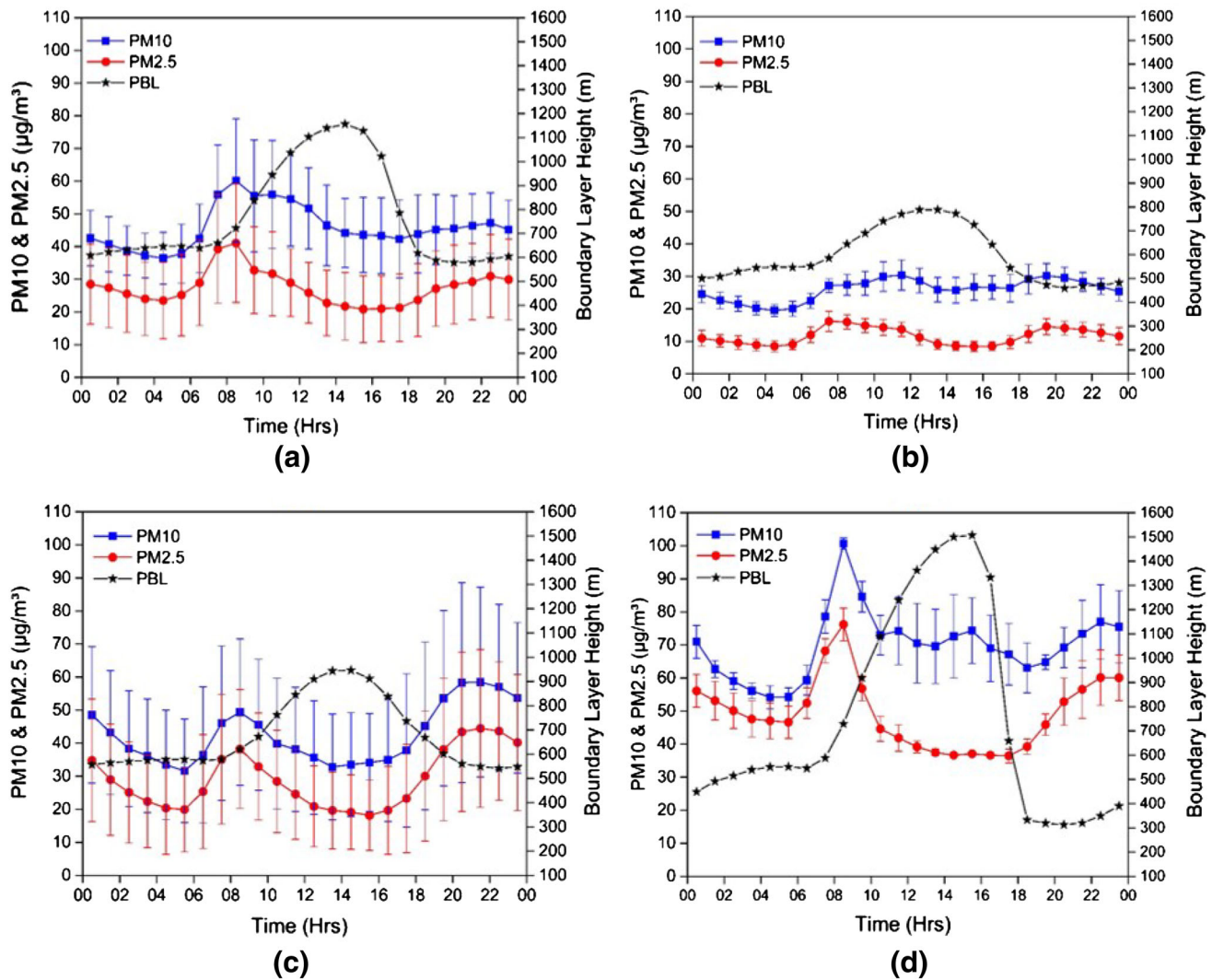


Fig. 7 Seasonal-mean diurnal variations of PM10 and PM2.5 concentrations during **a** pre-monsoon, **b** monsoon, **c** post-monsoon, and **d** winter seasons along with boundary layer height at Thiruvananthapuram

factors are responsible for the higher peak of PM2.5 in the daytime.

After the morning peak period, the mass concentration appeared to drop gradually between 10:00 IST and 12:00 IST and the concentrations remain more or less steady (12:00 IST–17:00 IST). Moreover, solar heating increases the boundary layer height to 600–800 m near the coast and much higher deeper in land (Kunhikrishnan et al. 1993). The solar radiation recorded at the observation site, during the hours of 10:00–17:00 peaks of about 520 W m^{-2} , that resulted an increased convective activity leading to an increase in the boundary layer height and this eventually increases the ventilation coefficient (defined as the product of boundary layer height and horizontal wind speed) leading to a much faster dispersion of fine aerosol particles (Saha and Despiau 2009).

In the evening, the solar forcing is cut-off and the boundary layer height decreases, thus at around 18:00 IST the concentration gradually increased. The development of nocturnal boundary layer height is an important aspect that favors the stagnation of pollutants near the surface. During night to morning hours (22:00 IST–6:00 IST) even though there were less traffic emissions, the concentrations of aerosols were stagnant due to the low ($0.5\text{--}0.8 \text{ ms}^{-1}$) wind speed. In early morning hours, relative humidity prevailed over the sampling site was higher (with 85–87% range). Higher relative humidity in the atmosphere could make the particulate to coagulate and to become heavier thereby helping in quick settling (Sivaramasundaram and Muthusubramanian 2010). Similar nocturnal behavior of particulate matter concentration was reported earlier over the tropical coastal site (Pillai et al. 2002) and urban areas (Latha and Badarinath 2005).

From the seasonal analysis, strong diurnal variation was observed for PM concentration in three seasons except in monsoon season (Fig. 7a–d). In the monsoon season, sharp gradient in the morning and evening peaks is not clearly visible. In the months of June to August, the episodes of frequent rain and cloudy sky conditions exhibit variations in meteorological parameters. The boundary layer height was less (800 m) comparatively in monsoon due to lower solar radiation and atmospheric temperature heating (Fig. 4a, c), which favored the trapping of pollutants near the surface (DeGaetano and Doherty 2004). Thus, the significant gradient in PM reduction is absent in the noon time. In post-monsoon and winter seasons, there is a gradual increase in PM values in the morning hours of 6–7 LST and it peaks around 8–9 LST. Also, in the afternoon (10–18 LST) the concentration were reduced and again peaks around 20–22 LST. In winter due to low boundary layer height and wind speed, dilution processes of aerosols are minimized. Resultantly the particulate matters emitted from anthropogenic activities are accumulated during the morning peak hours. Moreover, the atmospheric condition during winter season is almost stable that may not favor the vertical mixing of aerosols.

4.4 Influence of meteorological parameters

A correlation study was performed between mass concentrations of PM₁₀, PM_{2.5}, and meteorological parameters, such as temperature (AT), relative humidity (RH), wind speed (WS), rainfall (RF), to understand the impact of local meteorology on PM concentration over Thiruvananthapuram. The daily mean values of meteorological parameters were used to correlate with particulate matter (figures not shown).

The correlation analysis revealed that PM₁₀ and PM_{2.5} concentrations were negatively correlated with the wind speed (significant at 0.05 level). The correlation coefficients were -0.32 and -0.41 , respectively. The slightly high correlation value between PM_{2.5} and wind speed in comparison with PM₁₀ indicates the dominance of local sources. The correlation analysis was performed on a seasonal basis in which negative correlations were observed in winter (PM₁₀; -0.33 and PM_{2.5}; -0.28). However, in monsoon season a positive correlation (PM₁₀; $r = 0.35$) was observed, as the coarse particles are easily reloaded by the marine air masses. The stronger wind speeds during monsoon season are responsible for the long range transport of air pollutants, which favors the re-suspension of dust particles over the observational site and these pollutants are trapped due to low boundary layer height.

The daily mean values of boundary layer height varied from 490.3 to 1033.4 m (mean 685.9 m) for years 1 and 2, respectively. The PBL height determines the volume

through which surface-emitted pollutants are diluted and reflect the boundary layer turbulence (Stull 1988). PBL height was inversely correlated with fine particle during post-monsoon (-0.64) and winter (-0.54). This indicates that the accumulation of fine mode particles during post-monsoon is strongly influenced by the reduced PBL values. A negative correlation was found between PM₁₀ and PBL during post-monsoon season (-0.58). Diurnal average of PBL height also showed a negative correlation with PM_{2.5} in post-monsoon (-0.58).

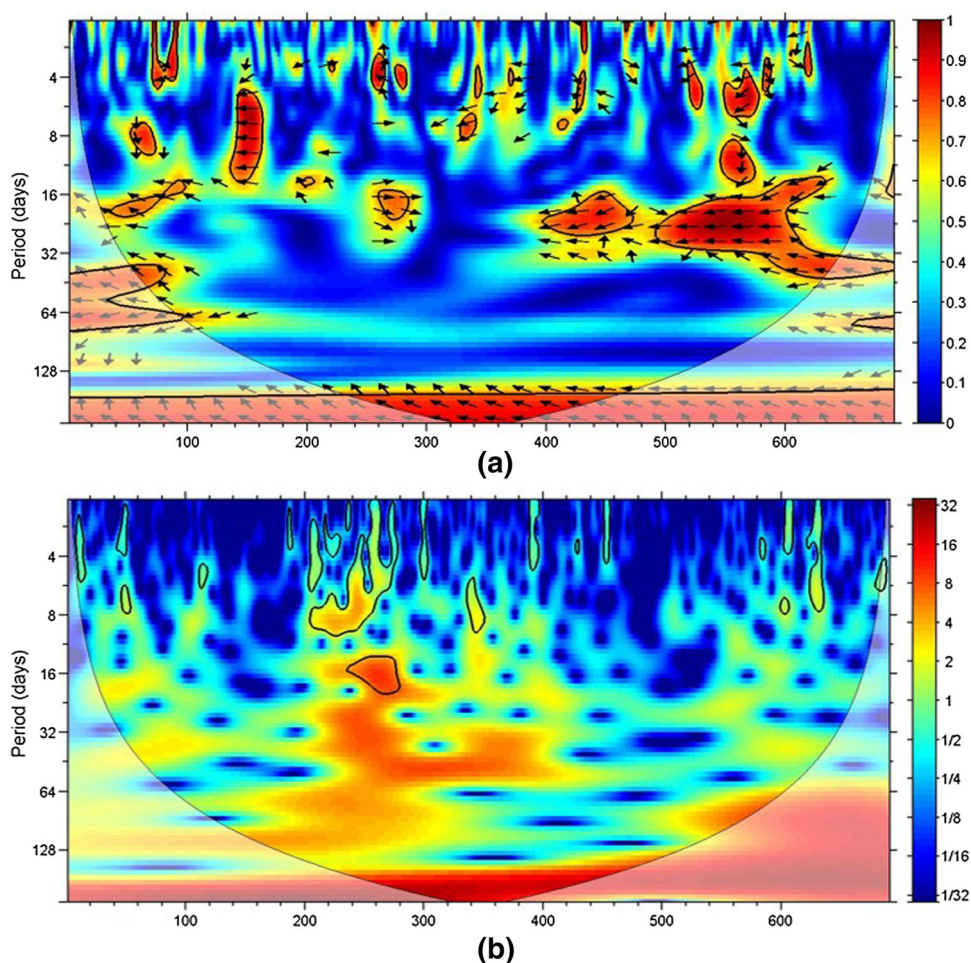
The relationship between PM mass concentrations and temperature was analyzed; no significant correlation was observed between PM_{2.5} and temperature, except post-monsoon ($r = -0.51$). In the case of PM₁₀, a positive correlation during pre-monsoon ($r = 0.48$) and monsoon ($r = 0.56$) and a negative correlation during post-monsoon (-0.51) season was observed. The particulate matter is subjected to a scrubbing process due to rain, and the possibility of reduction of particulate matter in the atmosphere is increased in monsoon season. But during non-rainy days of monsoon, the atmospheric temperature increases and the concentration of PM₁₀ is found to be higher due to the failure of the natural scrubbing process (Jayamurugan et al. 2013), thus producing a positive correlation. The photochemical reactions are enhanced at higher temperatures, resulting in the formation of secondary aerosol particles that combine with the already persisting particulates in the atmosphere. This can cause an increase in the concentration of particulate matter during warmer days/hours (Li et al. 2012). Jayaraman et al. (2007) studied seasonal variations of particulate matter concentrations in Delhi, reporting that the concentrations were positively and significantly associated with temperature. The dilution of pollutants was observed along with the surface temperature increase. Positive correlation between temperature and particulate matter during pre-monsoon and monsoon seasons indicates that an increase of ambient temperature will always elevate the particulate level at the observation site.

The probable impact due to rainfall and humidity on particulate matter in pre-monsoon and monsoon seasons was generally high when compared to post-monsoon and winter seasons. The PM₁₀ and PM_{2.5} were negatively correlated with relative humidity in all four seasons. The correlation coefficients were -0.56 and -0.53 for PM₁₀ and PM_{2.5} in pre-monsoon and -0.48 and -0.36 for PM₁₀ and PM_{2.5} in monsoon, respectively. The absorption rate of PM₁₀ particulates increases with an increase in humidity, and the rain which acts as a natural scrubber during monsoon brings down the particulate level. A higher level of atmospheric moisture helps the suspended particles to stick together and become heavier particles that settle down. Thus, during pre-monsoon both coarse and fine aerosols became heavier particles and settled down. But

during monsoon season, even though the suspended coarse particles were settled down due to high humidity, the higher wind speed associated with the monsoon circulation continuously supply the coarser sea spray aerosols to the inland area. Thus, re-suspension of PM10 dust particles are maintained over the observational site. These are possible reasons that there was less correlation in monsoon season. However, this effect was less in pre-monsoon season resulting in a higher correlation. The frequent rain and clouds induces nucleation and in-cloud scavenging of finer aerosols and were not replenished by sea spray activity (Pruppacher and Klett 1978). Therefore, the correlation of PM2.5 with relative humidity was less compared to PM10. Studies conducted at a tropical coastal station in Trivandrum on atmospheric boundary layer aerosols discussed the variation of aerosols in monsoon, which is attributed to the increase in sea spray aerosols (Vijayakumar et al. 1998). Rainy washout of aerosol particles and their wet deposition are the most effective mechanisms for atmospheric cleansing (Yamagata et al. 2009). The high RF in August 2014 and June 2015 (Fig. 3) was associated with lower concentration for both PM10 and PM2.5

The overall correlation with rainfall for both PM10 and PM2.5 was in -0.3 range irrespective of seasons. Furthermore, the relationship was investigated by applying a coherence wavelet transform to PM10 and rainfall time series data for the period of March 2014–February 2016. Wavelet coherence values range between 0 and 1 and provide a quantitative representation of the covariance between two time series as a function of frequency (Lakshmi et al. 2004). Significant relationships between PM10 and RF were obtained for 5–10 and 30 day oscillations (Fig. 8a). This co-variation between rainfall and PM10 is an indication of the role of rainfall as wet deposition, which removes particulate matter from the atmosphere. The atmosphere tends to be more stable after a rainfall event, thus inhibiting dispersion and promoting re-accumulation of particulates (Shaharuddin et al. 2008). The power spectrum of the wavelet analysis on daily values of PM10 shows 3–4 day oscillations, which are related to the synoptic weather activities (Fig. 8b). Our results show that MJO (30–60 day oscillations) influences is not much significant compared to 10 day oscillations in PM concentrations during monsoon season.

Fig. 8 **a** Plot showing coherence wavelet transform applied in PM10 and rainfall time series data. **b** Wavelet power spectrum of PM10 time series data



To examine the precipitation effect of rainfall on PM concentration, rainfall during the monsoon season was classified into three groups: (1) no rain, (2) rainfall less than 1 mm, and (3) rainfall greater than 1 mm. Therefore, in 2014, the PM₁₀ concentrations corresponding to each group were found to be, 26.8 ± 12.2 , 17.9 ± 8.9 , $16.3 \pm 8.8 \mu\text{g m}^{-3}$ and for PM_{2.5}, it was 11.5 ± 7.2 , 7.9 ± 5.7 , $7.1 \pm 5.5 \mu\text{g m}^{-3}$. The concentration levels of PM₁₀ for each group in monsoon 2015 were 26.5 ± 11.7 , 23.5 ± 9.5 , $20.3 \pm 9.0 \mu\text{g m}^{-3}$ and 12.4 ± 7.7 , 10.8 ± 6.2 , $9.7 \pm 5.8 \mu\text{g m}^{-3}$ for PM_{2.5}.

The particulate matter concentration was decreased by 33% (PM₁₀) and 32% (PM_{2.5}) in 2014 for rainfall less than 1 mm. In the same for the category of rainfall greater than 1 mm, the reduction of suspended pollutants was 40 and 38%, respectively. This indicates that higher rainfall reduces concentration percentage more than the lower rainfall. But percentage of reduction contributed only 7%. In 2015, the station receives less continuous rainfall episodes as compared to 2014 monsoon. The concentration decreased 12% (PM₁₀) and 10% (PM_{2.5}) during low rainfall (category 2) and 24% (PM₁₀) and 22% (PM_{2.5}) for

high rainfall (category 3), respectively. This indicates that high rainfall events sustained for long periods of time reduce the pollutants, but suspended particles are replenished easily after a heavy rainfall that happens in a short period. Light rainfall that sustains for a long time effectively removes the suspended aerosol particles from the atmosphere (Sarkar et al. 2015).

The bivariate polar plot (Fig. 9) shows the season-wise mean concentration of PM₁₀ (upper panel) and PM_{2.5} (lower panel). The wind speed and direction were used to identify the sources responsible for concentrations (Carslaw and Ropkins 2012). In all seasons, the high PM_{2.5} concentrations (Fig. 9e–h) are associated with low wind speed indicating the presence of local pollutants, but for PM₁₀ the highest concentrations are found mostly with strong winds, representing the presence of local emissions, as well as long-range transport. For PM₁₀, air masses with maximum concentration of particles are transported from each quadrant i.e., from southwest (in pre monsoon), northwest (in monsoon), northeast (in post-monsoon), and southeast (in winter) (Fig. 9a, d). Distinct features are observed in the PM_{2.5} concentrations in each season. In Fig. 9e–h, the air masses show a bi-directional pathway in

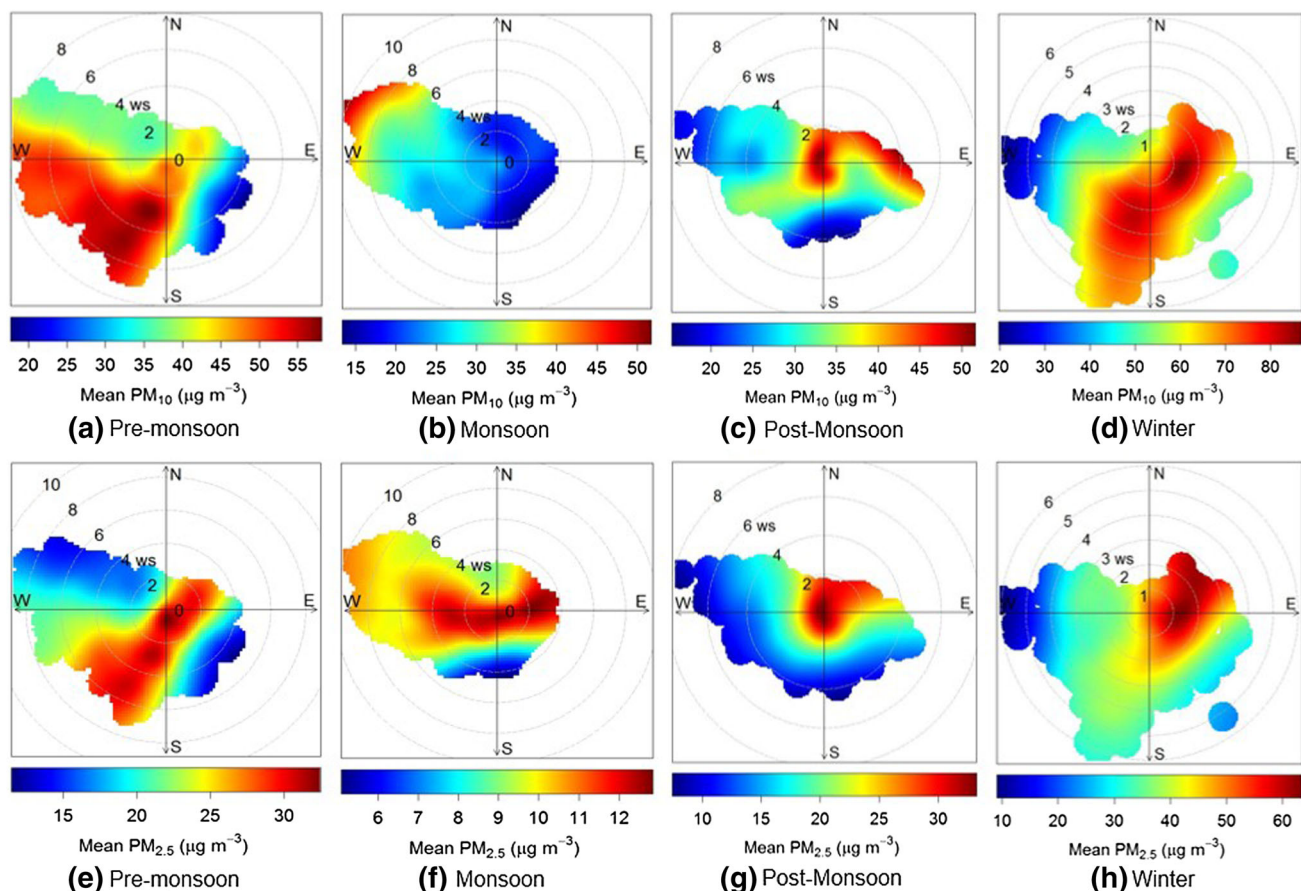


Fig. 9 Season-wise bivariate polar plots of the PM₁₀ (a–d) and PM_{2.5} (e–h) concentrations with wind speed and direction over Thiruvananthapuram. The mean concentrations are shown by the color scale

two seasons. i.e., from southwest and northeast (in pre-monsoon), west and east (in monsoon) direction. During post-monsoon, PM_{2.5} has higher concentrations in every direction with low wind speed ($<2 \text{ ms}^{-1}$), which does not favor the dispersion processes. Most importantly in winter, higher concentrations of PM_{2.5} are associated with light breeze from the northeast direction, which is due to local emissions from the urban area (densely populated including schools, colleges, and highly traffic roads). Eventually, the study area is influenced by the highest PM₁₀ and PM_{2.5} concentrations from the southwest direction by the moderate breeze in pre-monsoon. This indicates the influence of emissions from an industrial area (3 km away from the observation site) consisting of Travancore Titanium industry, English Indian clays, wood industries.

4.5 Exploration of particulate matters source directions

The conditional bivariate probability function (CBPF) (Uria-Tellaetxe and Carslaw 2014) was used to identify and characterize the source contributions at receptor locations. The CBPF analysis is an extension of the conditional probability function (CPF) combines with bivariate polar

plots for additional source characterization. The CPF (Ashbaugh et al. 1985) is a statistical method that identifies the emission sources of PM based on the probability that the measured concentration exceeds a predetermined threshold level for a particular wind direction.

$$\text{CPF}_{\Delta\theta} = \frac{m_{\Delta\theta}|c \geq x}{n_{\Delta\theta}}, \quad (1)$$

where $m_{\Delta\theta}$ is the number of samples in the wind direction θ with concentration c greater than or equal to a threshold value x , and $n_{\Delta\theta}$ is the total number of data samples from same wind direction.

But the CBPF uses wind speed as a third variable in the CPF and allocates the measured PM concentrations to cells defined by ranges of wind direction and wind speed rather than to wind direction only.

$$\text{CBPF}_{\Delta\theta, \Delta u} = \frac{m_{\Delta\theta, \Delta u}|c \geq x}{n_{\Delta\theta, \Delta u}}, \quad (2)$$

where $m_{\Delta\theta, \Delta u}$ is the number of samples in the wind direction $\Delta\theta$ with wind speed interval Δu with concentration c greater than or equal to a threshold value x , and $n_{\Delta\theta, \Delta u}$ is the total number of data samples in that wind direction and wind speed interval.

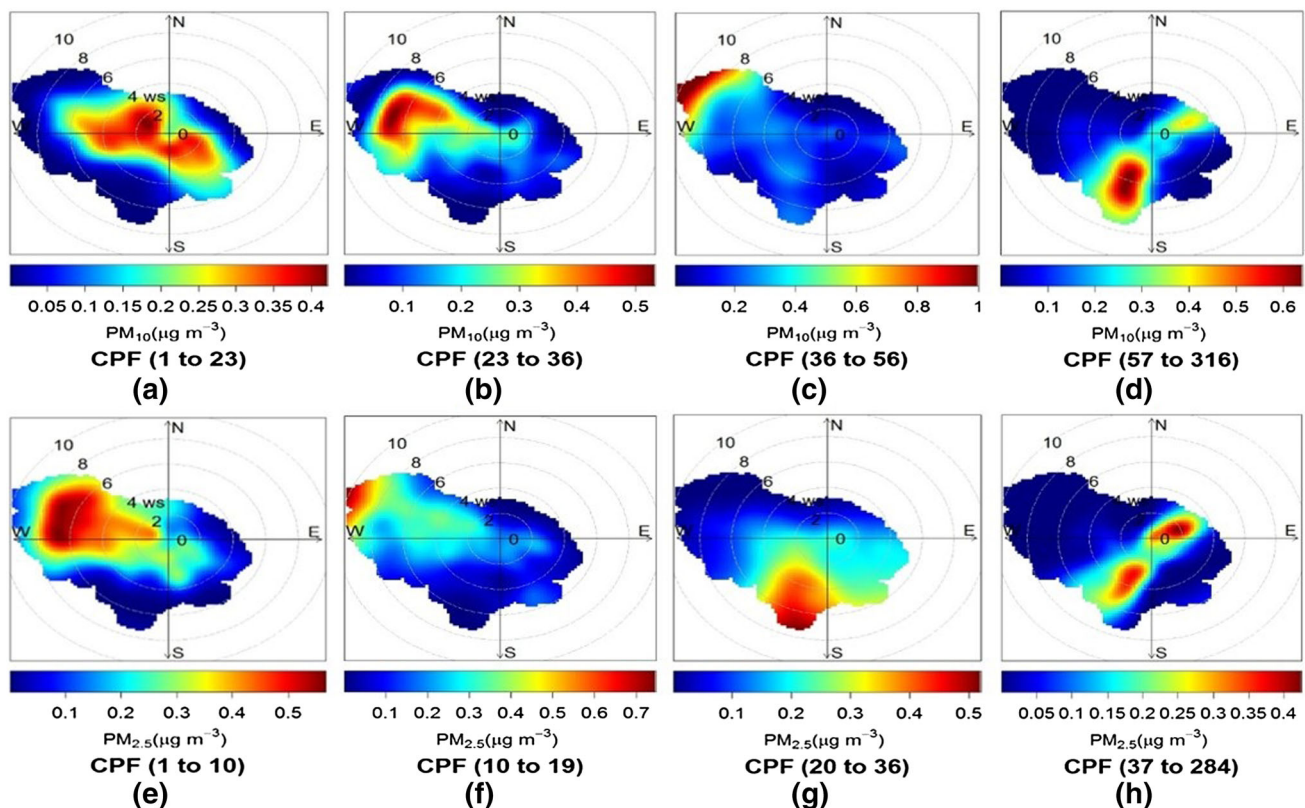


Fig. 10 Conditional bivariate probability function plots of PM₁₀ (a–d) and PM_{2.5} (e–h) concentrations during the study period from March 2014 to February 2016. The colored scales represent the CPF probability

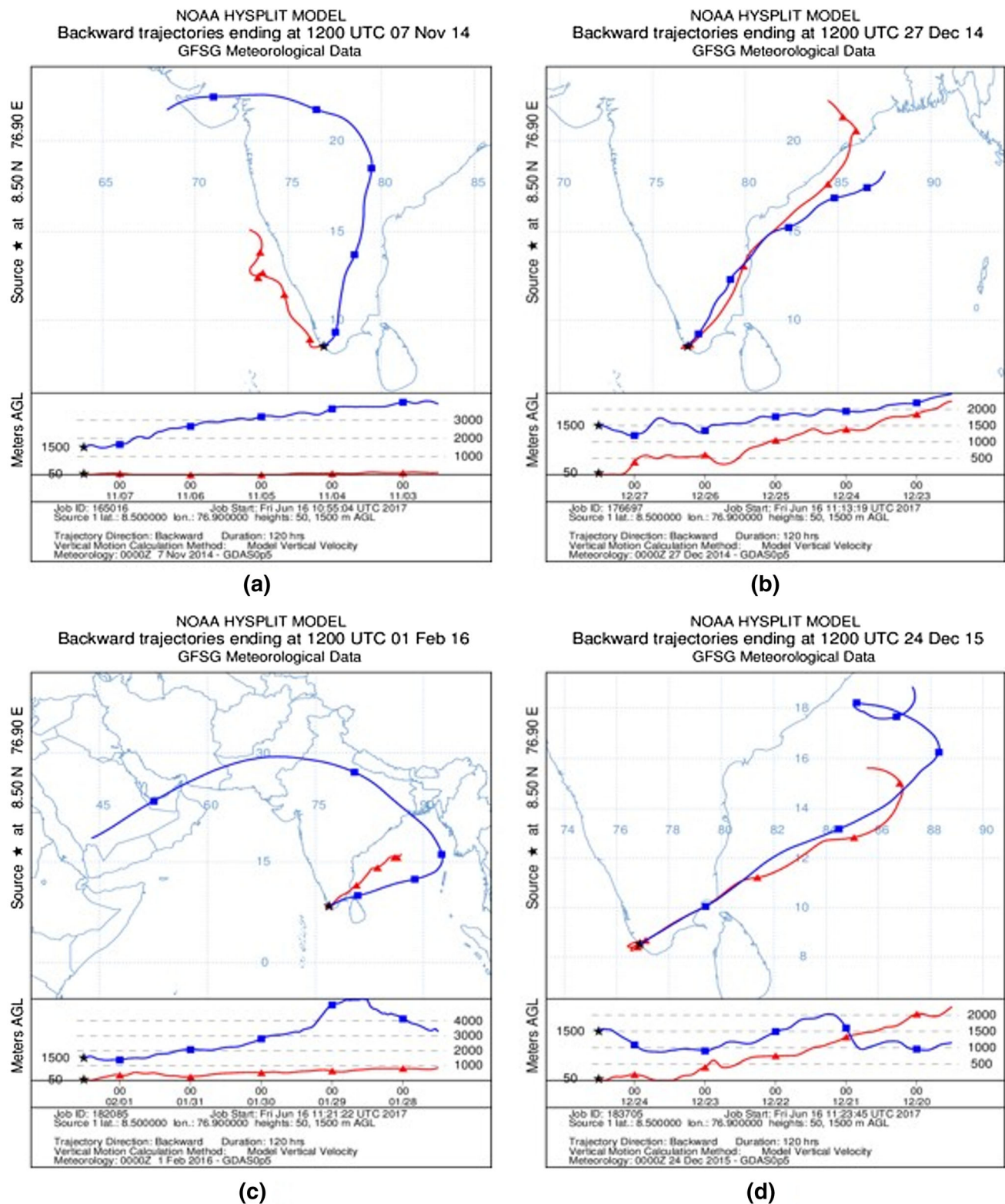


Fig. 11 5-day back trajectory (HYSPLIT model) analysis of the air masses in the observation site during **a** 7th November 2014, **b** 27th December 2014, **c** 1st February 2016, and **d** 24th December 2015

The CBPF can be used to consider full distribution of concentration rather than only values greater than threshold, and it is defined as:

$$\text{CBPF}_{\Delta\theta, \Delta u}(i) = \frac{m_{\Delta\theta, \Delta u|y \geq c \geq x}}{n_{\Delta\theta, \Delta u}}, \quad (3)$$

where $m_{\Delta\theta, \Delta u}$ is the number of samples in the wind sector $\Delta\theta$ with wind speed interval Δu with a concentration c between the intervals x and y , and $n_{\Delta\theta, \Delta u}$ is the total number of samples in that wind direction-speed interval. This CBPF methodology provides a much more comprehensive information for source identification rather than the ordinary CPF method.

Previous study (Uria-Tellaetxe and Carslaw 2014), suggested that in order to identify concentration intervals that best highlight a particular source, a large series of CBPF plots across a range of percentile intervals needs to be generated. For example, a large series of plots can be generated by considering particulate matter concentration from 0 to 100, at percentile intervals of 10. Therefore, here we used four intervals of PM10 (Fig. 10a, d) and PM2.5 (Fig. 10e, h) concentration, which represent percentile concentration values of 0–25, 26–50, 51–75, and 76–100%. For PM10, CBPF shows that a source located in west-northwest

direction with PM10 emissions from 36 to 56 $\mu\text{g m}^{-3}$ range of concentrations have high probabilities (Fig. 10c). Also, for PM2.5, high probabilities are in the same direction with concentrations in the range of 10–19 $\mu\text{g m}^{-3}$ (Fig. 10f). Thus, the CBPF method distinguish sources from the same wind direction, depending on their characteristics in high wind speed. The nearby sources from south-southwest directions have an impact in high concentrations of both PM10 and PM2.5 because high values of PM10 (57–316 $\mu\text{g m}^{-3}$) and PM2.5 (37–284 $\mu\text{g m}^{-3}$) were obtained during wind speed $< 5 \text{ ms}^{-1}$ as shown in (Fig. 10d, h). Figure 10h indicates that there are two sources for the highest PM2.5 concentrations, one dominant from the southwest direction and the other from the east-northeast for wind speeds below 6 ms^{-1} , which represents the contribution of local emissions. The lower PM2.5 concentrations are from the west-northwest with high wind speed suggesting transported dust from the populated area in that direction. The low concentrations of PM10 are also in same direction, but for low wind speed ($< 2 \text{ ms}^{-1}$), shows that the traffic emissions from the nearby roads. In contrast, the CBPF plots provide an idea of the various emissions sources affecting the PM10 and PM2.5 concentrations at Thiruvananthapuram.

Table 2 PM 10 concentration ($\mu\text{g m}^{-3}$) greater than mean + 3SD during study period

Year 1		Year 2	
Day	Concentration ($\mu\text{g m}^{-3}$) > PM (mean + 3SD)	Day	Concentration ($\mu\text{g m}^{-3}$) > PM (mean + 3SD)
07 November 2014	91	11 March 2015	84
08 November 2014	91	20 March 2015	85
25 November 2014	84	21 March 2015	91
26 November 2014	102	24 December 2015	96
27 November 2014	87	16 January 2016	85
04 December 2014	100	18 January 2016	89
05 December 2014	87	19 January 2016	83
06 December 2014	96	21 January 2016	90
07 December 2014	110	28 January 2016	88
08 December 2014	105	29 January 2016	90
09 December 2014	111	30 January 2016	83
10 December 2014	112	1 February 2016	87
11 December 2014	153	3 February 2016	83
12 December 2014	98	5 February 2016	85
26 December 2014	87	6 February 2016	91
27 December 2014	102	8 February 2016	103
16 January 2015	83	13 February 2016	85
20 January 2015	92	25 February 2016	83
22 January 2015	92		
23 January 2015	105		
11 February 2015	97		
18 February 2015	85		

The concentration of particulate matter and its impact are influenced by its source regions and the pathway through which it transports. In order to know the possible transport pathways of particulate matter aerosols, air mass back-trajectories obtained from the HYSPLIT model was computed. Figure 11 shows 5-day air mass back-trajectories for the days (7 Nov 2014, 27 Dec 2014, 24 Dec 2015, and 1 Feb 2016) in which the PM₁₀ concentration exceeds the mean + 3SD values. Table 2 shows the observation days and higher PM₁₀ values. The back-trajectories were calculated at 12:00 IST local time for two different altitudes at 50 and 1500 m (i.e., below boundary layer). It is clearly seen from the figure that the back-trajectories over the observation site are coming from the northeast region at all the altitude level. At that time, the mean concentration of PM is higher, which confirms that the major source of particulate matters during winter period corresponds to both localized and long-range transport of pollutants. This results shows that the model back-trajectories (50 and 1500 m) of high PM₁₀ episodes at observation site show two dominant stream lines, one originate from Bay of Bengal region and another from Middle East Asia. This result shows the important role of long-range transport of PM over the southern tip of India.

5 Conclusion

Particulate matter (PM₁₀ and PM_{2.5}) mass concentration measurements were conducted over a coastal location in Thiruvananthapuram from March 2014 to February 2016. The daily, monthly, and seasonal time scale variations of PM values and the influence of meteorology were examined. The analyses revealed that the annual mean concentrations of PM₁₀ were $44.5 \pm 14.6 \mu\text{g m}^{-3}$ (March 2014–February 2015), $40.3 \pm 11.9 \mu\text{g m}^{-3}$ (March 2015–February 2016) and PM_{2.5} were $27.7 \pm 10 \mu\text{g m}^{-3}$ (March 2014–February 2015), $24.4 \pm 7.8 \mu\text{g m}^{-3}$ (March 2015–February 2016), respectively. At present, the concentrations are under standard level, but ongoing urban activity may increase these concentration levels in near future. The PM₁₀–2.5 coarse particle concentrations during the study period were found to be $17.38 \pm 3.64 \mu\text{g m}^{-3}$. The mean PM_{2.5}/PM₁₀ were found to be $0.56 \pm 0.13 \mu\text{g m}^{-3}$ (March 2014–February 2015) and $0.54 \pm 0.08 \mu\text{g m}^{-3}$ (March 2015–February 2016) over the station. On a seasonal basis, the PM₁₀ was highest during winter and low during monsoon. PM_{2.5} followed a similar seasonality. However, coarse particles PM₁₀–2.5 were higher during winter and low during post-monsoon. PM_{2.5} and PM₁₀ concentrations showed a diurnal pattern with two maximum peaks one during the morning (~08:00 IST) and other in the evening (~20:00 IST) and minimum between

12:00 IST to 17:00 IST. The percentage of larger PM₁₀ concentration is almost equal (~50.4%) during day and night, while for PM_{2.5} it was observed during the daytime (~55%). The anthropogenic activities during the morning and evening hours, along with the wind speeds and boundary-layer height are the most important controlling factors for the diurnal variations of particulate matter at Thiruvananthapuram.

To determine the impact of meteorological parameters on particulate matters correlation analysis was performed between PM₁₀ and PM_{2.5} with wind speed, temperature, relative humidity, and rainfall. The PM concentrations were reduced by rainfall and wind speed, which resulted in a negative correlation. The bivariate plots show that high PM_{2.5} concentrations are associated with low wind speeds indicating the presence of local pollutants, while for PM₁₀ the highest concentrations are found with calm to strong winds, which represents the presence of local emissions as well as long-range transport. The CBPF analysis was performed to identify the probability of source contributions to PM concentrations at different wind speed and direction. The results show that the highest probability to get high PM concentration is from the south-southwest direction with wind speed $<5 \text{ ms}^{-1}$. The HYSPLIT model back-trajectories (50 and 1500 m above mean sea level) of high PM₁₀ episodes show two dominant streamlines, one from the Bay of Bengal and another from the Middle East Asia. This highlights the importance of long-range transport of PM towards the southern tip of India.

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