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Seasonal Variation of Ultrafine Particulate Matter (PM1) and Its Correlation with Meteorological Factors and Planetary Boundary Layer in A Semi-Arid Region

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ABSTRACT

The present study critically investigated the effect of meteorological parameters on the mass concentration of Ultrafine Particulate Matter **(**PM$_1$) between October 2018 and September 2019 (n=102) in a semiarid region of Rajasthan, India. The concentration of PM$_1$ ranged between 72-110.85 µg.m^{-3} with distinct seasonal variation. Higher PM$_1$ concentrations are closely linked to decreased wind speeds and colder temperatures, according to the findings. The winter season showed the highest concentration followed by post monsoon and pre monsoon season. The cumulative effect of environmental variables such as temperature, relative humidity, and wind speed, as well as the height of the planetary boundary layer, was investigated using multiple regression analysis (HPBL). A significant negative correlation ($p < 0.001$) with HPBL and wind speed was observed in all three seasons. The temperature was found to have a significant ($p<0.001$) negative correlation during winters whereas in other seasons there was a positive but no significant ($p>0.001$) relationship. Relative humidity showed a negative relationship during withers and pre-monsoon season. The multiple regression model indicated a significant negative ($p<0.001$) relationship with HPBL in winters ($R^2=0.70$) explaining the 70% effect of HPBL on mass concentration of PM$_1$.During the post-monsoon ($R^2 = 0.69$) and pre-monsoon ($R^2= 0.91$) explains 69% and 91% effect of HPBL on mass concentrations of PM$_1$.The results indicate that the concentration of PM$_1$ cannot be explained by a single meteorological parameter but all the parameters show a cumulative effect.

INTRODUCTION

Atmospheric aerosols have increased drastically in the last few decades and continuously deteriorating the air quality, impacting the quality of life, and have become an important parameter for evaluation in developed as well as in developing countries. The ubiquitous nature of atmospheric aerosol shows an intense impact on the earth's atmospheric system, climatic conditions, atmospheric chemistry, influence weather conditions, ecosystem, air quality, and public health (Pöschl 2005, Solomon et al. 2007). High-intensity exposure may cause both acute and chronic effects on different organs of the body by interacting with the immune system leading to the risk of chronic respiratory and heart diseases, lung cancer, acute respiratory infections in children, chronic bronchitis in adults, and asthmatics attacks (Chen et al. 2007, Kampa & Castanas 2008). Particulate matter with an aerodynamic size of less than 10 microns deposits primarily in the upper respiratory tract, whereas fine particles with an aerodynamic size of less than 2.5 microns and ultrafine particles with an aerodynamic size of less than 0.1 microns reach the alveolar spaces of the lungs and cause or exacerbate respiratory diseases (Satsangi et al. 2011), making it a major concern. Because fine particle matter contains a higher concentration of toxins, ultrafine and fine particulate matter provide a greater risk of cardiovascular and respiratory consequences, as well as mortality than coarse particulate matter.

Increased consumption of fossil fuels as a result of rapid urbanization and industrialization has resulted in significant emissions of pollutants into the lower atmosphere (Sharma et al. 2014), prompting major worry in Asian countries (Baldasano et al. 2003). Due to rapidly increasing population numbers, expanding industrialization and vehicular density, traffic jams, poor road conditions, and poor regulation of industrial emissions, air quality in Jaipur city has reached hazardous levels. (Dhamaniya & Goyal 2004, Kala et al. 2014). Meteorological parameters are one of the most important factors to influence particulate matter concentrations. Among them, temperature, relative humidity, wind speed, and direction play a crucial role in dispersion, accumulation, removal process, and formation of atmospheric aerosols in the lower atmosphere (Galindo et al. 2011, Goyal &Rao 2007), therefore they significantly control the concentrations of pollutants. In addition to other factors, the height of the planetary boundary layer (HPBL) also plays a critical role in regulating pollutant concentration in any region. Evaluating mass concentration of Particulate matter in alliance with HPBL is important for identifying air pollution (Du et al. 2013) with varying seasonal changes.

This paper presents a seasonal variation in mass concentration of particulate matter (PM_1) in Jaipur city of Rajasthan state in India, for a period between October 2018 to June 2019. The main objective of the study is not only to quantify the concentration of PM_1 but also to investigate the statistical relationship between PM_1 and meteorological parameters (Temperature, Relative humidity, wind speed, and Planetary Boundary layer height.

MATERIALS AND METHODS

Site Description

The present study was conducted at Albert hall museum in Jaipur, the capital city of Rajasthan located at 26°1'36" north latitude and 75°4'32" east longitude in the eastern parts of Thar Desert (Fig. 1). Albert hall museum built in the year 1876, is located near the walled city is a heritage site with a large number of tourists from all over the world. The area experiences a very heavy traffic load due to the proximity to commercial areas like Ajmeri gate, Johri Bazar, Sanganer gate. Jaipur city covers an area of 200.4 km^2 with a population size of 4007505 (year 2021). The semi-arid land of the city is surrounded by rugged Aravali hills on three sides. The desert state Jaipur faces seasonal dust storms every year due to the downwind location of the Thar desert which accumulates a huge amount of dust loading aerosol over the city (Verma et al. 2013). Frequent dust storms and variation in climatology throughout the year lead to significant variability in air quality from summers to winters (Mohan & Kandya 2007, Ram et al. 2010). Average wind speed varies between 3.0 to 10.0 mph. The maximum temperature is observed in the summer season (March to May) varying between 40°C to 47°C rising 4-6°C at times when heatwave prevails, whereas winters (December to February) are quite cold with a minimum temperature of 4-9°C or can be below 0°C when chilly winds northerly blows from the Himalayan region. Western disturbances lead to increased humidity, cloudiness, and rainfall activities during the monsoon period (July to September) in Jaipur city. July and August are the rainiest months and they last up to mid-September. Rainfall decreases sharply in October month so October and November are transitional months or post-monsoon seasons. The annual rainfall is 492 mm (Singh et al. 2012). The humidity reaches the highest value in August and gradually decreases in November again rising in December and January whereas the lowest value for humidity is observed in April (Tyagi et al. 2012).

Data Collection

Ambient air Sampling and analysis were done as per the guidelines given by Centre Pollution Control Board (CPCB) New Delhi. Aerosol sampling for PM_1 was done for nine months considering pre-monsoon, winter, and post-monsoon seasons. Twelve hours of continuous sampling (9:00-21:00) was done thrice in a weak for collection of fine particulate matter (PM_1) using a fine particulate sampler (Envirotech APM 577) with a flow rate of 10 L per minute. PTFE What man filter paper with pore size 0.2 μm and a diameter of 25 mm was used for the collection of particulate matter. The sampler was placed at a height of 14 m. The filter paper was desiccated for 24 hours before and after sampling before proceeding further for estimation of mass concentration with weighing balance with a precision of 0.01 mg. For further analysis, the filter paper was wrapped in aluminum foil to avoid moisture and loss of particles. After weighing, the filters were placed in the refrigerator at (-20° C) before extraction for chemical analysis. Mass concentration of particulate matter was estimated gravimetrically by the following formula:

$$PM_1\ (\mu g/m^3) = \frac{(w_f - w_i) * 10^6}{v}$$

Where,

w_f = Final weight of sample filter paper (g)
w_i = Initial weight of blank filter paper (g)
v = Volume of air sampled in (m^3)
10^6 = Conversion of g to μg ($1g = 10^6$ μg)

Meteorology Data

Data for metrological parameters such as wind speed, relative humidity, the temperature were collected from weather underground https://www.wunderground.com/.

Backward Trajectory Simulation

Planetary boundary layer data was achieved from reanalysis data by running a backward trajectory simulation model obtained from the National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory (ARL) or the provision of the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) transport and dispersion model from the website (http://www.arl.noaa.gov/ready.html) (Draxler & Hess 1998, Draxler & Rolph, 2003). Both forward and backward trajectories can be calculated to interpret the airflow patterns and dispersion of air pollutants in spatial and temporal boundaries. Further, these trajectories

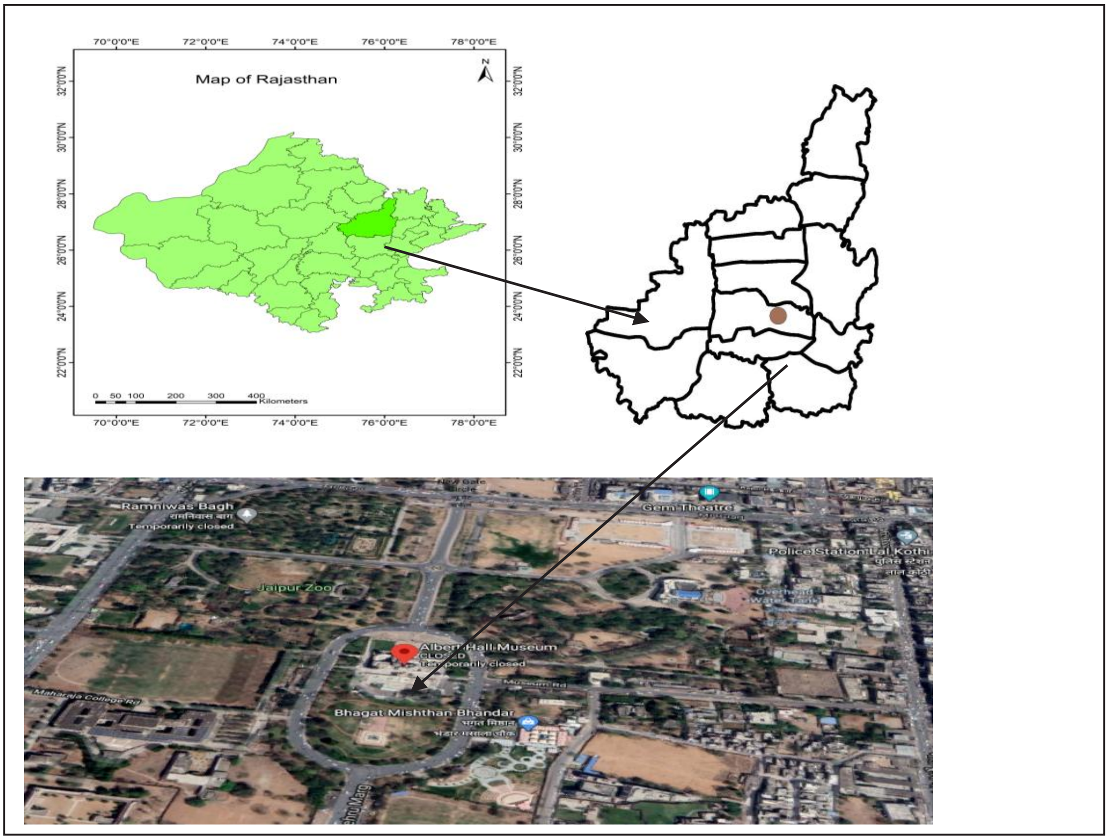

Fig. 1: Geographical map showing Rajasthan state and Jaipur district map with a satellite image of Albert Hall sampling site. source: Google

can be used to forecast the history of air mass movement and wind patterns (Fleming et al. 2012). The nine months data was achieved with GDAS metrological data at the altitude of 500 m above the ground level.

Statistical Analysis

Data analysis was performed to evaluate the impact of metrological parameters and HPBL on PM_1 concentration. Descriptive statistics were applied to examine the average seasonal value and trend followed by PM_1 concentrations and metrological parameters in three different seasons. Based on the data set we analyzed seasonal variation of PM_1 by using Pearson's correlation coefficient and multiple regression model (MLSR).

RESULTS AND DISCUSSION

Seasonal Variation in PM_1

In this study, the data was collected and analyzed for Particulate Matter (PM_1) and metrological parameters from October 2018 to June 2019 in Jaipur city. The study focused on three important seasons: post-monsoon (October-November), winter (December-February), and pre-monsoon (March-June) because these seasons show the most fluctuation in meteorological parameters in semi-arid areas like Jaipur. As depicted in Fig. 2PM_1 shows prominent seasonal variation among all three seasons in which maximum concentration was observed in winter seasons followed by post-monsoon and pre-monsoon season with the average value of 110.85±15.78 µg.m^{-3}, 90.63±9.75 µg.m^{-3}, and 72±2.20 µg.m^{-3} respectively. The observed levels of PM_1 are higher when compared with the annual standard limit of National ambient air quality standards (i.e. 60µg.m^{-3} for 24 h) for $PM_{2.5}$ or size less than 2.5 microns provided by the Central Pollution Control Board. Almost similar concentrations of PM_1 (135.0µg.m^{-3}) in the foggy period and (54.0 µg.m^{-3}) in the non-foggy period were reported by Mangal et al. (2020) in Agra city, India. Whereas PM_1in the present study was found lower than reported by Zhang et al. (2015a) in China (212 µg.m^{-3}) in the heavy haze-fog period, but higher than reported in Turkey (30.2 µg.m^{-3}) and at the urban areas of China (5.44-105.91 µg.m^{-3}) (Onat et al. 2013, Wang et al. 2020).

The concentration of PM_1 is highly influenced by different metrological parameters (Stull 2012, Dadhich et al. 2017) like Planetary boundary layer (PBL), Temperature, Relative Humidity, Wind Speed. The planetary boundary layer (PBL) showed a wide range of variation in all seasons, and the mean height for PBL was maximum in pre-monsoon season followed by post-monsoon and winters season (Fig. 2a). Similarly, the average daily temperature in winters was minimum followed by post-monsoon and maximum in pre-monsoon season (Fig. 2b). Wind speed showed a gradual increase from winters to pre-monsoon and found maximum in the post-monsoon season with a very narrow range (Fig. 2d). The highest RH values were found in winters. Generally, RH shows an increasing trend from pre-monsoon to post-monsoon and winters (Fig. 2c).

Influence of Metrological Factor on PM_1 Concentration

The relationship between PM_1 and metrological parameters was evaluated by using the Pearson correlation coefficient for nine months including the three major seasons mentioned earlier. The dispersion of particulate particles across time Chemical composition and pollutant concentrations in the lower atmosphere are influenced by meteorological factors such as relative humidity, wind speed, and temperature (Yin et al.2016, Asl et al. 2018). In the winter, low temperatures, thermal inversion, stagnant air, and calm weather conditions mostly hampered the dispersion of air pollution (Tripathi et al. 1996).

PM_1 and Temperature

Pearson correlation revealed different seasonal patterns for PM_1 and temperature (Table 1 and Fig. 3 (b,f,j). A significant negative correlation ($R = -0.59$ $p<0.01$) between PM_1 and atmospheric temperature in winter season and moderate positive correlation in Post-monsoon ($R = 0.22$ $p>0.05$) and pre-monsoon seasons ($R = 0.12$, $P>0.05$) was observed. PM_1 pollution is more severe in winters which can be attributed to the stable atmospheric conditions and low boundary layer height as temperature inversion during winters in semiarid regions like Jaipur prevails with high frequency. In addition to this, high atmospheric pressure and lower mixing height, stable atmospheric conditions facilitate the high concentration of pollutants in the winter season by restricting the vertical diffusion of air pollutants (Gamo et al. 1994, Lorga et al. 2015, Malandrino et al. 2013, Xu et al. 2018). Similar results indicating a higher concentration of PM with the decrease in temperature were reported by Asl et al. (2018) in Iran, Dadhich et al. (2017) in Jaipur, Galindo et al. (2011) in Spanish Mediterranean, and Lorga et al.(2015) in Bucharest. Whereas in summers atmospheric temperature near the earth surface is maximum, and heat flux value is high, which intensify the vertical mixing height, cause stronger convection

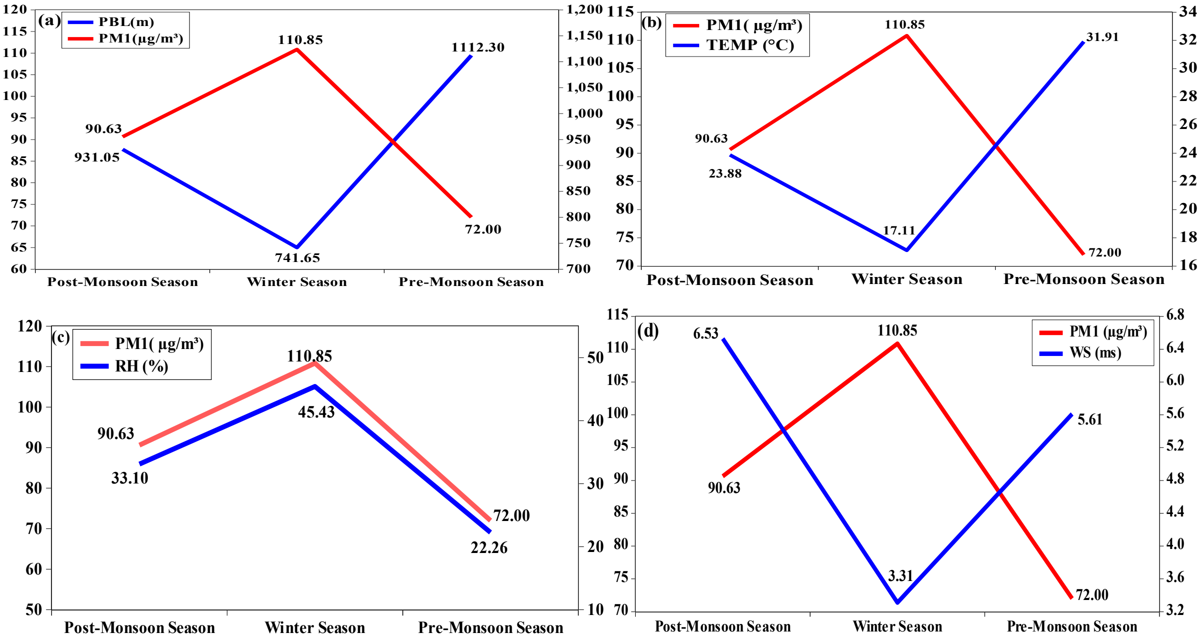

Fig. 2: Average seasonal variability of PM_1 concentration with varying metrological parameters in post-monsoon, winters, and pre-monsoon season. (a) PM_1 concentration and Planetary boundary layer (PBL) (b) PM_1 concentration and Temperature (Temp) in (c) PM_1 concentration and relative humidity (RH) (d) PM_1 concentration and wind speed (WS) for the period of sampling.

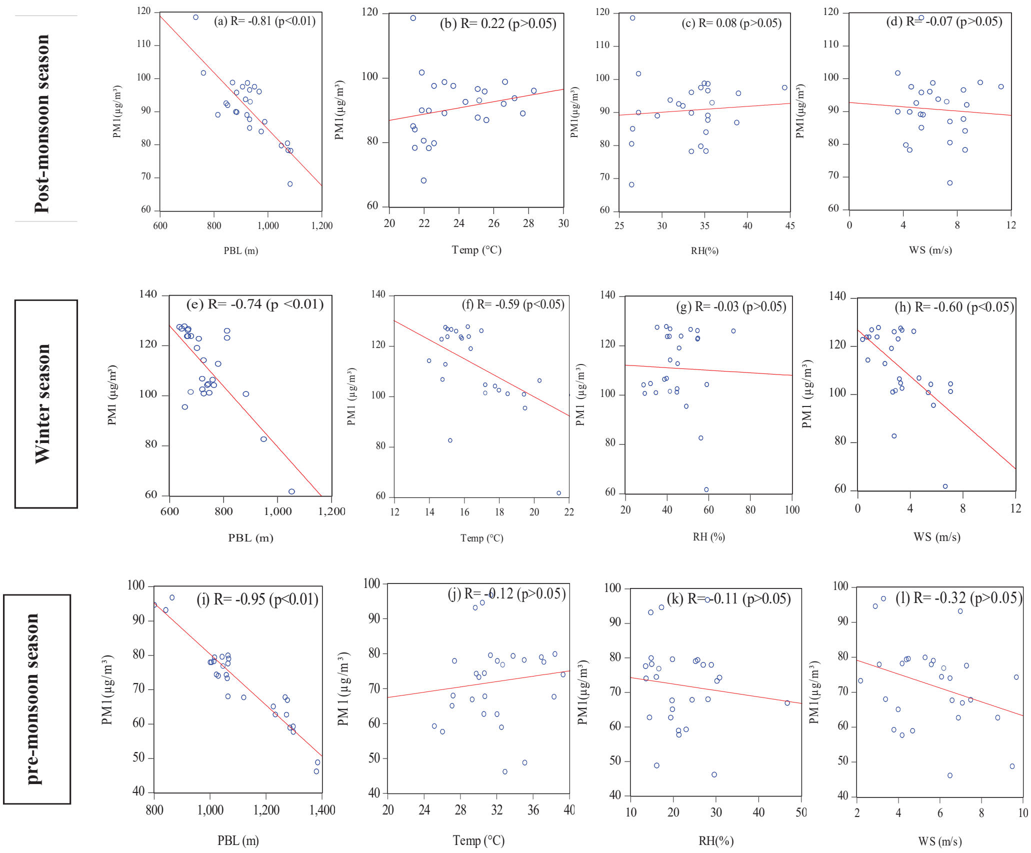

Fig. 3: Scatter plot for the monthly mean concentration of PM1 with various metrological parameters, i.e. (a,e,i) mean height of planetary boundary layer (PBL) in meters, (b,f,j) mean temperature in degree Celsius, (c,g,k) mean relative humidity in (%) and (d,h,l)mean wind speed in meter/second in three different seasons Post-monsoon season, winter season and pre-monsoon season.

condition and unsteady atmospheric conditions eventually minimizing the concentration of Particulate matter (Gamo et al. 1994, Jayamurugan et al. 2013, Sari et al. 2019, Asl et al. 2018). In addition to this geographical position of the Thar desert, Rajasthan has much influence on aerosol loading and dispersion of pollutants in the summer season, facilitating the accumulation of aerosol in the lower atmosphere leading to the problem of particulate pollution (Kisku et al. 2013).

PM_1 and Relative Humidity

Elevated levels of RH accelerate the formation of secondary pollutants and split semi-volatile species into aerosol further contributing to the fine particulate matter (Hu et al. 2008, Sun et al. 2013). Meanwhile, the moist atmosphere generally forms a lower boundary layer, enhancing the concentration of primary pollutants in the lower atmosphere (Sandeep et al. 2014). High concentrations of PM_1 generally coexist with high relative humidity during winters. As the percentage of RH increases in winters, simultaneously hydrophilicity of aerosol increases, and the radius of particle increases to double by adsorbing water droplets on the surface of the particle (Liu et al. 2011).In the present study, when mean Relative humidity increased from 22.26 to 33.1% and then to 45.43% from the pre-monsoon to post-monsoon and to the winters, the mean concentration of $PM_1$was found to increase from 72 µg.m^{-3} to 90.63 µg.m^{-3}and 110.85 µg.m^{-3}respectively. The impact of RH on PM_1 concentration is high during winters whereas in summers high RH is associated with precipitation and cleaning the air (Meng et al. 2019). PM_1 and RH were found to be moderately negatively correlated in the pre-monsoon season (R=-0.11, p>0.05) whereas statically no significant correlation was observed during the winter season (R= -0.03 p>0.05) and a positive correlation in the post-monsoon season (R = 0.08 p>0.05) was observed (Table1and Fig. 3(c,g,k)

PM_1 and Wind Speed

Wind speed and directions are the two most important factors for diluting the concentration of particulate matter from the atmosphere in any region (Asl et al. 2018).In the present

Table 1: Pearson correlation analysis between PM_1 and meteorological parameters.

Post-monsoon season	Metrological parameters	PM1	PBL	TEMP	RH	WS
	PM1	1.00	-0.81^{**}	0.22	0.08	-0.07
	HPBL		1.00	-0.13	0.08	0.19
	TEMP			1.00	0.30	0.27
	RH				1.00	0.47^{*}
	WS					1.00
Winter season	PM1	1.00	-0.73^{**}	-0.59^{**}	-0.03	-0.59^{**}
	HPBL		1.00	0.38	0.30	0.39^{*}
	TEMP			1.00	-0.04	0.71^{**}
	RH				1.00	0.08
	WS					1.00
Pre- monsoon season	PM1	1.00	-0.95^{**}	0.12	-0.11	-0.32
	HPBL		1.00	-0.12	0.17	0.36
	TEMP			1.00	-0.38^{*}	0.33
	RH				1.00	-0.13
	WS					1.00

**Correlation is significant at the 0.01 level (2-tailed)
*Correlation is significant at the 0.05 level (2-tailed)

study, Pearson's correlation coefficient between wind speed and $PM_1$was found to be is negatively correlated in all seasons. Similar results are reported by Galindo et al. (2011) at a traffic site in the city of Elche, Spain, and by Kozakova et al. (2017) in the Czech Republic, Central Europe. Negative associations between PM fractions (Coarse and Fine) and wind speed can be an indicator of the presence of significant local source(s) of PM (Chaloulakou et al.2003). As shown in Fig. 3 (d,h,l) the results of the present study revealed moderate negative correlation in post-monsoon ($R= -0.07$, $p>0.05$) and pre-monsoon season ($R= -0.32$, $p>0.05$) whereas significant negative correlation in the winter season ($R= -0.59$, $P<0.01$) between PM_1 levels and Wind speed (Table 1). Similar results were reported by Lorga et al. (2015) at Bucharest, Romania where a negative correlation was observed with wind speed. Higher PM_1 pollution in winters can be explained by low wind speed and a large decrease in planetary boundary layer height (PBLH) where both the facts restrict the horizontal and vertical dilution of pollutants in winters (Miao & Liu2019). Thus in a specific area wind plays a vital role in particulate concentration.

PM_1 and HPBL

As shown in Table 1 and Fig. 3 (a,e,i), PM_1 concentration was found to show a significant negative correlation with HPBL (height of planetary boundary layer) in all seasons, ($R= -0.8$, $p<0.01$) during the post-monsoon season, ($R= -0.74$, $p<0.01$) during Winters and ($R= -0.95$, $p<0.01$) during pre-monsoon season. As the mean height of planetary boundary increases from winters to post-monsoon then to pre-monsoon season i.e.741.65 m to 931.05 m and then to 1112.30 m respectively, the concentration of PM_1 decreased from winter (110.85 $\mu g.m^{-3}$) to post-monsoon (90.63 $\mu g.m^{-3}$) and further in pre-monsoon season (72 $\mu g.m^{-3}$). Normally there is a barrier (very low mixing rate) on the top of the planetary boundary layer (PBL) which bound the transportation of particles to the free troposphere (Sun et al. 2006, Yao et al. 2012). Less solar heating, Strong thermal stratification, and weak winds in the lower troposphere in winters lower the PBL height, which bounds the diffusion of pollutants and accumulates the particulate matter in shallow layer and reduces visibility (Medeiros et al. 2005, Miao et al. 2015, Zhang et al. 2015b). Whereas, intense solar radiation during the summer months creates favorable surface thermal conditions which destabilize the lower atmosphere, encouraging vertical mixing and increasing PBL height (Guo et al. 2016, Miao et al. 2012). In the present study, good air quality was observed in the pre-monsoon season (March-June) whereas the highest concentrations of PM_1 were observed in the winter season.

Multiple Regression Model

Normally change in metrological conditions causes more variation in aerosol concentration rather than pollutant emissions from the primary or secondary sources over a monthly or seasonal period (Chang and Lee 2007).To understand the cumulative effect of all the metrological factors on PM_1concentration, a multiple regression model was used in addition to person correlation in this study. A multiple regression model was performed between PM$_1$ (as dependent variable) and metrological parameters (as an independent variable). For presenting reliable results, Multicollinearity diagnosis was done using VIF variance inflation factor, Heteroskedasticity was checked using Goldfeld Quandt test in addition to Durbin Watson value for the obtained data.

Post monsoon season

$PM_1 = 153.80 – 0.085^* \mathit{PBL} + 0.313\ \mathit{TEMP} + 0.263\ \mathit{RH} + 0.070\ \mathit{WS}$
(7.783) (-6.485) (0.535) (0.857) (0.102)
0.00 0.00 0.598 0.400 0.919
$R^2 = 0.69$ $\bar{R}^2 = 0.63$ F = 11.70* P =0.00
D-W = 2.30 n = 26

Notes: Figures in parentheses are computed- t values
*indicates significance at 1% level.

The value of R^2 for the estimated regression model is 0.69 which implies that four explanatory variables explain 69% of the total variation in the dependent variable (PM$_1$). The value for R^2 is statically significant at a 1% level of significance as $p< 0.01$.The results show that a 1-meter increase in the height of PBL would reduce the PM$_1$ by nearly 0.085 µg.m^{-3}. The marginal effect of PBL on PM$_1$ is significant whereas the effects of other variables (Temp, RH, and WS) are not statically significant. The results are presented in the following equation.

Winter season

$PM_1 = 200.27 – 0.0102^* \mathit{PBL} – 1.094\ \mathit{TEMP} + 0.265\ \mathit{RH} – 1.944\ \mathit{WS}$
(14.22) (-6.160) (-1.134) (1.114) (-2.517)
0.00 0.00 0.26 0.27 0.01
$R^2 = 0.70$ $\bar{R}^2 = 0.64$ F= 12.98* P = 0.00
D-W = 1.42 n = 27

Notes: Figures in parentheses are computed- t values
*indicates significance at 1% level.

The value of R^2 for the estimated model is 0.70 which implies that four explanatory variables together explain 70% of the total variation in the dependent variable (PM$_1$). The value for R^2 is statically significant at a 1% level of significance at $P < 0.01$. The value of each estimated coefficient interprets the marginal effect on the dependent variable (PM$_1$). A 1-meter increase in the height of PBL would reduce the PM$_1$ by nearly 0.010 µg.m^{-3} during winters. Similarly, a 1 m.s^{-1} increase in WS would reduce the PM$_1$ by 1.94 µg.m^{-3}. Therefore, the marginal effect of WS in the winter season was highest followed by PBL. Whereas the effect of other variables (Temp and RH) is statically insignificant. The study by Manju et al., 2018 reported r^2 value for PM$_{2.5}$ is 0.79. Another study from China by Meng et al. (2019) reported r^2 (0.96) for PM$_{2.5}$ and metrological parameters in the winter season.

Pre-monsoon season

$PM_1 = 151.69 – 0.075^* \mathit{PBL} + 0.040\ \mathit{TEMP} + 0.098\ \mathit{RH} + 0.161\ \mathit{WS}$
(14.81) (-13.90) (0.174) (0.878) (0.362)
0.00 0.00 0.86 0.38 0.71
$R^2 = 0.91$ $\bar{R}^2 = 0.89$ F= 59.15* P = 0.00
D-W = 2.29 n = 28

Notes: Figures in parentheses are computed- t values
*indicates significance at 1% level.

The Value of R^2 for the estimated model for pre-monsoon season is 0.91 which implies that four explanatory variables together explain 91% variation in PM$_1$. The value for R^2 is statically significant at a 1% level of significance $P< 0.01$. The 1-meter increase in the height of PBL would reduce the PM$_1$by nearly 0.075 µg.m^{-3}. Therefore the marginal effect of PBL on PM$_1$ is significant whereas the effects of other variables (Temp, RH, WS) is insignificant.

CONCLUSION

The concentration of PM$_1$ was higher as compared to the standard prescribed by CPCB (60 µg.m^{-3} for PM size less than 2.5 microns).The linkages between PM$_1$ and metrological parameters studied at a semi-arid region, Jaipur by conducting the field measurements indicated a strong influence of HPBL, which is formed as a result of different metrological parameters like Temp, RH, WS. It is evident from the outcome of the study that the concentration of PM$_1$ cannot be determined based on the single metrological parameter, whereas all the metrological conditions cumulatively show a significant impact. In addition to this, a significant ($p < 0.001$) negative correlation with HPBL and WS was observed in all three major seasons, the temperature was also a significant negative correlation. The relative humidity also showed negative relationship during the winter and pre-monsoon seasons. The multiple regression model analysis showed a negative relationship with HPBL in the winter, post-monsoon, and pre-monsoon seasons. While observing an annual cycle, a prominent seasonal variability with the highest concentrations in the winter season was shown due to the lower height of PBL followed by the post-monsoon and pre-monsoon season due to higher PBL.

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