



# Spatial and seasonal variations in particulate matter and gaseous pollutants around integrated industrial estate (IIE), SIDCUL, Haridwar: a case study

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## Abstract

The holy city of Haridwar is now emerging as a key industrial destination of the north India due to the presence of IIE Haridwar. Due to increased anthropogenic activities like vehicular emissions and industrial discharges in the Haridwar region, air pollution levels has raised greatly. The objectives of the study were (1) to determine the Spatial and seasonal variations of air quality in and around IIE, Haridwar and (2) to find out the Air Pollution Index (API). For this monthly monitoring of pollutant gases (SO<sub>2</sub> and NO<sub>2</sub>) and particulate matter (SPM and RSPM) for two years (2012 and 2013) were done at six ambient air quality monitoring locations, one at each land use area (rural/agricultural, urban residential, protected area, industrial and commercial land use). The average SPM, RSPM, SO<sub>2</sub>, NO<sub>2</sub> in the study area were found as 86.92 µg/m<sup>3</sup>, 81.04 µg/m<sup>3</sup>, 23.23 µg/m<sup>3</sup>, 24.73 µg/m<sup>3</sup>, respectively. It was observed that the average SPM levels exceed WHO standard limit of 120 µg/m<sup>3</sup> at IIE Haridwar during study period. The average API during study period was 50.14 which indicates light air pollution. Meteorological conditions along with the distance from sources, particularly road transportation play a major role in deciding the ambient air quality over a region.

**Keywords** Air pollution index · SPM · RSPM · Land use · Health effects · Ambient air quality

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## 1 Introduction

Air pollution and the resultant impacts in India can broadly, be attributed to emissions from vehicles, industrial and domestic activities. These emissions are of two forms, viz. solid particles (SPM) and gaseous emissions (SO<sub>x</sub>, NO<sub>x</sub>, CO, etc.) and can damage human respiratory and cardio-respiratory systems in various ways. Suspended particulate matter (SPM) arises from incomplete combustion of solid, liquid or gaseous fuels; wastes from metallurgical, chemical and refining operations; incineration and numerous other processes. With the increase in the concentration of pollutants worldwide and their adverse health effects, the concern about this problem is gaining importance (Künzli et al. 2000). Besides, natural sources also contribute suspended materials like spores, salt water spray, pollens, etc. Particulate Matter related health effects include cardiovascular risks, (Pope & Dockery 2006; BIS 1999). The potential source of Respirable Suspended Particulate Matter (RSPM) generation includes exhaust from diesel engines. RSPM exposure can lead to respiratory ailments viz. chronic obstructive pulmonary disease, respiratory infections (Abdo et al. 2016; BIS 2006b). Study of air quality in Roorkee in Haridwar district showed main sources of RSPM<sub>10</sub> at sub-urban sites were road dust, traffic exhaust, tire abrasion and industrial emissions (Tyagi et al. 2012; Nayak et al. 2012). Air quality data in India suggest that the pollutant is of most concern from the point of view of environmental health risk in the airborne particulate matter (MoEF 2009a). Ambient concentrations of fine particulate matter have been exceeding by far with WHO limits and national air quality standards (Amann et al. 2017). Epidemiological evidence supports an association between exposure to ambient air pollutants (particulate matter (PM), nitrogen oxides (NO<sub>x</sub>), sulfur oxides (SO<sub>x</sub>), metals, volatile organics (VOCs), Ozone (O<sub>3</sub>), etc.) and various health effects, such as respiratory symptoms or illness impaired cardiopulmonary function, reduction of lung function and premature mortality (Gul et al. 2011). It has been reported that high levels of pollution affect mental and emotional health too (MoEF 2009a). Sulfur is usually present in fuels in varying quantities and is discharged as sulfur dioxide. Sulfur dioxide causes air pollution on large scales as fossil fuels are being consumed universally, however their concentration in atmosphere is governed by number of potent sources acting locally, height of chimneys, meteorological conditions. Industrial processes like combustion, incineration also contribute in SO<sub>2</sub> generation. Industrial sources, thermal power plants and transport sectors are identified as sole contributors for SO<sub>2</sub> emissions (Rai et al. 2011). Increase in SO<sub>2</sub> emissions is also linked to increase in motor vehicle population (Rai et al. 2013).

NO<sub>2</sub> enters the atmosphere from various natural and anthropogenic sources, including lightning, action of microorganisms on nitrogen-based fertilizer, the result from chemical reaction between atmospheric N<sub>2</sub> and O<sub>2</sub> in the presence of heat to form NO which then reacts again with O<sub>2</sub> to form NO<sub>2</sub>. The rate of reaction is determined by the temperature of combustion. Thus, unlike SO<sub>2</sub>, NO<sub>2</sub> is not a component of fossil fuel, but results from a catalytic reaction of heat with atmospheric N<sub>2</sub> and O<sub>2</sub> in the combustion process. The most important and major anthropogenic source is the combustion of fossil fuel in traffic and power plants (Salam et al. 2008). Mobile sources such as automobiles contributed 58%, and stationary point sources such as industries and power plants accounted for 32% of NO<sub>x</sub> emissions in the USA (Rai et al. 2013). With the establishment of integrated industrial estate (IIE), Haridwar in the year 2000, the pollution levels has increased drastically over the period of 10 years (Joshi & Mahadev 2011). The central pollution control board (CPCB), India assigned a Comprehensive Environmental Pollution Index (CEPI) score of 51.75 to air quality, 48 to water quality and 40 to land quality and a cumulative score of

61.01 to industrial clusters in Haridwar. Ambient air quality was studied to assess impact of thermal power plant emissions in Angul District, Orissa (Nayak et al. 2012). Air quality monitoring in Haridwar district showed that total average value for the concentration of  $PM_{10}$  was higher at the sub-urban as compared to the rural sites (Tyagi et al. 2012).

Researchers in several countries studied correlations between ambient air quality and meteorological parameters (Tyagi et al. 2012; Martuzevicius et al. 2004; Jacob & Winner 2009; Maraziotis et al. 2008; Sivaramasundaram & Muthusubramanian 2010) and found that ambient air quality is impacted by meteorological conditions. Literature shows that industrial activities and vehicular pollution are the predominant source of gaseous pollution (Bhanarkar et al. 2005; Azmi et al. 2010; Rintone et al. 2015; Chauhan & Pawar 2010).

With the extension of fiscal incentives to IIE Haridwar and resulting influx of additional industries, there is a need to assess the environmental impacts of the industrial area on its immediate environs. Therefore, in the present study, an attempt has been made to study the spatial and seasonal variations of trace species ( $NO_2$ ,  $SO_2$ ) and particulate matter (SPM, RSPM) for a period of 2 years (2012–2013). During the course of study, the data for different environment types (rural/agricultural, urban residential, protected area, industrial and commercial land use) were separately extracted from monitoring stations in Haridwar, India.

## 2 Material and methods

### 2.1 Location

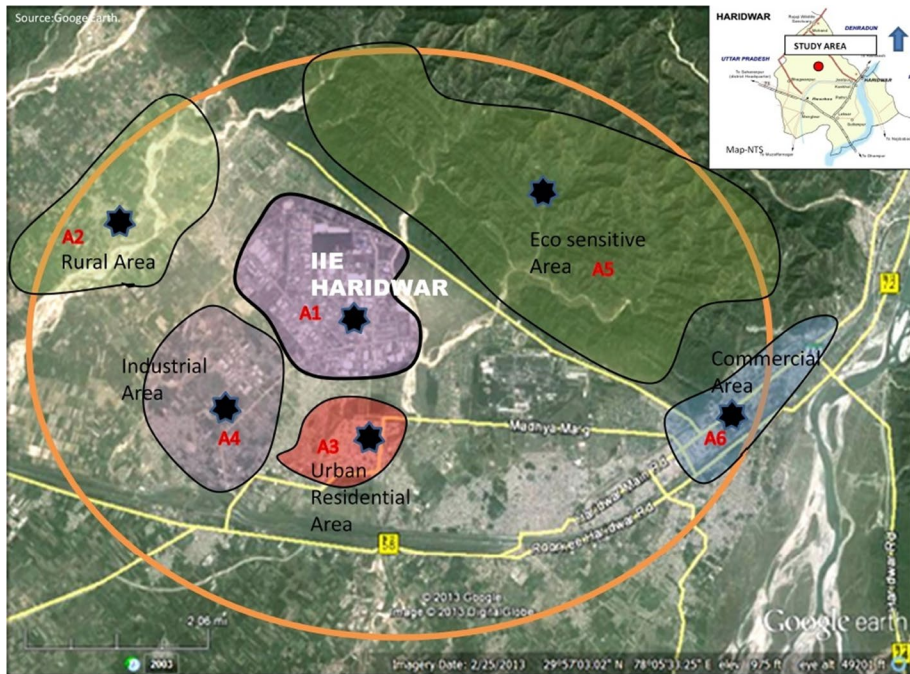
Haridwar district, covering an area of about 2360 sq km, is located in the western part of Uttarakhand state of India and came into existence on 28th December 1988. Haridwar the largest city and headquarters of Haridwar district extends from latitude  $29^{\circ} 58'$  in the north to longitude  $78^{\circ} 13'$  in the east. The city grew between the Shivalik mountain range in the north and northeast and Ganga river in the south. The height from the sea level is 249.7 m.

### 2.2 Study area

IIE Haridwar is located just 3 kms from the Delhi-Haridwar National Highway NH-58 and 52 kms from the state capital, Dehradun. The study area selected for the present study includes a geographical area of approximately 6–8 km radius and includes rural/agricultural, urban residential, protected area, industrial and commercial land uses (Fig. 1).

### 2.3 Sampling methodology

Air quality monitoring and analysis was carried out as per standard methods (BIS 1999, 2001). Ambient air quality was monitored monthly at six locations in the study area. Frequency of monitoring was 2 day continuous each month at each location in the study area. Air quality monitoring was carried out for a period of two years from January 2012 to December 2013. Air quality monitoring program was initiated in morning hours between 7.00 am and 11.00 am and was carried out using a respirable dust sampler (RDS) (Enviro-tech-APM-460) with gaseous sampling attachment in accordance with CPCB's Conceptual guidelines and common methodology for air quality monitoring, emission inventory and



**Fig. 1** Map of study area showing different landuses

source apportionment studies for Indian cities. Filter papers were folded lengthwise after monitoring, kept in plastic zip bags and transported to the laboratory. Plastic bags with ziplock containing SPM were transported to laboratory immediately after sampling and weighed.

Meteorological data were procured from pollution control research institute (PCRI) Haridwar. The meteorological station is located near Rajaji national park (RNP) in the study area. Data were monitored monthly.

## 2.4 Analytical methods

### 2.4.1 Suspended particulate matter (SPM)

*Monitoring Method:* Suspended particulate matter (SPM) was collected through a sampling train equipped with a cyclone separator, where suspended particles above 10 microns were collected in a plastic bag. Initial weight of the plastic bag with press lock was taken on the digital weighing balance and recorded. On site, the RDS was run before setting up the filter paper, and the manometer reading was checked to ensure it was at “zero”. Manometer reading was set at zero by adding DW if required. The RDS was then switched off, and the plastic bag was fitted into the cyclone cup fitted to the bottom of the cyclone hopper. The initial flow rate of manometer was recorded at the start of sampling, and final flow rate reading was taken after 24 h. Several intermediate flow meter readings were taken in between the sampling period. After 24 h, the plastic bag was removed carefully from the

cyclone cup, locked and transported to the laboratory. The plastic bag was then weighed again for final weight.

Concentration of SPM was calculated using formula.

Calculation:

$$\text{SPM } (\mu\text{g}/\text{m}^3) = [(\text{FW} - \text{IW}) \times 10^6] / V$$

where IW = Initial weight of plastic bag in gm. FW = Final weight of plastic bag in gm.  $10^6$  = Factor for converting g into  $\mu\text{g}$ .  $V$  = Volume of air sampled in  $\text{m}^3$ .

Here,

$$V = \{(Q_i + Q_f)/2\} \times T$$

where  $V$  = volume of air sampled ( $\text{m}^3$ );  $Q_i$  = Initial flow rate recorded by manometer ( $\text{m}^3/\text{min}$ );  $Q_f$  = Final flow rate recorded by manometer ( $\text{m}^3/\text{min}$ );  $T$  = sampling time in minutes.

## 2.4.2 Respirable suspended particulate matter (RSPM)

*Monitoring method:* The Glass fiber filter paper was inspected for pinholes by exposing to light in the laboratory. It was then numbered on the smooth side and equilibrated in the filter conditioning environment for 24 h. The filter paper was then weighed and stored in zip lock bag for sampling. On site, the RDS was run before setting up the filter paper, and the manometer reading was checked to ensure it was at "zero". Manometer reading was set at zero by adding DW if required. The RDS was switched off before setting up the filter paper on the filter holder with rough side up. The filter cover was fastened by securing the wing nuts, and RDS was switched on. The initial reading on the flow meter was recorded at start of sampling, and final reading was taken at end of 24 h. Several intermediate flow meter readings were taken in between the sampling period. After switching off, the RDS the filter paper was removed carefully, folded length wise, stored in a ziplock bag and transported to the laboratory. In the laboratory the used filter paper was again equilibrated for 24 h in filter conditioning environment and weighed again.

Concentration of RSPM was calculated using formula.

Calculation:

$$\text{RSPM } (\mu\text{g}/\text{m}^3) = [(\text{FW} - \text{IW}) \times 10^6] / V$$

where IW = Initial weight of filter paper in gm; FW = Final weight of Filter paper in gm;  $10^6$  = Factor for converting g into  $\mu\text{g}$ .  $V$  = Volume of air sampled in  $\text{m}^3$ .

Here,

$$V = ((Q_i + Q_f) / 2) \times T$$

where  $V$  = volume of air sampled ( $\text{m}^3$ );  $Q_i$  = Initial flow rate recorded by manometer ( $\text{m}^3/\text{min}$ );  $Q_f$  = Final flow rate recorded by manometer ( $\text{m}^3/\text{min}$ );  $T$  = sampling time in minutes.

## 2.4.3 Gaseous pollutants

**2.4.3.1 Nitrogen dioxide ( $\text{NO}_2$ )** *Monitoring method:* 50 mL of absorbing reagent was taken in impinger and placed in gaseous sampling attachment box of RDS. The manometer was set at zero mark before starting the RDS. The timer was set for 24 h. The air flow pipe was attached to the impinger, and rotameter reading was adjusted to ensure air bubbled in the

impinger at flow rate of 1.0 lpm. After 24 h, initial and final manometer and rotameter readings were taken. Absorbing reagent was transferred from impinger to dark glass sample bottle and stoppered tightly. Sample was stored in refrigerator and analyzed within 24 h.

Calculation:

$\mu\text{g NO}_2/\text{m}^3$  was computed from the slope and intercept values of standard equation.

$\mu\text{g NO}_2/\text{m}^3 = [\mu\text{g (graph factor } \times \text{ sample absorbance)} \times \text{dilution factor} \times \text{final volume of sampling solution (mL)}] / [\text{volume of air sample in m}^3 \times 0.82 \times \text{aliquot taken for sample analysis}]$ .

**2.4.3.2 Sulfur dioxide (SO<sub>2</sub>)** Monitoring method: 50 mL of absorbing reagent was taken in impinger and placed in gaseous sampling attachment box of RDS. The manometer was set at zero mark before starting the RDS. The timer was set for 24 h. The air flow pipe was attached to the impinger, and rotameter reading was adjusted to ensure air bubbled in the impinger at flow rate of 1.0 lpm. After 24 h, initial and final manometer and rotameter readings were taken. Absorbing reagent was transferred from impinger to dark glass sample bottle and stoppered tightly. Sample was stored in refrigerator and analyzed within 24 h.

Conversion of volume calculation:

$V_a = V \text{ air sampled} \times P \times (298/(273 + t))$ , where  $V_a$  = Volume of Air at 25 °C, 760 mm Hg;  $P$  = Barometric pressure (mmHg/760).

$t$  = temperature of air sampled in °C.

Calculation of SO<sub>2</sub> in  $\mu\text{g}/\text{m}^3$  in standard and sample:

$\text{SO}_2 \text{ conc} = (\text{Sample Absorbance} - \text{Bank Absorbance}) \times 10^3 \times \text{Calculation factor} \times D / (\text{Volume of Air Sample collected at } 25 \text{ }^\circ\text{C, } 760 \text{ mm Hg})$ . Where  $D = 1$  (for 30 min-1 h samples) and  $D = 10$  (for 24 h sample).  $10^3$  = conversion factor from liters to  $\text{m}^3$ .

## 2.5 National ambient air quality standards (NAAQS)

Country permissible standard limits for PM and gaseous pollutants (CPCB 2009) selected for the study have been given in Table 1.

**Table 1** National ambient air quality standards (NAAQS)

S.No	Pollutant	Time weighted average	Concentration in ambient air	
			Industrial, Residential, Rural and other area	Ecologically Sensitive area
1	Sulfur dioxide (SO <sub>2</sub> ) in $\mu\text{g}/\text{m}^3$	24 h*	80	80
2	Nitrogen dioxide (NO <sub>2</sub> ) in $\mu\text{g}/\text{m}^3$	24 h*	80	80
3	Particulate matter (size less than 10micron) or PM10 in $\mu\text{g}/\text{m}^3$	24 h*	100	100
4	Ozone (O <sub>3</sub> ) in $\mu\text{g}/\text{m}^3$	1 h*	180	180
5	Nickel (Ni) in $\mu\text{g}/\text{m}^3$	Annual	20	20

\*24 hourly or 8 hourly or 01 hourly, monitored values, as applicable, shall be complied with 98% of the time in a year, 2% of the time they may exceed the limit but not on two consecutive days of monitoring

## 2.6 Air pollution index (API)

API is the aggregated ratio of pollutant concentration in ambient air to permissible standard limit of pollutant in ambient air. API is an empirical number used by several researchers (Joshi & Mahadev, 2011; Chauhan, 2010b, 2010a; Joshi & Bora, 2011) to understand the cumulative impact of pollutants. API has been calculated for three criteria pollutants RSPM, SO<sub>2</sub> and NO<sub>2</sub> using following formula.

$$\text{API} = 1/4 \times \left( \frac{\text{RSPM}}{\text{RSPM}(s)} + \frac{\text{SO}_2}{\text{SO}_2(s)} + \frac{\text{NO}_2}{\text{NO}_2(s)} \right) \times 100$$

Where RSPM=pollutant concentration in ambient air; RSPM (s)=permissible standard limit for pollutant in ambient air; SO<sub>2</sub>=pollutant concentration in ambient air; SO<sub>2</sub>(s)=permissible standard limit for pollutant in ambient air; NO<sub>2</sub>=pollutant concentration in ambient air; NO<sub>2</sub>(s)=permissible standard limit for pollutant in ambient air.

The pollutant with the highest API number became the criteria pollutant for the location. The higher the API value, greater is adverse impact of pollution and greater the damage to health. The API scale has been divided into five categories based on impact on human health (Table 2).

## 2.7 Statistical methods and software's used

Result database has been evaluated using Ms Excel and SPSS. Graphing and Mapping have been done with Ms Excel, SPSS and Surfer 11. The average mean values are means of two values obtained from two days of monitoring each month. Paired t-test has been computed to test the difference in parameter means over years of study. Welch ANOVA has been used to assess spatial and seasonal variation in parameter concentrations. Games Howell post hoc test has been applied where required to further test significant results of one-way ANOVA. ANOVA test tells us whether we have an overall difference between the groups, but it does not tell us which specific groups differed—the post hoc tests do. Because post hoc tests are run to confirm where the differences occurred between groups, they should only be run when we have shown an overall statistically significant difference in group means (i.e., a statistically significant one-way ANOVA result). Post hoc tests attempts to control the experiment wise error rate (usually alpha=0.05) in the same manner that the one-way ANOVA is used instead of multiple t-tests. Pearson Correlation analysis has been carried out to understand the relationship between parameters of study. The Shapiro–Wilk Test is appropriate for small sample sizes (<50 samples), but can also handle sample sizes as large as 2000. For this reason, the Shapiro–Wilk test has been used as numerical means of assessing normality. As distribution of parameters has been found be normal for all

**Table 2** API index values and interpretation

Index value	Interpretation of index value
0–25	Clean air
26–50	Light air pollution
51–75	Moderate air pollution
76–100	Heavy air pollution
> 100	Severe air pollution

parameters except RSPM ( $p > 0.01$ ), the data are assumed to be normal. If the test concludes the correlation coefficient is significantly different from zero, we say the correlation coefficient is significant. While if the test concludes the correlation coefficient value is not significantly different from zero (it is close to zero), we say the correlation coefficient is not significant.

### 3 Results

Results of ambient air quality and meteorological conditions during study period are given in Table 3 and Figs. 2 and 3, Table 4 shows the spatial variation in ambient air quality during the study period while Tables 5 and 6 shows seasonal variation and one-way ANOVA in ambient air quality, respectively.

*Suspended particulate matter (SPM)*: During the study period SPM in the study area ranges from a minimum of  $6.04 \mu\text{g}/\text{m}^3$  at RNP in August 2012 to a maximum of  $442.47 \mu\text{g}/\text{m}^3$  at IIE Haridwar in Feb 2013 (Fig. 2 & Fig. 3). The average SPM in the study area was found as  $86.92 \mu\text{g}/\text{m}^3$ . Average SPM levels were observed in decreasing order of landuse during study period i.e., IIE Haridwar > Bahadradab old industrial area > Commercial area > Aneki > Shivalik Nagar > RNP (Tables 4 and 5). One-way ANOVA shows statistically significant difference in SPM levels between different landuses  $F(5,138) = 7.824$ ,  $p = 1$  (Table 6).

*Respirable suspended particulate matter (RSPM)*: During the study period, RSPM in study area ranges from minimum of  $6.04 \mu\text{g}/\text{m}^3$  at RNP in September 2012 and maximum of  $241.51 \mu\text{g}/\text{m}^3$  at IIE Haridwar in October 2012. The average RSPM during the study period was found as  $81.04 \mu\text{g}/\text{m}^3$ . During study period, average RSPM levels were observed in decreasing order of landuse i.e., IIE Haridwar > Bahadradab old industrial area > Aneki > Commercial area > Shivalik Nagar > RNP.

*Sulfur dioxide ( $\text{SO}_2$ )*: During the study period,  $\text{SO}_2$  ranged between  $1.33 \mu\text{g}/\text{m}^3$  at RNP during August 2013 and  $78.16 \mu\text{g}/\text{m}^3$  at Bahadradab old industrial area during July 2013. The average concentration of  $\text{SO}_2$  in ambient air of the study area was found as  $23.23 \mu\text{g}/\text{m}^3$ . During study period, average  $\text{SO}_2$  levels was observed in decreasing order by landuse i.e., Bahadradab > IIE Haridwar > Aneki > commercial area > Shivalik Nagar > RNP. Studies have reported high  $\text{SO}_2$  levels in industrial areas as compared to other land use areas (Joshi & Mahadev 2011; Joshi & Bora 2011; Kabir & Madugu 2010; Jeong et al. 2011; Jha et al. 2010).

*Nitrogen dioxide ( $\text{NO}_2$ )*:  $\text{NO}_2$  ranges from a minimum of  $1.41 \mu\text{g}/\text{m}^3$  at Aneki rural area during August and September 2012 and  $73.43 \mu\text{g}/\text{m}^3$  at Railway station road (commercial area) during July 2013. Average  $\text{NO}_2$  level during the study period was found as  $24.73 \mu\text{g}/\text{m}^3$ . Average  $\text{NO}_2$  levels during study period were observed in decreasing order of landuse i.e., IIE Haridwar > Commercial area > Bahadradab old industrial area > Shivalik Nagar > Aneki > RNP.

Correlation between PM (SPM and RSPM), gaseous pollutants ( $\text{SO}_2$  and  $\text{NO}_2$ ) and meteorological parameters (Table 7).

Significant positive correlations at (0.800,  $p < 0.01$ ) have been found between SPM and RSPM during the study period which indicates that sources of emission of SPM and RSPM are similar during study period. Similar findings have been reported in many studies (Bamniya et al. 2011; Maraziotis et al. 2008). Strong positive correlation (0.775,  $p < 0.01$ ) exists between gaseous pollutants  $\text{SO}_2$  and  $\text{NO}_2$  in the study area indicating sources of emission



**Table 3** Ambient air quality and meteorological conditions during study period (Year 2012 and Year 2013)

S.No	Month	SPM ( $\mu\text{g}/\text{m}^3$ )		RSPM ( $\mu\text{g}/\text{m}^3$ )		SO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )		NO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
1	Jan	143.46	99.35	119.07	49.79	19.54	11.14	16.81	10.12
2	Feb	126.76	135.55	112.57	51.88	29.14	7.60	42.40	16.87
3	March	88.05	61.87	66.93	38.42	28.58	17.99	50.52	14.93
4	April	92.10	93.77	69.52	38.23	28.94	8.85	21.31	16.36
5	May	120.45	136.94	90.24	44.76	26.26	18.75	32.29	9.43
6	June	50.99	32.84	95.78	55.97	29.25	9.07	40.10	20.42
7	July	48.70	35.25	67.02	41.82	49.50	20.57	38.92	19.75
8	Aug	22.92	29.25	40.62	33.15	5.33	4.63	3.44	2.40
9	Sept	33.72	28.65	26.56	32.04	3.74	1.83	3.70	2.24
10	Oct	77.73	49.96	96.37	64.04	18.26	10.18	17.49	13.06
11	Nov	117.97	61.60	89.11	40.81	25.04	14.03	14.65	10.93
12	Dec	120.19	73.97	98.68	47.82	15.19	6.65	15.05	9.42
13	Avg	86.92	40.19	81.04	27.77	23.23	12.20	24.73	15.64
S.No	Month	API	Temp ( $^{\circ}\text{C}$ )		RH (%)		WS (m/s)		WD
			Mean	SD	Mean	SD	Mean	SD	
1	Jan	54.84	13.29	3.97	80.24	11.87	1.74	0.80	SW
2	Feb	67.33	19.63	3.92	56.70	16.59	2.86	1.35	SW
3	March	55.02	27.51	4.04	36.32	14.11	4.66	1.79	SW
4	April	43.82	30.45	6.16	36.82	20.34	4.54	2.55	SW
5	May	54.48	37.97	2.67	23.96	10.38	5.34	1.80	SW
6	June	60.82	35.26	5.00	48.77	16.36	3.92	2.04	SW
7	July	59.18	31.90	4.00	72.35	13.89	2.47	1.57	SE
8	Aug	17.20	30.19	3.15	75.99	11.65	2.40	1.43	SE
9	Sept	11.95	30.00	1.86	72.03	8.72	3.04	0.51	SW
10	Oct	47.02	29.56	3.24	52.94	11.42	1.82	0.73	SW
11	Nov	46.24	22.23	4.61	56.41	12.06	2.09	0.65	SW
12	Dec	45.49	18.29	3.79	65.05	10.02	3.09	2.21	SW
13	Avg	46.95	27.19	7.33	56.47	17.67	3.16	1.19	SW

Note: API=Air pollution index, RH=relative humidity, WS=wind speed, WD=predominant wind direction, sd=standard deviation

of the gases are similar. Insignificant but positive correlation exists between RSPM and gaseous pollutants SO<sub>2</sub> and NO<sub>2</sub> during study period. Negative correlation has been found between RH and SPM during the study period, and SPM levels are lowest during monsoon. Increase in humidity during monsoon results in precipitation of particulate matter and gases from the atmosphere and hence during monsoon SPM levels are low in the study area. PM is efficiently scavenged by precipitation, and this is its main atmospheric sink (Jacob & Winner 2009). During the high humidity episodes, the particle hygroscopic growth and condensation likely result in an increase of the coarse aerosol fraction or SPM (Martuzevicius et al. 2004). Negative correlation between RH and SPM /RSPM has been reported by several researchers (Bhaskar 2010; Martuzevicius et al. 2004; Jacob & Winner 2009).

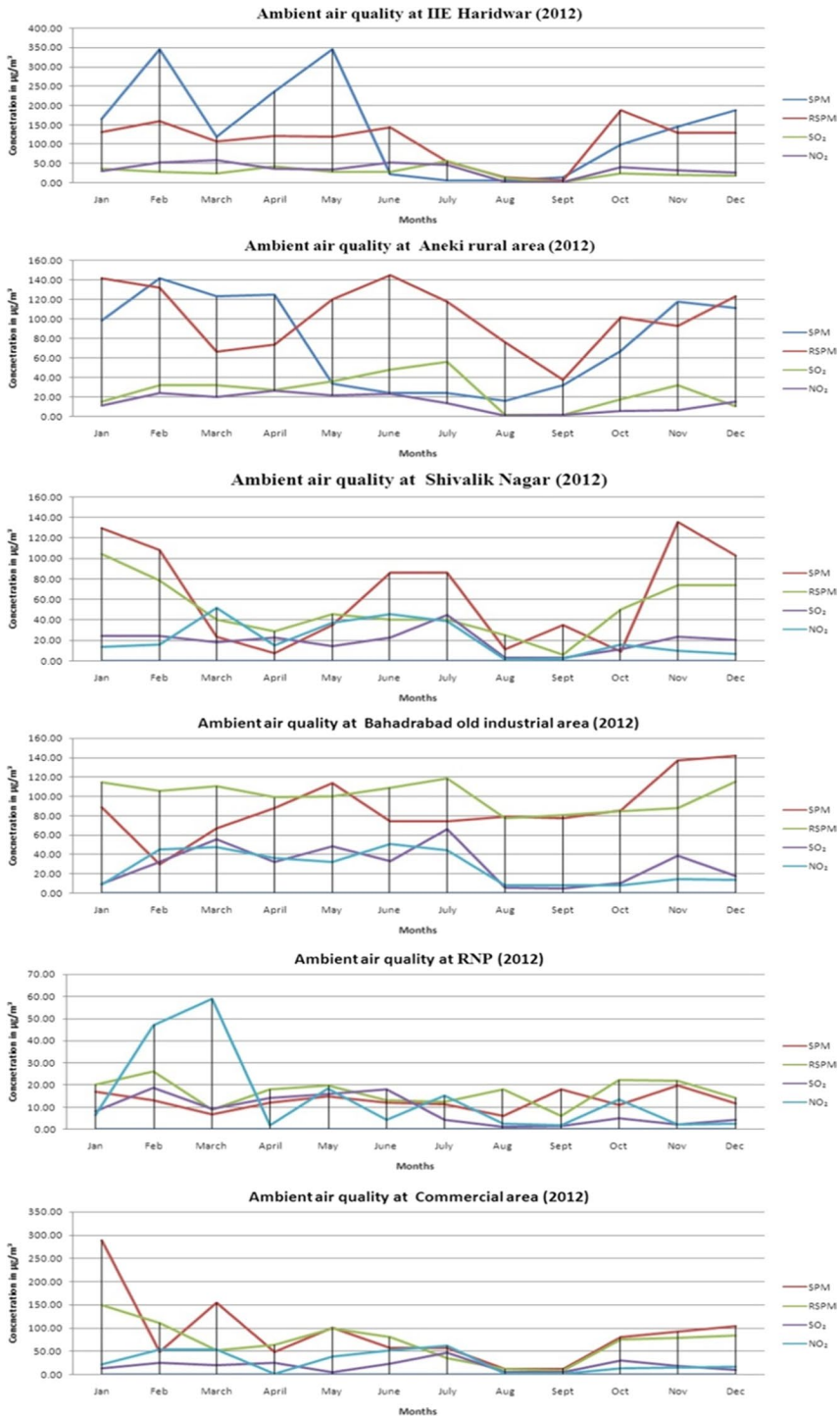


Fig. 2 Ambient air quality in study area in Year 2012

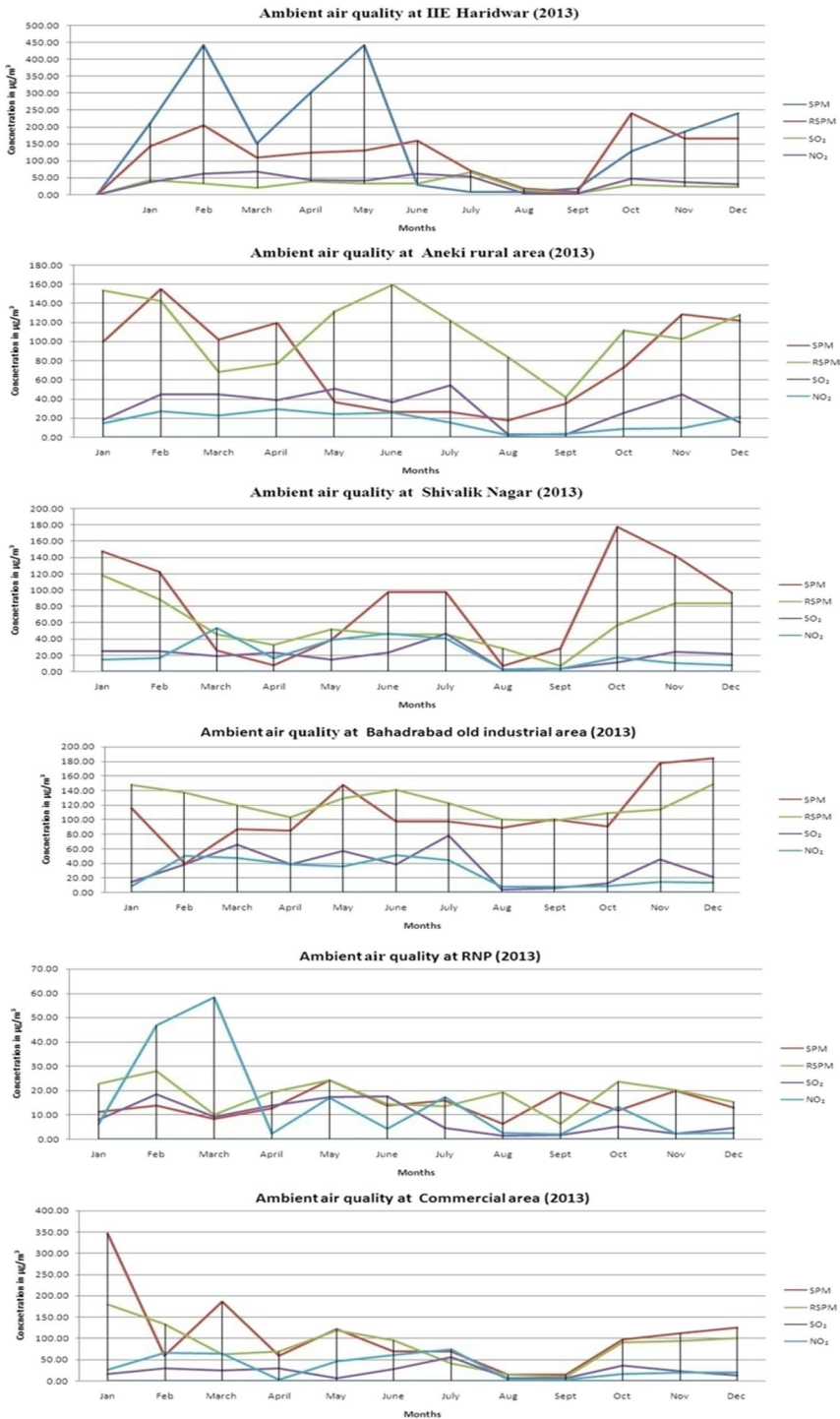


Fig. 3 Ambient air quality in study area in Year 2013

**Table 4** Spatial variation in ambient air quality during study period (Year 2012 and Year 2013)

S.No	Landuse	Month	SPM ( $\mu\text{g}/\text{m}^3$ )		RSPM ( $\mu\text{g}/\text{m}^3$ )		SO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )		NO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )		API
			Mean	SD	Mean	SD	Mean	SD	Mean	SD	
1	IIE Haridwar	Min	7.14	1.89	7.14	2.22	2.45	2.20	3.45	2.10	6.66
		Max	442.47	6.10	241.51	6.10	66.26	11.33	68.92	6.62	112.26
		Avg	161.44	138.45	119.31	62.48	28.98	14.42	38.66	19.12	72.92
2	Aneki rural area	Min	15.87	2.20	38.10	3.12	1.80	1.44	1.41	1.89	16.36
		Max	155.29	11.33	159.64	6.62	56.02	6.20	29.43	6.10	79.14
		Avg	77.27	47.05	106.52	34.37	28.71	17.40	15.82	9.12	56.98
3	Shivalik Nagar	Min	7.20	1.90	6.31	2.10	3.00	1.89	1.73	3.12	5.55
		Max	177.93	9.72	118.56	6.62	46.36	6.10	53.66	6.62	56.14
		Avg	73.40	56.30	54.25	28.69	19.84	11.04	21.98	17.20	37.00
4	Bahadradabad old industrial area	Min	30.18	1.56	77.16	2.20	4.34	1.78	7.68	1.56	38.58
		Max	184.21	8.79	149.12	9.88	78.16	5.67	51.63	5.66	92.12
		Avg	98.62	37.23	111.39	19.93	32.60	21.71	27.16	17.84	67.16
5	RNP	Min	6.04	1.10	6.04	2.20	1.33	1.78	1.78	1.30	3.45
		Max	24.18	8.79	27.93	11.33	18.79	9.76	59.03	6.10	36.52
		Avg	13.54	4.55	17.44	6.02	8.67	6.42	14.53	18.55	15.69
6	Commercial area	Min	11.93	2.20	8.22	1.89	4.91	1.10	2.10	1.30	6.94
		Max	346.15	11.33	179.49	6.10	55.75	8.79	73.43	4.20	83.66
		Avg	97.25	81.32	77.33	44.41	20.59	13.33	30.20	24.34	51.10
7	Study area	Min	6.04	1.10	6.04	1.89	1.33	1.10	1.41	1.30	3.45
		Max	442.47	138.45	241.51	62.48	78.16	21.71	73.43	24.34	112.26
		Avg	86.92	47.87	81.04	39.53	23.23	8.73	24.73	9.17	50.14

Note: API= Air pollution index, SD=standard deviation

During study period, negative and insignificant correlation has been found between wind speed and RSPM ( $r = -0.145$ ). This indicates that wind (predominant direction-south-west) is causing dispersion of RSPM fraction. Negative correlation between wind speed, RH and RSPM has been attributed (Martuzevicius et al. 2004) to dispersion of particulates and greater release of dust particles due to resuspension by high wind speed. However, positive but insignificant correlation between wind direction and SPM indicates that resuspension of dust in the study area at ground level is taking place especially in rural area; causing high SPM and RSPM levels during summer. Emissions of gaseous pollutants are dependent upon sunlight and temperature. Negative correlation between RSPM and temp indicates that low temperatures favor accumulation of particulate matter in ambient air. This has been reported in literature (Tyagi et al. 2012; Maraziotis et al. 2008). However, other researchers have found positive correlations between RSPM and Temperature (Sivaramasundaram & Muthusubramanian 2010; Martuzevicius et al. 2004).

Air pollution index (API) during study period.

API during study period ranges from a minimum of 3.45 indicating clean air at RNP in Sept 2012 to a maximum of 112.26 at IIE Haridwar in October 2013. The average API during study period is 50.14 which indicate light air pollution. Seasonal variation shows that highest API occurs during summer (54.12 = moderate air pollution)

**Table 5** Seasonal variation in ambient air quality during study period (Year 2012 and Year 2013)

S.No	Landuse	Month	SPM ( $\mu\text{g}/\text{m}^3$ )		RSPM ( $\mu\text{g}/\text{m}^3$ )		SO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )		NO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )		API
			Mean	SD	Mean	SD	Mean	SD	Mean	SD	
1	IIE Haridwar	Summer	206.50	151.37	127.80	17.44	31.48	6.99	50.73	12.08	66.96
		Monsoon	36.84	48.80	75.43	90.19	26.71	23.65	25.75	23.54	66.09
		Winter	240.97	101.97	154.71	25.93	28.74	8.39	39.50	11.96	65.16
2	Aneki rural area	Summer	73.73	47.16	105.45	37.90	39.17	7.91	24.37	2.92	53.62
		Monsoon	36.35	21.81	86.79	32.98	20.32	23.24	6.59	5.45	53.56
		Winter	121.73	19.65	127.32	20.40	26.63	13.47	16.50	7.33	51.41
3	Shivalik Nagar	Summer	40.33	33.86	41.75	7.55	19.90	3.57	38.37	14.89	32.81
		Monsoon	56.55	63.42	32.67	19.12	15.91	18.71	15.41	16.43	33.28
		Winter	123.32	58.03	88.33	15.63	23.71	1.62	12.16	3.71	32.61
4	Bahadradab old indus-trial area	Summer	94.93	25.35	114.02	15.21	46.19	12.27	42.75	7.63	64.50
		Monsoon	86.62	9.28	98.77	17.24	23.91	30.11	17.40	16.85	64.31
		Winter	114.31	58.00	121.39	21.73	27.69	12.90	21.33	16.68	62.53
5	RNP	Summer	13.12	5.26	16.01	5.22	14.50	3.54	20.66	24.36	13.52
		Monsoon	12.47	4.93	15.20	6.74	3.14	1.84	8.48	6.89	13.55
		Winter	15.04	3.48	21.10	4.80	8.37	6.76	14.47	20.12	12.67
6	Commercial area	Summer	99.84	50.62	80.01	22.57	19.89	9.43	39.44	24.08	44.78
		Monsoon	44.73	35.36	35.67	31.68	23.65	20.77	21.73	28.85	45.54
		Winter	147.18	109.12	116.30	35.20	18.24	6.80	29.42	18.80	44.02
7	Study area	Summer	88.07	66.80	80.84	43.92	28.52	12.49	36.05	11.41	46.03
		Monsoon	45.59	24.75	57.42	33.96	18.94	8.58	15.89	7.42	46.05
		Winter	127.09	72.30	104.86	46.22	22.23	7.77	22.23	10.44	44.73

Note: API = Air pollution index, SD = standard deviation

**Table 6** One-way ANOVA to assess spatial and seasonal variation in air quality parameters during study period (Year 2012 and Year 2013)

S.No	Parameter	Spatial variation					Seasonal variation						
		Sum of squares	df	Within groups	Mean square	F	Sig	Sum of squares	Df	Within groups	Mean square	F	Sig
1	SPM	279,870.814	5	138	55,974.163	7.824	.000	121,983.040	2	141	60,991.520	7.509	.001
2	RSPM	163,585.907	5	138	32,717.181	21.093	.000	25,107.923	2	141	12,553.962	5.021	.008
3	SO <sub>2</sub>	7763.200	5	138	1552.640	6.618	.000	1486.960	2	141	743.480	2.712	.070
4	NO <sub>2</sub>	8509.971	5	138	1701.994	4.781	.000	9691.901	2	141	4845.950	14.251	.000

**Table 7** Correlations between ambient air quality and meteorological parameters during study period (2012 and 2013)

	SPM	RSPM	SO <sub>2</sub>	NO <sub>2</sub>	API	Temp	RH	WS
SPM	1							
RSPM	.800**	1						
SO <sub>2</sub>	.178	.331	1					
NO <sub>2</sub>	.194	.350	.775**	1				
API	.574	.794**	.790**	.819**	1			
Temp	-.615*	-.488	.177	.192	-.142	1		
RH	-.250	-.140	-.311	-.518	-.371	-.507	1	
WS	.039	-.145	.210	.479	.167	.557	-.827**	1

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

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

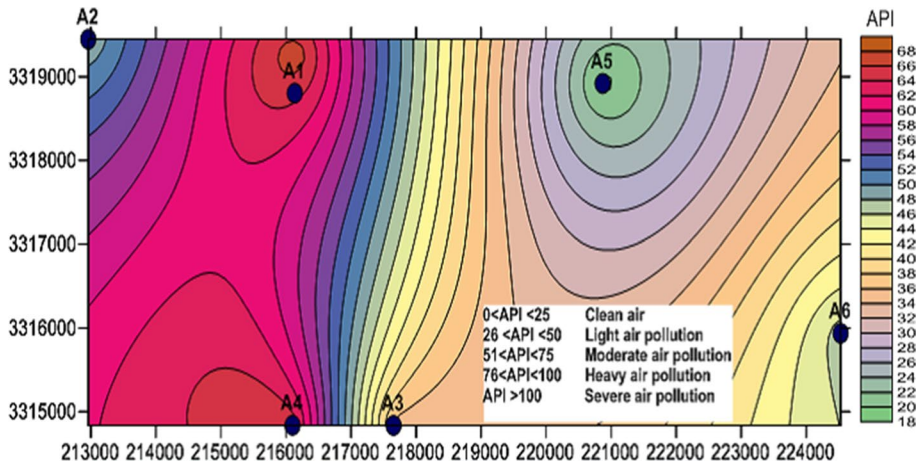
followed by winter (50.13 = moderate air pollution) and monsoon (36.76 = light air pollution), respectively. Average API levels during study period in decreasing order of landuse are IIE Haridwar (moderate pollution) > Bahadradab old industrial area (moderate pollution) > Aneki rural area (light pollution) > commercial area (light pollution) > Shivalik Nagar (light pollution) > RNP (clean air). Air pollution index (API) for criteria pollutants, SPM, RSPM, SO<sub>2</sub> and NO<sub>2</sub> shows that average ambient air quality in the study area can be categorized as clean, only during August and September, when API ranges between 0 and 25. API ranges between 50 and 75 in January, April, May, June, July, October, November and December indicating moderate air pollution in study area. During February and May air quality in the study area shows heavy pollution with API levels exceeding 75. API results indicate that air quality is a function of anthropogenic activities and does not show seasonal variation.

Air quality condition is the consequence of many determinants (Nematchoua et al. 2015; Papamanolis 2015; Root et al. 2015; Schucht et al. 2015). The impact of socio-economic factors on the API level has been discussed (Zhao et al. 2012). Zheng et al. (2015) have studied the impact of provincial energy and environmental policies on API level. The relationship between each pollutant (NO<sub>2</sub>, PM<sub>10</sub>, SO<sub>2</sub>) and urban structure is also conducted (Cárdenas Rodríguez et al. 2016).

Seasonal trend of API is similar to seasonal variation in API found in Dehradun city (Chauhan 2010b, Rai & Panda 2015). Figure 4 shows spatial variation in Air Pollution index (API) during study period. A1, A2, A3, A4, A5 and A6 depict air quality monitoring locations at IIE Haridwar, Aneki rural area, Shivalik Nagar, Bahadradab old industrial area, RNP and Commercial area, respectively.

*T* Test to assess variation in air quality parameters for study years (2012 and 2013).

*T* test has been applied to assess variation between parameter concentrations over study years 2012 and 2013. Results of test for two sample means are significant for all parameters (SPM, RSPM, SO<sub>2</sub>, NO<sub>2</sub> at ( $p < 0.05$ ) except for meteorological parameters Temp, RH and wind speed. Results of *T* test (Table 8) indicate that variation (increase) in PM and gaseous pollutants could be attributed to increased pollution emissions and micro level dust resuspension due to vehicular and anthropogenic activities in the study area.



**Fig. 4** Spatial variation in air pollution index (API) during study period

**Table 8** *T*-Test of air quality parameters between study years

S.No	Parameter	Mean	Variance	Df	<i>t</i> Stat	<i>P</i> ( <i>T</i> < = <i>t</i> )	<i>t</i> Critical two-tail
1	SPM	101.088	7628.477	47.000	-6.728	0.000	2.012
2	RSPM	88.087	1871.343	47.000	-7.872	0.000	2.012
3	SO <sub>2</sub>	25.214	255.025	47.000	-5.609	0.000	2.012
4	NO <sub>2</sub>	29.547	472.109	47.000	-3.119	0.003	2.012

### 3.1 Discussion

SPM levels are found minimum at RNP, which is a protected area with restricted vehicular entry. Low SPM levels can also be attributed to the monitoring location which is a raised secluded spot covered with vegetation in the park. High SPM levels in industrial and commercial/urban areas can be attributed to industrial emissions and dust resuspension from vehicular traffic and pollution (Chauhan & Pawar 2010; Joshi & Mahadev 2011; Tyagi et al. 2012; Khanna et al. 2013). Average SPM levels in the study area during summer, monsoon and winter was observed as 88.07  $\mu\text{g}/\text{m}^3$ , 45.59  $\mu\text{g}/\text{m}^3$  and 127.09  $\mu\text{g}/\text{m}^3$ , respectively (Table 5). SPM levels were found lowest during monsoon season this may be due to high humidity and rainfall which ultimately result in precipitation of the dust fraction from the atmosphere. One-way ANOVA shows statistically significant differences in SPM levels between the different seasons,  $F(2, 141) = 7.509$ ,  $p = 0.001$ . High SPM levels in ambient air during the winter have been reported by many researchers (Salam et al. 2008; Bhaskar 2010; Nayak et al. 2012). Average SPM levels exceed WHO standard limit of 120  $\mu\text{g}/\text{m}^3$  at IIE Haridwar during the study period.

Studies have attributed high RSPM levels in industrial and urban areas to pollutant emissions, automobile exhaust, traffic and dust resuspension (Rai et al. 2013; Radhapiya et al. 2012; Rao et al. 2009; Singh & Agrawal 2005; Singh et al. 2008). Average RSPM levels exceed NAAQS limit of 80  $\mu\text{g}/\text{m}^3$  at IIE Haridwar, Bahadradabad old industrial area and



Aneki rural area. Average RSPM levels were found to be maximum in winter as  $104.86 \mu\text{g}/\text{m}^3$  followed by  $80.84 \mu\text{g}/\text{m}^3$  in summer and  $57.42 \mu\text{g}/\text{m}^3$  in monsoon, respectively. It is evident that in winter season temperature inversion occurs easily in mornings and evenings which suppresses the diffusion of pollutants in to the atmosphere; meanwhile in summer season fuel is consumed in large quantities, which makes bad situation of air pollution. Similarly the spring season is the season of frequent dust storms that affect many cities through long-range transport of pollutants. Hence an increase in dust eventually affects air quality in the spring season. High levels of particulate matter during winter can be attributed to low wind velocity, low temperature and relatively stable atmospheric conditions with a low dispersion rate (Haritash & Kaushik 2007). High SPM and RSPM levels in winter have been reported in several studies (Miranda et al. 2012; Chauhan & Pawar 2010; Mishra et al. 2013).

Development of industry consumes amount of energy and emits many pollutants (e.g., NO<sub>x</sub>, PM<sub>2.5</sub>) into the atmosphere (Zhang et al. 2015c). Haridwar has experienced rapid urbanization and modernization over the past two decades. It is estimated that approximately 90% of SO<sub>2</sub> and 80% of particulate matter result from coal burning by organizations such as industries. Statistics show that the SO<sub>2</sub> emissions for the same quantity of heat generated by burning coal and gas is 119:1, and the emission of particulate matter from those fuel sources is 615:1 (Zhang et al. 2011). High SO<sub>2</sub> levels in Aneki rural area were found during the course of study this may be due to burning of biofuels, wood and coal for cooking and the proximity of the rural area to both industrial areas. Average SO<sub>2</sub> levels were found to be highest during summer at  $28.52 \mu\text{g}/\text{m}^3$  followed by  $28.23 \mu\text{g}/\text{m}^3$  in winter and  $18.94 \mu\text{g}/\text{m}^3$  in monsoon, respectively. Gaseous pollutants (SO<sub>x</sub> and NO<sub>x</sub>) being water-soluble get washed away from atmosphere due to rainfall and high humidity, hence the concentration of these gases was found minimum in rainy season. Contrary to this, in the winter season, relative humidity of the atmosphere being the minimum and atmosphere being sluggish due to the stability of temperature; distribution of gaseous pollutants is not so prominent (Jha et al. 2010). The average SO<sub>2</sub> level has been found to be within NAAQ limit of  $80 \mu\text{g}/\text{m}^3$  at all the landuse areas during the study period.

As per Oiamo et al. (2012) traffic, dwelling density and railway yard and railway lines have been reported to cause variability in NO<sub>2</sub> levels similar observations were reported during the present study. In the present study, the commercial area has the city bus stand and railway station all of which generate high density of vehicular traffic resulting in high NO<sub>2</sub> levels. However, NO<sub>2</sub> levels have been found to be within NAAQ limits of  $60 \mu\text{g}/\text{m}^3$  at all landuse areas during the study period. One-way ANOVA shows statistically significant differences in NO<sub>2</sub> levels between different seasons,  $F(2, 141) = 14.251, p = 0.000$ . Games Howell post hoc test reveals that there is statistically significant ( $p = 0.00$ ) increase in NO<sub>2</sub> levels from monsoon to summer and from winter to summer ( $p = 0.000$ ). SO<sub>2</sub> and NO<sub>2</sub> levels have been found to be within limits in Haridwar region by several researchers (Joshi & Mahadev 2011; Joshi & Swami 2007; Chauhan & Pawar 2010).

### 3.2 Conclusion

The results of the above study can be summarized as 1. Increase in concentrations of SM over the year at industrial site, this may be attributed to industrial emissions and dust resuspension from vehicular traffic. SPM levels were found lowest during monsoon season as high humidity and rainfall result in precipitation of the dust fraction from the atmosphere. 2. Average RSPM levels are found to be maximum in winter and can be attributed to low

wind velocity, low temperature. 3. Average SO<sub>2</sub> and NO<sub>2</sub> levels have been found to be within NAAQ limits 4. Air pollution index (API) for criteria pollutants, SPM, RSPM, SO<sub>2</sub> and NO<sub>2</sub> shows that average ambient air quality in the study area can be categorized as clean, only during August and September, when API ranges between 0–25. Hence from the above study it may be concluded that ambient air quality in the study area can be categorized as clean in Rajaji National Park (RNP), while in both industrial areas air quality can be said to be severely polluted. Study shows there is light air pollution in Shivalik Nagar, Aneki rural area and commercial area. Study also shows that meteorological parameters have an impact on pollutant concentrations during study period. High temperatures during summer are causing high NO<sub>2</sub> levels in ambient air. RH lowers pollutant concentrations, whereas wind speed is causing dispersion of RSPM fraction of ambient air.

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## Compliance with ethical standards

**Conflict of interest** The authors have no conflict of interest arising from the direct application of this research work.

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