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ABSTRACT

A key challenge in controlling Delhi's air quality is a lack of clear understanding of the impacts of emissions from the surrounding National Capital Region (NCR). Our objectives are to understand the limitations of publicly available data, its utility to determine pollution sources across Delhi-NCR and establish seasonal profiles of chemically active trace gases. We obtained the spatiotemporal characteristics of daily-averaged particulate matter (PM_{10} and $PM_{2.5}$) and trace gases (NO_x, O_3, SO_2, and CO) within a network of 12 air quality monitoring stations located over $2000\,km^2$ across Delhi-NCR from January 2014 to December 2017. The highest concentrations of pollutants, except O_3, were found at Anand Vihar compared with lowest at Panchkula. A high homogeneity in $PM_{2.5}$ was observed among Delhi sites as opposed to a high spatial divergence between Delhi and NCR sites. The bivariate polar plots and k-means clustering showed that $PM_{2.5}$ and PM_{10} concentrations are dominated by local sources for all monitoring sites across Delhi-NCR. A consequence of the dominance of local source contributions to measured concentrations, except to one site remote from Delhi, is that it is not possible to evaluate the influence of regional pollution transport upon PM concentrations measured at sites within Delhi and the NCR from concentration measurements alone.

1. Introduction

Air pollution remains one of the major threats to human health and wellbeing in cities (WHO, 2016). Ambient air pollution was estimated to cause nearly 4.2 million premature deaths worldwide in 2016 (WHO, 2016). Given the rapid rates of growth and urbanisation in Indian cities, air pollution is increasingly becoming a critical threat to the environment, human health, and to the quality of life among the urban population in India (Kumar et al., 2013). Recent studies have revealed that the public health importance of improving poor air quality in India, where approximately 600,000 premature deaths annually are associated with ambient air pollution (Ghude et al., 2016; Lelieveld, Evans, Fnais, Giannadaki, & Pozzer, 2015; WHO, 2016). Some of the highest levels of pollutants in ambient air globally are found in some Indian cities like Delhi (Kumar et al., 2015). Due to the growth of population and expansion of transportation and city infrastructure, Delhi is one of the most polluted cities in the world (Kumar, Gulia, Harrison, & Khare, 2017; WHO, 2016). Particulate matter exposure is linked with an average lost life expectancy of three years across India and approximately six years in Delhi city (Ghude et al., 2016).

Over the last two decades, several policies have been implemented to tackle air pollution in Delhi, including shutting down 1328 factories responsible for hazardous emissions, renewing the public transport system by introducing the Delhi Metro, reduction of sulphur content in diesel and conversion of fleet transport vehicles to compressed natural gas (Chowdhury et al., 2017; Goel & Pant, 2016; Kumar et al., 2017;

Table 1

Summary of relevant past studies for assessing secondary data of particulate matter and gaseous pollutants in Delhi and its NCR.

Pollutant type	Data source	Time period	City	Study
$PM_{2.5}$ and PM_{10}	CPCB and DPCC	Jan and April 2016	Delhi	Kumar et al. (2017)
O_3, NO, NO_2, CO, $PM_{2.5}$ and PM_{10}	SAFAR	October 2010–December 2014	Delhi-NCR	Peshin et al. (2017)
NOx, O_3, and CO	SAFAR	Jan-Dec 2014	Delhi-NCR	Tyagi et al. (2016)
$PM_{2.5}$ and PM_{10}	CPCB	2011–2013	Delhi	Tiwari, Hopke et al. (2015)
PM_{10}, SPM, SO_2, and NO_2	CPCB	–	Delhi	Sharma et al. (2013)
PM_{10}, $PM_{2.5}$, SO_2, O_3, CO, and NO_2	CPCB	2006-2010	Delhi	Guttikunda and Gurjar (2012)

Narain & Krupnick, 2007). In 2016, an odd-even traffic intervention, allowing odd- and even-numbered cars as classified by their last digit of the number plates to be used on alternate days, was implemented in Delhi twice (winter and summer schemes) for fifteen days, from January to April without clear concentration reduction benefits (Kumar et al., 2017) that is again being repeated in November 2019. However, very few policies have been implemented to reduce pollution in the NCR region, particularly Haryana and Uttar Pradesh (Chowdhury et al., 2019). Despite the implementation of these policies in Delhi, ambient $PM_{2.5}$ concentrations in Delhi and its NCR region remain many-fold higher than annual NAAQS (Chowdhury et al., 2019). Multiple factors elucidate the lack of success of those policies, owing to concrete interventions but also due to its geographic location (land-locked) and the prevailing meteorological conditions (Kumar et al., 2015). In addition, dust emissions during the summer, transport of pollution emitted from open biomass burning in upwind rural regions during the crop burning season, and those from the brick kilns in the surroundings of Delhi throughout the year (Cusworth et al., 2018) add to local pollutant sources such as transportation and traffic emissions, construction activities and resuspension dust emission, diesel generators, power plants, industries and roadside biomass burning (Kumar et al., 2013, 2015; Nagpure, Ramaswami, & Russell, 2015). In addition, there is a need to reduce air pollution and a planned sustainable system in cities such as Delhi. Some of the primary initiatives towards reaching a more sustainable city and low pollution level include, include limiting car use, improving public transport services and encouraging their use, increasing the opportunities for walking and cycling, controlling roadside and open biomass burning within Delhi and its NCR region, and the implementation of policies at a larger spatial scale considering the land-locked nature of the city covering Delhi and NCR region (Chowdhury et al., 2017; Kumar et al., 2013, 2015).

Air pollutants such as particulate matter (PM), nitrogen oxides (NOx), carbon monoxide (CO), sulphur dioxide (SO_2) and ground-level ozone (O_3) have often been recorded to exceed the National Ambient Air Quality Standards (NAAQS; Sharma, Sharma, Jain, & Kumar, 2013). PM released from both natural and anthropogenic sources is considered as the main air pollutant, which is responsible for the deteriorating quality of ambient air in Delhi city. PM is classified into coarse ($PM_{2.5-10}$), and fine ($PM_{2.5}$) size fractions based on aerodynamic diameter. The main primary sources of PM in Delhi, are vehicular emissions, biomass burning, fossil fuel combustion, diesel generators, construction and road dust emissions (Kumar et al., 2017; Pant et al., 2015; Saxena et al., 2017; Sharma, Mandal et al., 2016; Villalobos et al., 2015). The secondary sources, arising from atmospheric oxidation of precursor gases and condensation of the products formed, also comprise a significant fraction of the observed PM concentration. The main gaseous air pollutants are oxides of nitrogen (NO, and NO_2, collectively NO_X), sulphur dioxide (SO_2), CO, and O_3, which are emitted by various sources such as transportation, power generations, and industrial and domestic fuel combustion, or formed from the oxidation of VOCs in the presence of NO_X the case of O_3. A major source of NO_X emissions is vehicle exhaust and power plants (Tyagi et al., 2016). For example, previous studies have reported that approximately 80–90 % of NO_X and CO are produced from the transport sector in Delhi (Gurjar, van Aardenne, Lelieveld, & Mohan, 2004; Gulia, Shiva Nagendra, Khare, & Khanna,

2015; Tyagi et al., 2016).

In addition to local pollutant emissions, meteorological conditions play a significant role in affecting the concentration of ambient air pollution. Among several meteorological parameters, the role of wind speed and direction is particularly significant in controlling the concentrations of atmospheric pollutants in urban areas. The impact of meteorological conditions on levels of PM and gaseous pollutants has been reported for urban areas in India (Guttikunda & Gurjar, 2012; Tiwari et al., 2014; Yadav, Beig, & Jaaffrey, 2014; Yadav, Sahu, Beig, & Jaaffrey, 2016). Previous work has reported approximately two-times higher concentrations of PM during winter as compared to the summer over Delhi, owing to the effect of local meteorology and high emissions during the winter season (Guttikunda & Gurjar, 2012). During the winter season, the atmosphere of Delhi is characterized by low mixing height, wind speed, and low ventilation that result in less dispersion and increase of air pollutants (Tiwari et al., 2013). However, in the summer season, very high temperatures dilute surface emissions into a deeper atmospheric boundary layer and strong winds effectively ventilate the area causing lower pollution in urban areas.

A number of studies have recently reported quantification the sources and processes that contribute to both $PM_{2.5}$ and PM_{10} and gaseous pollutants in Delhi and the NCR (Table 1). However, there is a lack of investigations to date that have focused on the quantification of the sources and processes that contribute to PM. In this context, the aim of this work is to establish a relationship between PM and metrological conditions through the application of polar plots and k-mean clustering at six sites each within Delhi and across the NCR. A specific focus is given to building an understanding of the relative contributions of primary and secondary sources to the observed PM and gaseous pollutants across Delhi-NCR. We have also examined spatiotemporal variations of PM and gaseous pollutants and their relationship with local meteorology over a four-year duration. The findings from this work will support the air pollution assessment and pollution control strategies in future within this region.

2. Methodology

2.1. Site description

Fig. 1 shows the locations of air quality monitoring stations across Delhi and its NCR. The daily averaged data for NO_X, CO, O_3, SO_2, $PM_{2.5}$, and PM_{10} were collected from January 2014 to December 2017 from 6 air quality stations in Delhi, 4 stations in Haryana, and 2 stations in Uttar Pradesh. Table 2 provides details of the monitoring stations.

Delhi city, the capital of India, is one of the most densely populated cities in the world. Delhi has a population of 16.7 million with an annual average growth rate of 1.92 % (http://census2011.co.in). The overall population density is 11,297 km^{-2}. It is located at an elevation of 216 m above the mean sea level (http://census2011.co.in). Delhi is geographically situated within the coordinates of 28.24 °N to 28.53 °N and 76.50 °E to 77.20 °E; it has a semi-arid climate. The city is surrounded by the mountain region of the Himalaya to the north, central hot peninsular region to the south, hilly region to the east and, to the west the Great Indian Desert (Sahay, 2018; Yadav & Sharma, 2018; Yadav et al., 2016; Yadav, Sharma, Peshin, & Masiwal, 2017). Delhi
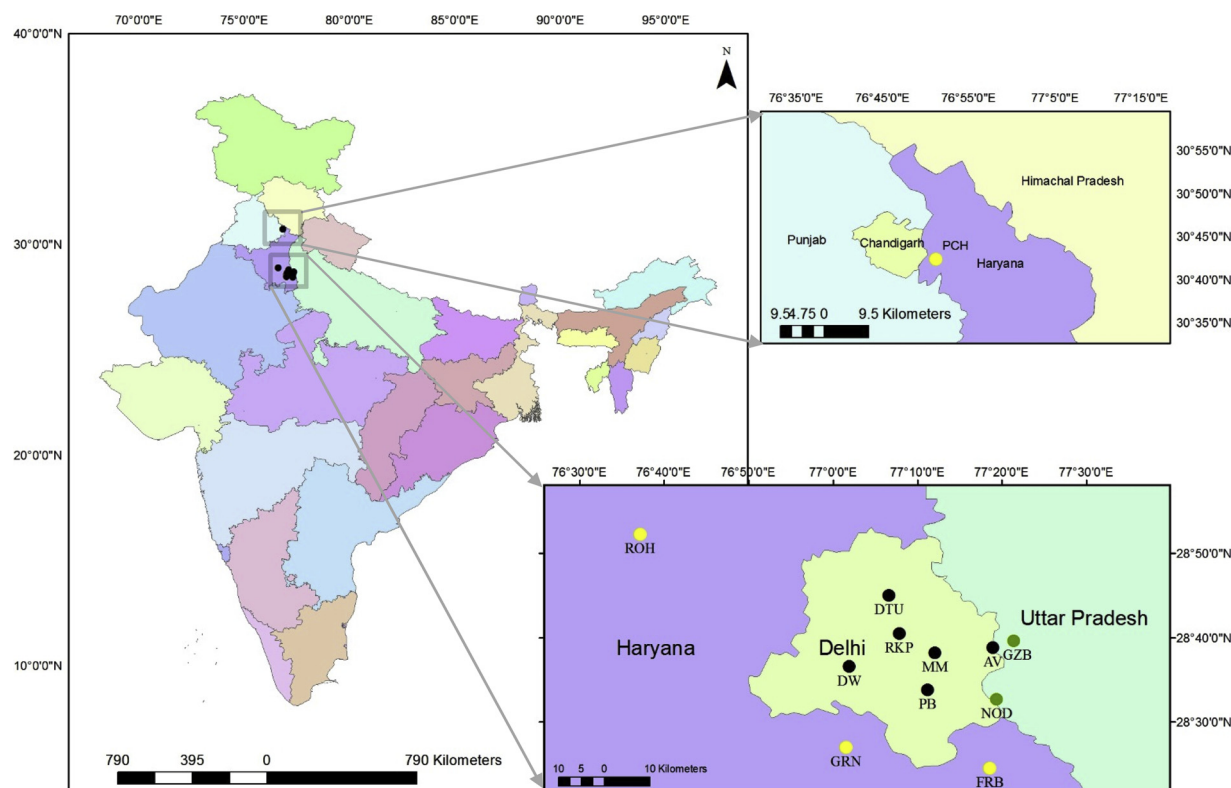
Fig. 1. Map of monitoring stations in the Delhi-NCR region. Black dots represent monitoring sites within Delhi, yellow dots are representing monitoring sites in Haryana state, and green dots are representing sites in Uttar Pradesh.

experiences four main seasons: winter (December-February), summer (March-May), monsoon (June-August) and post-monsoon (September-November). Temperatures range between 7 ± 3 °C in winter and 45 ± 3 °C in summer (Kumar et al., 2017). The city has 93 % population living in urban areas as compared to the national average of 31.16 % (SAD, 2014). Delhi has the highest number of registered motor

vehicles in India. There were about 6.93 million vehicles on the roads in 2011 in Delhi, and those are expected to increase to 25.6 million by 2030 (Kumar, Gurjar, Nagpure, & Harrison, 2011).

The NCR is geographically located between the coordinates 27.60 °N to 29.30 °N and 76.20 °E to 78.40 °E as an area of dense population ($\sim 800/km^2$) covering four states: National Capital Territory-

Table 2
Brief description of monitoring sites operated by CPCB (Central Pollution Control Board), DPCC (Delhi Pollution Control Committee), HSPCB (Haryana State Pollution Control Board) and UPPCB (Uttar Pradesh Pollution Control Board).

State	Monitoring station	Site code	Latitude Longitude	Type of site	Operated by	Data coverage
Delhi	R K Puram	RKP	28.674045 77.131023	Residential	DPCC	94.7 %
	Delhi Technological University	DTU	28.7500499 77.1112615	Residential & industrial	CPCB	27.8 %
	Dwarka	DW	28.60909 77.0325413	Residential	CPCB	60.5 %
	Punjabi Bagh	PB	28.563262 77.186937	Residential, industrial & commercial	DPCC	94 %
	Mandir Marg	MM	28.636429 77.201067	Residential & commercial	DPCC	87.8 %
	Anand Vihar	AV	28.646835 77.316032	Residential, industrial & commercial	DPCC	86.2 %
Haryana	Panchukla	PCH	30.7057778 76.85318055	Residential	HSPCB	54.1 %
	Rohtak	ROH	28.870083 76.620500	Residential	HSPCB	37.8 %
	Gurgaon	GRN	28.4501238 77.0263051	Residential	HSPCB	47.8 %
	Faridabad	FRB	28.4088421 77.3099081	Residential	HSPCB	56.4 %
Uttar Pradesh	Ghaziabad	GZB	28.6603346 77.3572563	Residential, industrial & commercial	UPPCB	27.8 %
	Noida	NOD	28.5447608 77.3231257	Residential, industrial & commercial	UPPCB	27.8 %

Table 3

Air quality instrumentation at selected monitoring sites across Delhi-NCR.

Air quality Parameters	Methods of Measurements	Principle of work
PM_{10}	BAM 1020	Beta ray attenuation
$PM_{2.5}$	BAM 1020	Beta ray attenuation
NO_X	Thermo 42i NO-NO2-NOx monitor (Thermo Fischer Scientific Inc., USA)	Chemiluminescence
O_3	UV photometric 49i (Thermo Fischer Scientific Inc., USA)	Absorption
CO	Non-Dispersive Infrared (NDIR) spectroscopy	Absorption
SO_2	Ultraviolet fluorescence	Ultraviolet fluorescence
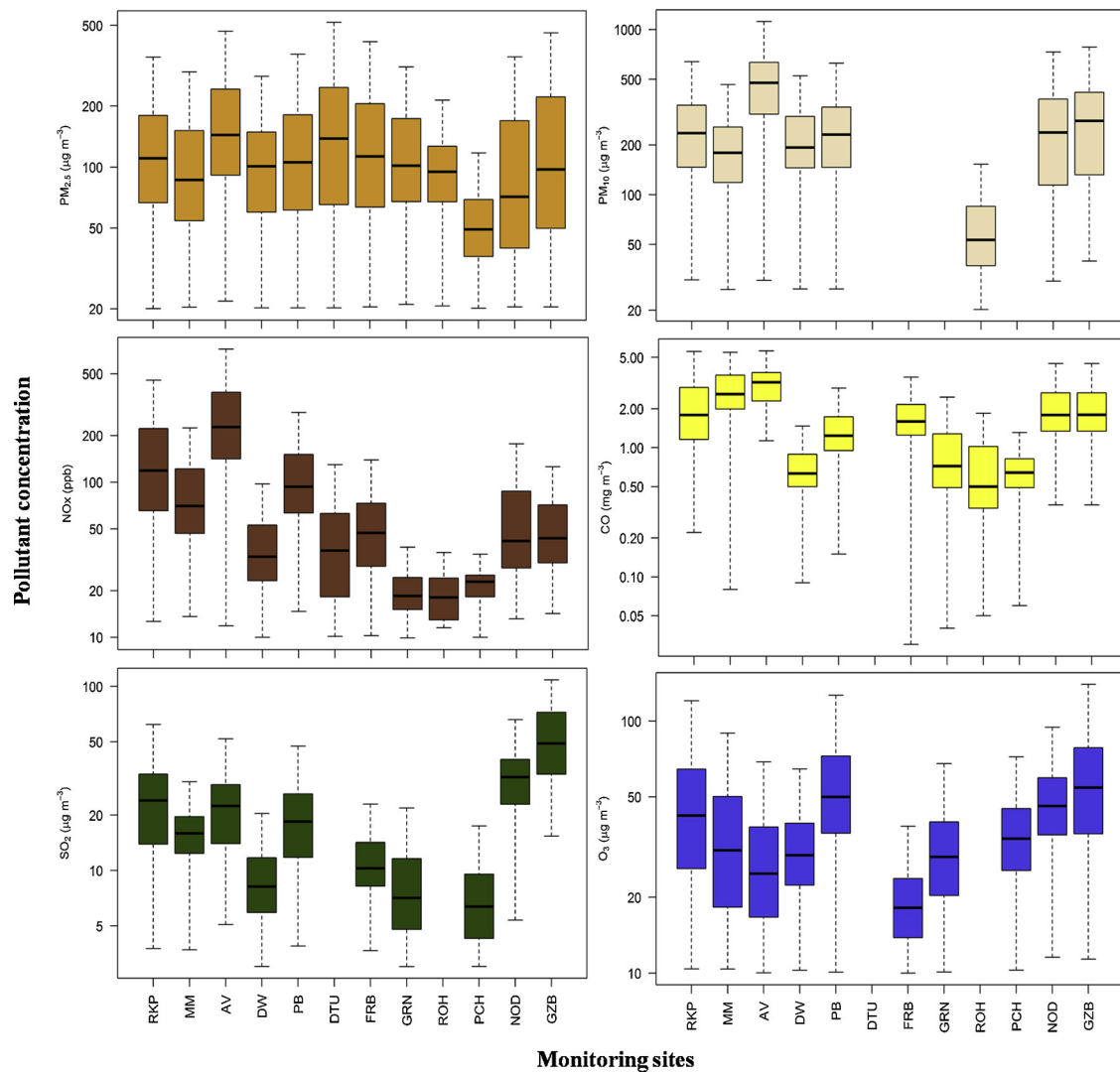
Fig. 2. Boxplots of daily concentrations of analysed pollutants; median is shown by the middle line of the box, the interquartile range is shown by box, and whiskers present the $\pm 1.5 \times$ inter-quartile range. Concentrations are expressed in $\mu g\,m^{-3}$ for $PM_{2.5}$, PM_{10}, SO_2 and O_3, $mg\,m^{-3}$ for CO, and ppb for NOx.

Delhi, Haryana, Uttar Pradesh, and Rajasthan with a total of twenty three districts (Hazarika et al., 2019). Haryana is a fast developing state of north India, situated at 30.30 °N, 74.60 °E and around 275 m above mean sea level; it covers Delhi from three sides and have an area of 44,000 km². The population is about 25.3 million (http://census2011.co.in). Uttar Pradesh state covers a total of 71 districts.

2.2. Data collection and instrumentation

Table 3 summarises the availability of equipment for PM_{10}, $PM_{2.5}$, and gaseous pollutants (NO_X, O_3, CO, SO_2) at the selected monitoring sites. In this work, we collected the data from the Central Pollution Control Board (CPCB), Delhi Pollution Control Committee (DPCC),

Hariyana Pollution Control Board (HSPCB) and Uttar Pradesh Pollution Control Board (UPPCB) run stations. Daily concentrations of all these pollutants were downloaded from the CPCB database (cpcb.nic.in) for all 12 monitoring sites across Delhi-NCR over a period of four years (2014–2017). The monitors are reported to be regularly calibrated by operating bodies in accordance with the instruction manual of the equipment for ensuring the quality of the data, as discussed in previous studies (Kumar et al., 2014; Tiwari, Dahiya, & Kumar, 2015). Daily meteorological data (ambient temperature, relative humidity, solar radiation and wind speed and direction) for each site separately were acquired from the CPCB database for the monitoring sites, which are operated by the CPCB and DPCC. The measurement errors in pollutant concentration data are typically reported to be smaller than 5 % (CPCB,

Table 4

The statistics of daily PM and gaseous pollutant concentrations at the six monitoring stations (2014–2017) across Delhi. Note that '- 'shows the unavailability of the data.

	year	2014		2015		2016		2017	
	Pollutants	mean ± SD	median	mean ± SD	median	mean ± SD	median	mean ± SD	median
RKP	PM_{10}	262 ± 139	229	247 ± 131	226	271 ± 154	255	245 ± 144	238
	$PM_{2.5}$	140 ± 87	112	125 ± 81	104	136 ± 99	112	131 ± 91	111
	NO_x	–	–	189 ± 140	151	174 ± 135	125	118 ± 90	93
	CO	–	–	2.1 ± 1.3	1.7	2.1 ± 1.3	1.8	2.2 ± 1.3	1.8
	O_3	–	–	49 ± 27	44	49 ± 28	42	46 ± 24	38
	SO_2	–	–	18 ± 14	12	26 ± 15	25	28 ± 11	26
MM	PM_{10}	194 ± 116	198	197 ± 105	172	241 ± 138	219	188 ± 106	171
	$PM_{2.5}$	129 ± 85	96	108 ± 72	82	121 ± 104	84	105 ± 85	80
	NO_x	–	–	–	–	98 ± 72	73	92 ± 67	67
	CO	–	–	6.9 ± 3.2	8.7	–	–	2.6 ± 1.3	1.8
	O_3	–	–	49 ± 18	49	–	–	24 ± 11	20
	SO_2	–	–	–	–	–	–	17 ± 8	15
AV	PM_{10}	581 ± 281	565	468 ± 204	461	424 ± 275	356	468 ± 168	449
	$PM_{2.5}$	190 ± 118	150	165 ± 99	140	172 ± 133	123	190 ± 117	157
	NO_x	–	–	–	–	251 ± 189	182	319 ± 175	266
	CO	–	–	–	–	–	–	3.1 ± 1	3.2
	O_3	–	–	–	–	27 ± 14	24	36 ± 25	25
	SO_2	–	–	–	–	20 ± 13	18	30 ± 13	26
DW	PM_{10}	232 ± 116	191	222 ± 154	203	–	–	254 ± 194	194
	$PM_{2.5}$	–	–	81 ± 51	62	140 ± 86	121	140 ± 137	105
	NO_x	53 ± 40	38	46 ± 32	34	27 ± 14	23	64 ± 56	36
	CO	0.7 ± 0.4	0.6	0.9 ± 0.6	0.7	0.7 ± 0.5	0.6	0.7 ± 0.3	0.6
	O_3	–	–	–	–	36 ± 20	31	30 ± 13	29
	SO_2	11 ± 8	9	10 ± 8	7	9 ± 6	7	10 ± 5	9
PB	PM_{10}	262 ± 136	237	263 ± 144	237	276 ± 174	236	245 ± 158	208
	$PM_{2.5}$	139 ± 88	112	131 ± 82	110	135 ± 117	95	125 ± 100	97
	NO_x	–	–	133 ± 94	103	130 ± 94	94	104 ± 83	79
	CO	–	–	1.3 ± 0.5	1.2	1.7 ± 1.1	1.3	1.6 ± 1.2	1.1
	O_3	–	–	62 ± 30	55	56 ± 76	49	53 ± 22	49
	SO_2	–	–	18 ± 12	13	19 ± 11	17	23 ± 10	20
DTU	PM_{10}	–	–	–	–	–	–	–	–
	$PM_{2.5}$	–	–	–	–	–	–	–	–
	NO_x	–	–	–	–	134 ± 124	75	43 ± 39	29
	CO	–	–	–	–	–	–	–	–
	O_3	–	–	–	–	–	–	–	–
	SO_2	–	–	–	–	–	–	–	–

2010; Tyagi et al., 2016).

As a quality control exercise, the data were screened for irregularities and removal of maintenance periods. Before further analyses, the data were first inspected for zero values, negative values and outliers by manual observations. Then, the whole data set was analysed using the R statistical package (R Core Team, 2015) in the Open-air software package (Carslaw & Ropkins, 2012; Carslaw, 2015) as 'summary plots' to identify missing periods and assess the basic statistics of the data. Finally, the data for each site were plotted and checked for 'outliers' by using R package and then the data were used for further analysis and interpretation.

2.3. Data analysis

The spatial characteristics of concentrations of $PM_{2.5}$ and PM_{10} between Delhi and NCR sites were evaluated by using Pearson correlation coefficients (r) and coefficients of divergence (COD). r shows the degree of correlation of PM concentrations between two sampling sites while the COD (Eq. 1) evaluates the degree of uniformity between sampling sites. COD values close to 0 represent homogeneity between pairs of sites, while values approaching 1 represent complete divergence (Contini, Donateo, Elefante, & Grasso, 2012; Jeong et al., 2010; Krudysz, Moore, Geller, Sioutas, & Froines, 2009; Turner & Allen, 2008;

Wilson, Kingham, Pearce, & Sturman, 2005).

$$COD_{ab} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left[\frac{(C_{ia} - C_{ib})}{(C_{ia} + C_{ib})}\right]^2}$$ (1)

where C_{ia} and C_{ib} are PM concentrations in the day i at sites a and b, respectively, and n is the number of observations (Krudysz et al., 2009; Wongphatarakul, Friedlander, & Pinto, 1998). A boundary COD value of 0.2 was adopted, where COD values greater than 0.20 are defined as heterogeneous spatial distributions, and values smaller than 0.20 indicate similarities between the sites (Cesari et al., 2016; Pinto, Lefohn, & Shadwick, 2004; Wilson et al., 2005).

A relationship between the air pollutant concentrations, the meteorological parameters and the transport pathways from different sources can be demonstrated by bivariate concentration polar plots and k-means clustering techniques (Carslaw & Beevers, 2013). Bivariate polar plots show how a pollutant concentration varies together with wind speed and wind direction in polar coordinates. These also provide a modelled surface that shows the relationship between pollutant concentration, wind speed and wind direction using a Generalized Additive Modelling (GAM) method to smooth the noisiness from the raw data (Carslaw & Beevers, 2013). The features observed on the polar plot are grouped using the k-means clustering technique.
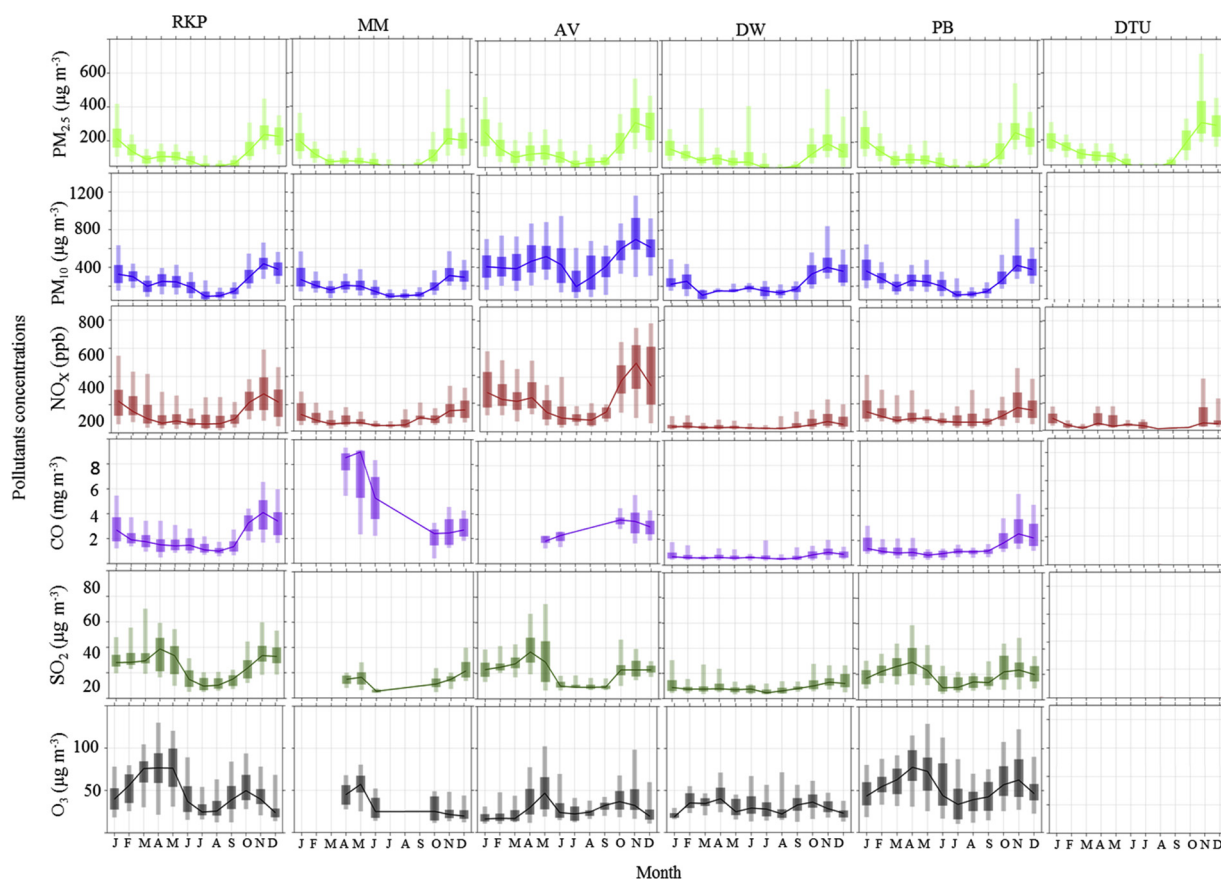
Fig. 3. Monthly variations in the median, 25th/75th and 5th/95th quantile values for all pollutants for 2014–2017 at six sites within Delhi.
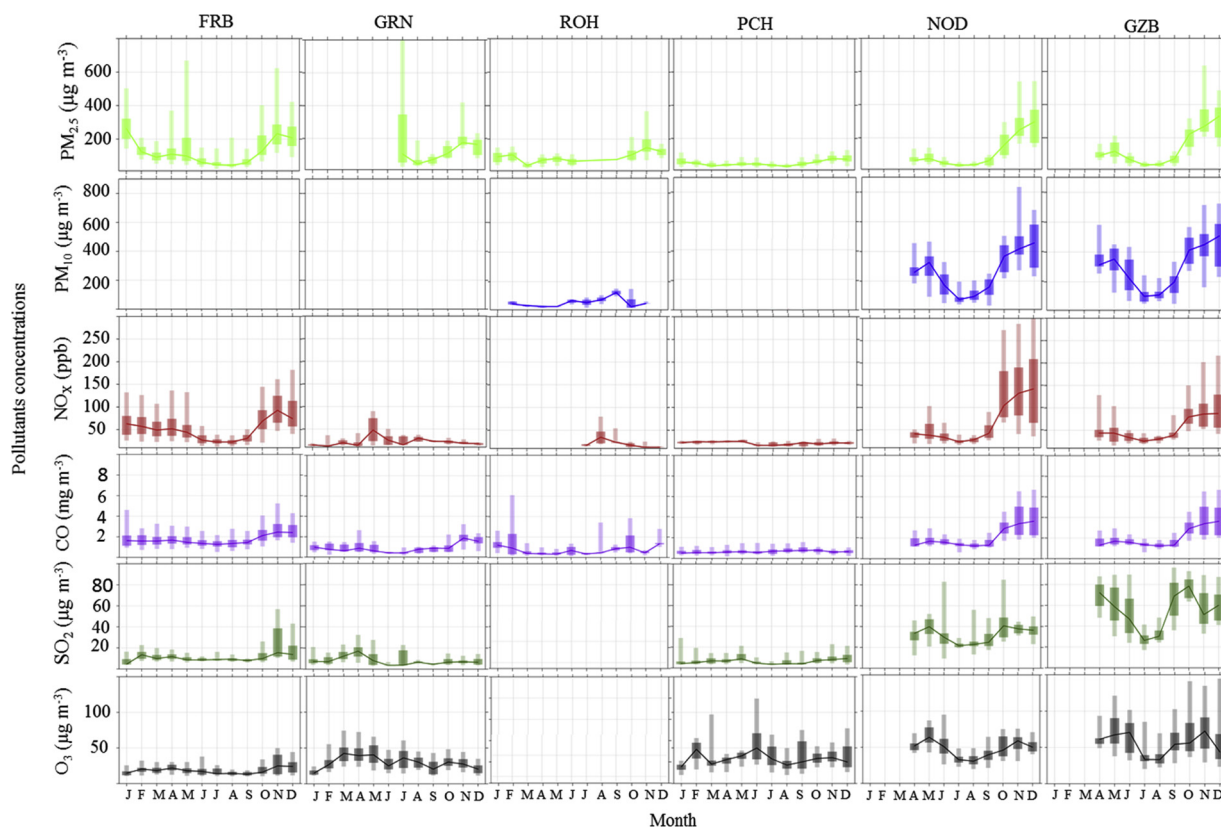

Fig. 4. Monthly variations in the median, 25th/75th and 5th/95th quantile values for all pollutants for 2014–2017 at six sites across NCR.

Table 5

Summary statistics of seasonal concentration of PM_{10} ($\mu g/m^3$), $PM_{2.5}$ ($\mu g/m^3$), NOx (ppb), CO (mg/m^3), SO_2 ($\mu g/m^3$), and O_3 ($\mu g/m^3$) at the 12 monitoring stations during the study period (2014–2017). (W = winter, S = summer, M = monsoon, P-m = post-monsoon). CO is kept to two decimals because the lower values. Note that '- 'shows the unavailability of the data.

Site	Season	PM_{10} $C_{mean}+SD$ (C_{med})	$PM_{2.5}$ $C_{mean} \pm SD$ (C_{med})	NO_X $C_{mean} \pm SD$ (C_{med})	O_3 $C_{mean} \pm SD$ (C_{med})	CO $C_{mean} \pm SD$ (C_{med})	SO_2 $C_{mean} \pm SD$ (C_{med})
RKP	W	348 ± 116 (338)	203 ± 84 (190)	238 ± 125 (213)	38 ± 21 (32)	3.1 ± 2.2 (2.8)	34 ± 10 (32)
	S	241 ± 91 (232)	106 ± 40 (102)	115 ± 84 (86)	74 ± 27 (73)	1.7 ± 0.8 (1.5)	34 ± 20 (31)
	M	139 ± 76 (121)	66 ± 30 (59)	86 ± 46 (79)	34 ± 18 (29)	1.4 ± 0.9 (1.2)	13 ± 4 (11)
	P-M	303 ± 158 (291)	163 ± 105 (139)	214 ± 128 (182)	47 ± 18 (44)	2.9 ± 1.5 (2.7)	24 ± 12 (20)
MM	W	275 ± 106 (252)	174 ± 75 (158)	134 ± 78 (104)	22 ± 10 (19)	2.9 ± 0.9 (2.7)	23 ± 9 (21)
	S	192 ± 84 (176)	76 ± 30 (72)	71 ± 44 (58)	50 ± 17 (46)	–	15 ± 6 (14)
	M	114 ± 59 (100)	52 ± 21 (48)	46 ± 24 (39)	–	–	–
	P-M	224 ± 126 (189)	143 ± 110 (110)	119 ± 73 (104)	20 ± 14 (17)	2.3 ± 1.25 (2.2)	13 ± 6 (12)
AV	W	486 ± 191 (467)	248 ± 108 (235)	–	20 ± 11 (17)	3.2 ± 0.8 (2.8)	24 ± 8 (23)
	S	473 ± 190 (455)	133 ± 64 (122)	232 ± 131 (202)	35 ± 22 (30)	–	32 ± 16 (28)
	M	286 ± 171 240	95 ± 47 87	107 ± 55 95	24 ± 12 22	–	17 ± 9 11
	P-M	621 ± 279 (594)	208 ± 135 (168)	–	35 ± 13 (32)	–	21 ± 14 (20)
DW	W	277 ± 131 (245)	147 ± 62 (134)	55 ± 40 (41)	26 ± 11 (23)	0.8 ± 0.4 (0.7)	11 ± 7 (9)
	S	–	113 ± 83 (95)	34 ± 16 (29)	35 ± 14 (34)	0.6 ± 0.3 (0.5)	9 ± 6.5 (7)
	M	144 ± 45 (137)	86 ± 57 (94)	28 ± 14 (25)	28 ± 22 (23)	0.8 ± 0.5 (0.7)	6.5 ± 3 (5)
	P-M	320 ± 210 (303)	147 ± 108 (126)	70 ± 46 (56)	34 ± 11 (33)	0.9 ± 0.4 (0.7)	12 ± 8 (10)
PB	W	353 ± 129 (327)	204 ± 81 (190)	159 ± 91 (130)	49 ± 18 (47)	1.9 ± 1.1 (1.4)	19 ± 7 (18)
	S	240 ± 100 (221)	97 ± 40 (92)	108 ± 69 (84)	77 ± 68 (71)	1.1 ± 0.4 (0.9)	28 ± 15 (25)
	M	148 ± 77 (131)	59 ± 25 (55)	87 ± 48 (79)	43 ± 26 (34)	1.2 ± 0.2 (1.1)	11 ± 4.7 (11)
	P-M	303 ± 183 (263)	165 ± 124 (129)	135 ± 102 (99)	58 ± 24 (57)	2.2 ± 1.1 (1.6)	21 ± 12 (19)
DTU	W	–	219 ± 99 (233)	68 ± 49 (60)	–	–	–
	S	–	110 ± 45 (108)	53 ± 35 (52)	–	–	–
	M	–	48 ± 24 (43)	27 ± 11 (22)	–	–	–
	P-M	–	205 ± 160 (183)	25 ± 13 (20)	–	–	–
FRB	W	–	212 ± 111 (185)	72 ± 37 (63)	22 ± 8 (21)	2.3 ± 0.99 (2.1)	14 ± 10 (12)
	S	–	155 ± 60 (94)	58 ± 41 (49)	–	1.9 ± 1.7 (1.8)	10 ± 4 (9)
	M	–	63 ± 47 (60)	28 ± 23 (24)	–	–	–
	P-M	–	170 ± 105 (147)	74 ± 44 (62)	21 ± 9 (20)	2.2 ± 0.9 (1.9)	18 ± 11 (17)

Table 5 (*continued*)

Site	Season	PM$_{10}$ C$_{mean}$ + SD (C$_{med}$)	PM$_{2.5}$ C$_{mean}$ ± SD (C$_{med}$)	NO$_X$ C$_{mean}$ ± SD (C$_{med}$)	O$_3$ C$_{mean}$ ± SD (C$_{med}$)	CO C$_{mean}$ ± SD (C$_{med}$)	SO$_2$ C$_{mean}$ ± SD (C$_{med}$)
GRN	W	–	–	21 ± 6 (16)	19 ± 10 (18)	1.2 ± 1 (1.18)	8 ± 5 (7)
	S	–	–	28 ± 16 (15)	41 ± 14 (40)	0.95 ± 0.3 (0.9)	13 ± 8 (11)
	M	–	–	15 ± 6 (15)	31 ± 12 (30)	0.5 ± 0.2 (0.4)	4 ± 3 (3)
	P-M	–	134 ± 86 (107)	31 ± 17 (20)	27 ± 10 (27)	1 ± 0.5 (0.8)	6 ± 3 (4)
ROH	W	–	108 ± 35 (111)	–	–	1.3 ± 0.8 (1.1)	–
	S	–	62 ± 27 (57)	18 ± 4 (18)	–	0.34 ± 0.2 (0.33)	–
	M	–	–	27 ± 17 (21)	–	0.97 ± 0.3 (0.4)	–
	P-M	–	157 ± 109 (137)	–	–	1.2 ± 0.8 (0.9)	–
PCH	W	–	64 ± 29 (62)	23 ± 3 (22)	30 ± 12 (27)	0.6 ± 0.3 (0.6)	9 ± 5 (7)
	S	–	44 ± 23 (40)	25 ± 3 (25)	36 ± 18 (33)	0.7 ± 0.4 (0.6)	8 ± 4 (7)
	M	–	38 ± 25 (37)	21 ± 11 (18)	±	0.7 ± 0.5 (0.7)	4.5 ± 3 (4)
	P-M	–	63 ± 26 (60)	20 ± 4 (20)	38 ± 13 (36)	0.9 ± 0.5 (0.7)	7 ± 4 (6)
NOD	W	487 ± 244 (496)	294 ± 146 (297)	161 ± 116 (141)	±	3.7 ± 1.5 (3.6)	35 ± 8 (36)
	S	293 ± 107 (292)	79 ± 33 (74)	45 ± 25 (38)	64 ± 22 (60)	1.7 ± 0.5 (1.6)	38 ± 15 (37)
	M	129 ± 78 (101)	40 ± 15 (37)	29 ± 11 (26)	41 ± 21 (35)	1.4 ± 0.4 (1.3)	28 ± 18 (23)
	P-M	324 ± 176 (305)	171 ± 128 (152)	108 ± 75 (83)	48 ± 18 (45)	2.7 ± 1.4 (2.5)	36 ± 14 (35)
GZB	W	460 ± 167 (507)	303 ± 122 (330)	106 ± 64 (87)	±	3.7 ± 1.5 (3.6)	61 ± 19 (60)
	S	345 ± 111 (352)	118 ± 52 (112)	49 ± 32 (43)	70 ± 24 (62)	1.8 ± 0.5 (1.7)	75 ± 32 (71)
	M	159 ± 101 (118)	46 ± 24 (41)	29 ± 10 (28)	54 ± 37 (39)	1.5 ± 0.4 (1.4)	37 ± 18 (32)
	P-M	363 ± 192 (340)	203 ± 145 (205)	75 ± 43 (63)	70 ± 37 (56)	2.7 ± 1.4 (2.5)	76 ± 28 (75)

k-means clustering is a method in which bivariate polar plot features can be identified and grouped. The main aim of grouping data in this technique is to identify recorded values in the original time series data by cluster to enable post-processing to well understand potential source features. K-means clustering, ranging from a two-cluster to a six-cluster solution, was considered in this study.

Data used in polar cluster analysis are daily averaged PM$_{2.5}$ concentration measured during the study period for both MM site in Delhi and NOD in NCR. The corresponding meteorological data for the wind speed and direction were taken from IGI airport and Noida for MM and NOD sites, respectively. In total, we have used 907 data points for MM, and 248 data points for NOD for plotting the polar clusters (Section 3.5). Details on the theoretical development of these plots can be found in the Open-air software package (Carslaw, 2015). In addition, the number of clusters (k) is determined for PM$_{2.5}$ for all monitoring sites in this study by using the elbow method. This method calculates the root mean square deviation (RMSD) as a function of *k* (Lee & Kim, 2018). When another cluster is added, it does not significantly improve and change the RMSD value of the population. Therefore, we selected *k* (= 6) as an optimal number.

3. Results and discussion

3.1. Overview of air pollutants over Delhi-NCR

Fig. 2 shows the daily PM and gaseous pollutant concentrations over four years at each of the 12 sites. The mean, standard deviation (SD) and median values are presented in Table 4. Within Delhi, AV site shows relatively higher values of PM and gaseous pollutants, except O$_3$. For example, the four-year average concentration (± SD) of PM$_{2.5}$ and NOx at the AV site were 179 ± 99 μg/m^3 and 285 ± 165 ppb, respectively. However, the corresponding values at RKP and DW sites were 133 ± 90 and 120 ± 96 μg m^{-3}, and 124 ± 104, 47 ± 35 ppb, respectively. In addition, it is observed that annual average PM$_{2.5}$ and NOx concentration data exceeded the NAAQS value of 60 μg/m^3 and 42 ppb, respectively, at all the monitoring stations (CPCB, 2010). The AV site showed PM$_{2.5}$ and NOx concentrations higher than the NAAQS standard, up to about 3- and 7-times, respectively. The high levels of pollutants at this site might be due to the location of the site, which is close to traffic and residential pollution sources (Gulia, Mittal, & Khare, 2018; Kumar et al., 2017).

In case of the NCR, the highest average concentration of PM$_{2.5}$ was observed at the FRB site, which was 159 ± 97 μg/m^3, followed by 149 ± 133 μg/m^3 at the GZB site (Fig. 2). In general, the NOD and GZB
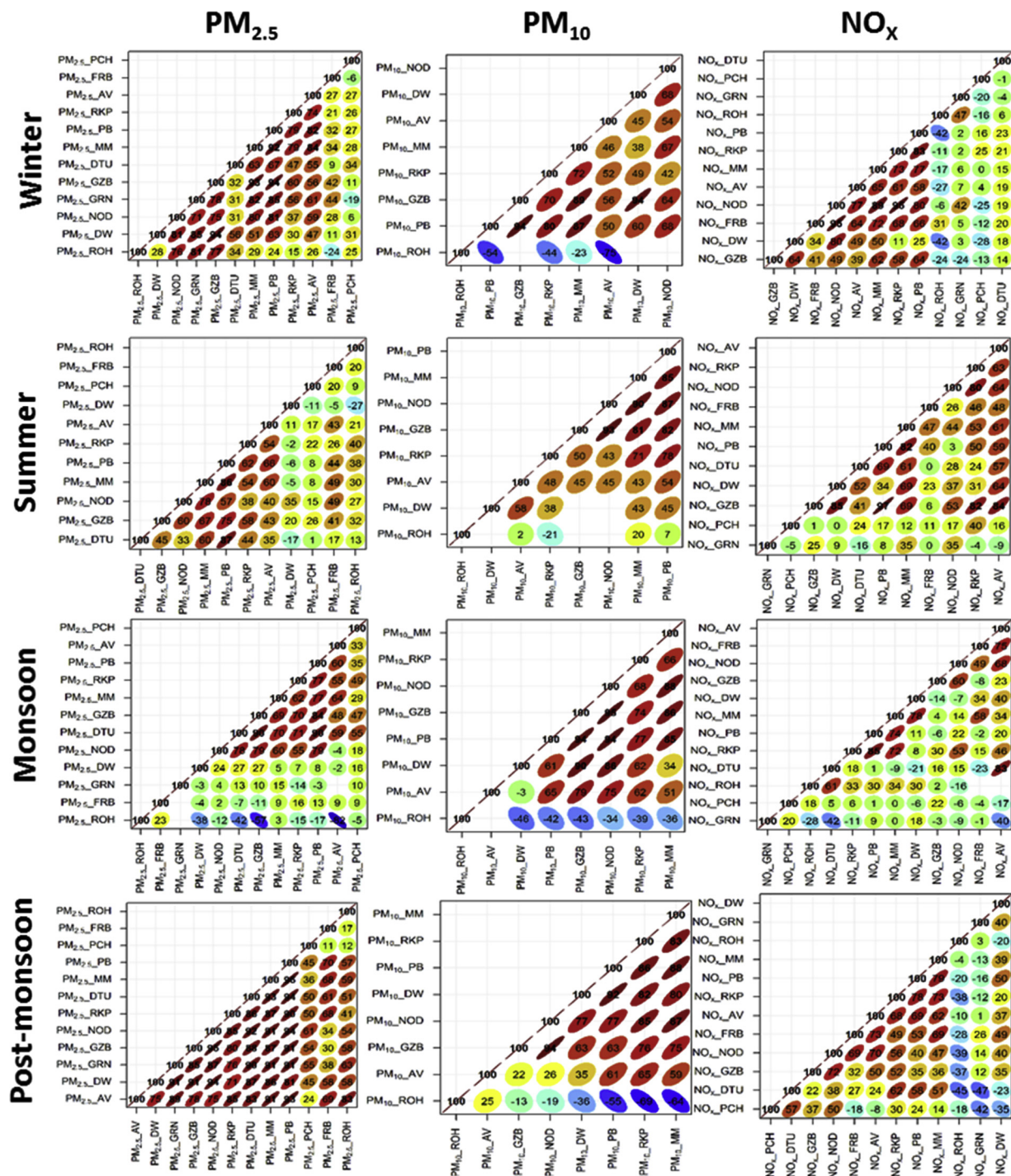
Fig. 5. Correlation matrix showing the relationships between $PM_{2.5}$, PM_{10} and NO_X over the period at all sites. The plot shows the Pearson correlation coefficients expressed as −100 to 100. 100 is perfect correlation, zero is no correlation and −100 is a perfect inverse correlation.

sites showed a relatively higher concentration of all pollutants. For instance, the average concentration for NOx were 76 ± 67 and 59 ± 45 ppb, CO were 2.2 ± 1.3 and 2.2 ± 1.3 mg/m³, and SO₂ were 33 ± 15 and 53 ± 23 µg/m³ at NOD and GZB sites, respectively. A very low concentration was observed at the PCH site, indicating that the PCH site can be considered as a possible background site for the Delhi city. The average concentration of $PM_{2.5}$, NOx, and SO₂ at PCH was 57 ± 27 µg/m³, 23 ± 8 ppb, 10 ± 7 µg/m³, respectively. The above finding suggests diversity in concentrations at different sites across the Delhi-NCR region. While most sites, despite some being in the NCR region, showed high concentrations only the PCH sites showed

potential to be considered as an urban background site, owing to the consistent lowest concentration observed across the years.

3.2. Annual variation

Fig. 3 shows the monthly variations of daily $PM_{2.5}$, PM_{10}, NOx, CO, SO₂ and O₃ concentrations from 1 January 2014 to 31 December 2017 over Delhi city. Similar patterns were observed for all species (except O₃) with the highest levels observed during the cold period, and the lowest concentration during the monsoon months in each of the examined years. Previous studies have also reported similar features in

Fig. 6. Correlation matrix showing the relationships between CO, SO$_2$ and O$_3$ over the period at all sites. The plot shows the Pearson correlation coefficients expressed as –100 to 100. 100 is perfect correlation, zero is no correlation and –100 is a perfect inverse correlation.

Delhi (Arif, Kumar, Kumar, Eric, & Gourav, 2018; Gupta, Gadi, Sharma, & Mandal, 2018; Kumar et al., 2017; Peshin, Sharma, Sharma, Naja, & Mandal, 2017). The monthly average concentrations of all parameters reached their maximum during winter and post-monsoon months while they fell to their minimum in the monsoon season (Figs. 3 and 4). The seasonal average concentrations of all pollutants are summarised in Table 5. The average PM$_{2.5}$ and PM$_{10}$ concentrations showed high intra-annual variation, with highest in winter and post-monsoon as opposed to the lowest levels in the monsoon season. The higher levels of PM$_{2.5}$ and PM$_{10}$ in the winter months are probably due to an increase in coal and biomass burning for residential heating as most parts of the region do not have a central heating system (Kumar et al., 2015; Masih, Singhvi, Taneja, Kumar, & Masih, 2012; Nagpure et al., 2015). Moreover, meteorological factors play a significant role in the accumulation

Table 6

COD values of the daily $PM_{2.5}$ (PM_{10}) mass concentrations ($\mu g\ m^{-3}$) between the monitoring sites within Delhi (2014–2017). Note that '- 'shows the unavailability of the data.

COD		RKP	MM	AV	DW	PB	DTU
Delhi	**RKP**	0	0.20 (0.20)	0.20 (0.34)	0.24 (0.21)	0.15 (0.14)	0.18 (-)
	MM		0	0.27 (0.44)	0.25 (0.26)	0.15 (0.17)	0.21 (-)
	AV			0	0.28 (0.43)	0.19 (0.34)	0.19 (-)
	DW				0	0.25 (0.19)	0.22 (-)
	PB					0	0.12 (-)
	DTU						0
NCR	**FRB**	0.24 (-)	0.26 (-)	0.25 (-)	0.31 (-)	0.23 (-)	0.27 (-)
	GRN	0.32 (-)	0.30 (-)	0.28 (-)	0.31 (-)	0.31 (-)	0.33 (-)
	ROH	0.32 (-)	0.26 (-)	0.38 (-)	0.33 (-)	0.33 (-)	0.37 (-)
	PCH	0.42 (-)	0.36 (-)	0.50 (-)	0.42 (-)	0.42 (-)	0.47 (-)
	NOD	0.20 (-)	0.16 (-)	0.27 (-)	0.21 (-)	0.12 (-)	0.16 (-)
	GZB	0.17 (-)	0.19 (-)	0.20 (-)	0.19 (-)	0.13 (-)	0.13 (-)

of $PM_{2.5}$ and PM_{10} during winter, owing to the lower boundary layer height and temperature, reduced precipitation, and low wind speed (Dumka et al., 2019; Ganguly, Sharma, & Kumar, 2019; Guo et al., 2017; He et al., 2017; Kumar, Ambade, Sankar, Sethi, & Kurwadkar, 2020). During post-monsoon, open biomass burning (for example, crop burning) leads to increased $PM_{2.5}$ and PM_{10} concentrations in Delhi and its NCR region (Liu et al., 2018). In addition to local emission and meteorological conditions, various factors such as long-range transport, and dust play an important role in the accumulation or dispersion of pollutants at urban sites (Arif et al., 2018). The average $PM_{2.5}$ concentrations over winter for RKP, MM, AV, DW, PB, and DTU in Delhi were 203 ± 84 $\mu g/m^3$, 174 ± 75 $\mu g/m^3$, 248 ± 108 $\mu g/m^3$, 113 ± 83 $\mu g/m^3$, 97 ± 40 $\mu g/m^3$, and 219 ± 99 $\mu g/m^3$, respectively. For FRB, ROH, PCH, NOD, and GZB in NCR region, these were, 212 ± 111 $\mu g/m^3$, 108 ± 35 $\mu g/m^3$, 64 ± 29 $\mu g/m^3$, 294 ± 146 $\mu g/m^3$ and 303 ± 122 $\mu g/m^3$, respectively. The winter average PM_{10} concentrations for RKP, MM, AV, DW, and PB in Delhi were, 348 ± 116 $\mu g/m^3$, 275 ± 106 $\mu g/m^3$, 486 ± 191 $\mu g/m^3$, 277 ± 131 $\mu g/m^3$, and 353 ± 129 $\mu g/m^3$, respectively. The corresponding values for NOD and GZB in NCR region were 487 ± 244 $\mu g/m^3$, and 460 ± 167 $\mu g/m^3$, respectively (Table 5). The annual average $PM_{2.5}$ and PM_{10} concentrations are ~5 and 5.7 times higher than the National Ambient Air Quality Standards (NAQS; http://cpcb.nic.in/air-quality-standard/ for $PM_{2.5}$ (40 $\mu g/m^3$) and PM_{10} (60 $\mu g/m^3$). They are ~6 and 2.3 times higher than the daily US EPA standards for $PM_{2.5}$ (35 $\mu g/m^3$) and PM_{10} (150 $\mu g/m^3$) (US EPA, 2012). The above values are approximately 20 and 17 times higher than the annual limits set by WHO for $PM_{2.5}$ (10 $\mu g/m^3$) and PM_{10} (20 $\mu g/m^3$) (WHO, 2005). They are also around ~7 and 8 times higher than the limits of European Union Air Quality Annual Standards for $PM_{2.5}$ (25 $\mu g/m^3$) and PM_{10} (40 $\mu g/m^3$; http://ec.europa.eu/environment/air/quality/standards. htm).

NOx, CO and SO_2 also exhibited similar seasonal variations, with highest levels in winter and post-monsoon and lowest in the monsoon (Figs. 3 and 4; Table 5), owing to a combined effects of primary emissions from traffic and domestic heating (Yin et al., 2019), weak photochemical reactions and adverse diffusion conditions (Ran et al., 2014; Zhao, Yu, Yin, & He, 2015). There was a peak in summer (or early pre-monsoon season) for SO_2 for RKP, AV, and PB sites in Delhi, and NOD, GZB sites in NCR region (Figs. 3 and 4), which could be attributed to the stable meteorological condition (high temperature, dry, and low wind speed) (Gaur, Tripathi, Kanawade, Tare, & Shukla, 2014). The monthly concentration of O_3 is shown in Figs. 3 and 4. The O_3 levels were lower in winter months and start increasing in summer and again

decreasing in the monsoon. This large seasonal variability of O_3 and its precursors is due to the effect of meteorological condition and secondary chemical factors (NOx and VOC variability). The lower concentrations in winter months may be attributed to shorter daylight hours and lower solar radiation (Guo et al., 2017; Liu et al., 2018; Maji, Ye, Arora, & Nagendra, 2019), and also the effect of the chemical reaction between NO and O_3 (Wang & Hao, 2012). The higher concentrations in summer are consistent with enhanced photochemical oxidation of precursors by higher solar radiation and temperature (Sharma, Sharma, Rohtash, & Mandal, 2016; Wang & Hao, 2012). Non-significant daily variations were observed for all pollutants, but lower concentrations were found on Sunday, except for ozone which high levels were observed due to the weekend effect (Schipa et al., 2009) (Figs. S1 and S2). The above-finding highlight the clear seasonal variation of pollutants across the region. Highest levels of all pollutants, except O_3, occurred during winter as opposed to the lowest during monsoon.

3.3. Spatial variation of $PM_{2.5}$ and PM_{10}

Seasonal spatial correlations between all monitoring sites for daily concentrations of $PM_{2.5}$, PM_{10}, NOx, CO, O_3, and SO_2 are presented in Figs. 5 and 6. High correlations were observed among all the six sites in Delhi, while a low correlation was found between sites within Delhi and its NCR. The correlation was also observed to vary between different seasons. For example, all the pollutants showed a high correlation in winter and post-monsoon months (see in Figs. 5 and 6) and a low correlation in the monsoon season. The results showed a high homogeneity between Delhi monitoring sites. To further investigate homogeneity and heterogeneity in the correlation analysis using the methods described in Section 2.4, the COD for daily average $PM_{2.5}$ and PM_{10} were calculated for all sites in pairs (Table 6). COD values are consistent with the correlation matrix results (Figs. 5 and 6), showing high heterogeneity between Delhi and NCR monitoring sites, especially ROH and PCH sites and also showing $PM_{2.5}$ and PM_{10} to be more spatially homogeneous than gaseous pollutants (Figs. 5 and 6). According to previous studies (Cesari et al., 2016; Contini et al., 2012; Wilson et al., 2005), the threshold value was set to 0.2 for the comparison of COD values of $PM_{2.5}$, and PM_{10} between all monitoring sites. Most COD values for sites in Delhi were lower than the threshold (0.2), while most COD values for NCR sites compared with Delhi sites were higher than the threshold. Highest COD values were found between Delhi (all sites) and ROH and PCH (COD_{max} = 0.38 for ROH, and COD_{max} = 0.5 for PCH), indicating relatively heterogeneous spatial distributions (Tiwari, Dahiya et al., 2015; Tiwari, Hopke et al., 2015; Wang, Hopke, & Utell, 2011). The mean COD values for daily $PM_{2.5}$ were ~0.20 (varied from 0.12 to 0.28, p-value < 0.05), indicating spatial homogeneity between Delhi sites. The mean COD values were ~0.3 between Delhi and NCR monitoring sites, indicating a relatively heterogeneous spatial distribution. It can be concluded that $PM_{2.5}$ concentrations are relatively homogeneous in spatial distribution within Delhi, while $PM_{2.5}$ concentrations are relatively heterogeneous in spatial distribution between Delhi (all sites) and NCR (ROH and PCH).

Pearson correlation analysis was performed between $PM_{2.5}$, PM_{10}, gas-phase pollutants, and meteorological data (Table S1). $PM_{2.5}$ and PM_{10} were moderately correlated with gas-phase species and negatively correlated with wind speed. Overall, the abundance of $PM_{2.5}$ and PM_{10} co-varies within Delhi during winter.

3.4. Characteristics ratios

The $PM_{2.5}/PM_{10}$, $PM_{2.5}/NOx$, $PM_{2.5}/CO$, $PM_{2.5}/SO_2$ ratios for each site within Delhi for the study period are presented in (Figs. 7 and S3). The shapes of the trends were relatively constant through the years for all sites. In the case of the RKP site, the trend was increased for $PM_{2.5}/PM_{10}$, and $PM_{2.5}/NOx$ ratios (Fig. 7), while it was decreased
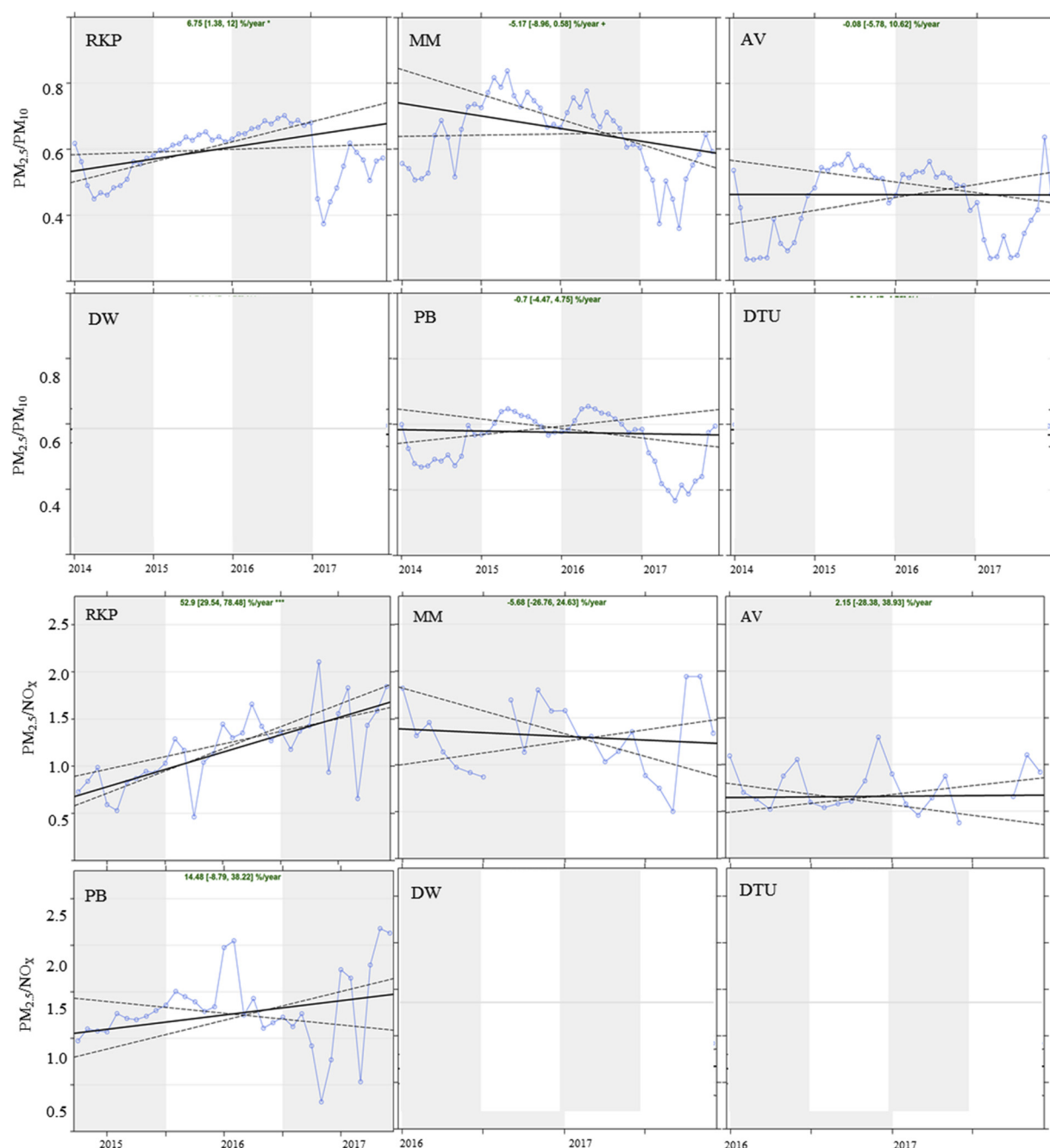
Fig. 7. Linear trends for $PM_{2.5}/PM_{10}$, and $PM_{2.5}/NOx$ calculated over the entire period. Trends expressed in percentage (%) y^{-1} along with the upper and lower 95th confidence intervals in the trends.

dramatically for $PM_{2.5}/CO$, and $PM_{2.5}/SO_2$ (Fig. S3). At MM and AV sites, the trend was decreased for $PM_{2.5}/PM_{10}$ and $PM_{2.5}/NOx$. At the PB site, the trend shows relatively constant over the studied period. To find a seasonal effect on the ratios of the pollutants across Delhi-NCR, the average values for the pollutant ratios are also presented in Table 7. The $PM_{2.5}/PM_{10}$ ratios were the highest, ranging in winter (0.53-0.64) and (0.60-0.65), and lowest in summer (0.30-0.44) and (0.28-0.35) for Delhi and NCR, respectively. In winter, the elevated ratios, combined with the high $PM_{2.5}$ concentrations and favourable meteorological conditions lead to enhanced formation of secondary particles. During summer, the lowest ratios were observed, indicating a higher fraction of coarse particle, probably due to entrainment of dust during dry and windy conditions (Chen et al., 2018; Clements, Hannigan, Miller, Peel, & Milford, 2016; Xu et al., 2017). In addition, the $PM_{2.5}/NOx$ ratio

follows the same variation as for $PM_{2.5}/PM_{10}$, i.e., highest in winter while lowest in summer and monsoon. The $PM_{2.5}/CO$ ratios (i.e. CO an excellent tracer for primary combustion sources) were found quite similar in winter and summer as opposed to lower ratios during the monsoon, presumably due to a wet deposition effect upon $PM_{2.5}$. At RKP and ROH sites, the $PM_{2.5}/CO$ ratio is apparently higher in summer than during the cold period; this suggests that the process of secondary PM formation is more significant in influencing the PM concentration in this region during summer. Moreover, the ratios of $PM_{2.5}/SO_2$, and NO_x/CO, and NOx/SO_2 are also presented in Table 7. They all show high values during the cold period, due to the high concentration of pollutants across Delhi-NCR, while in summer and monsoon relatively lower values were observed. The ratios of $PM_{2.5}/PM_{10}$ and $PM_{2.5}/NOx$ increased over the studied period at most of the monitoring sites, with

Table 7

Average values (mean ± sd) for ratios of $PM_{2.5}/PM_{10}$, $PM_{2.5}/NO_x$, $PM_{2.5}/CO$, $PM_{2.5}/SO_2$ for Delhi and NCR sites.

Delhi	season	RKP	MM	AV	DW	PB	DTU
$PM_{2.5}/PM_{10}$	Winter	0.57 ± 0.11	0.64 ± 0.13	0.53 ± 0.21	0.63 ± 0.05	0.57 ± 0.07	–
	Summer	0.44 ± 0.08	0.41 ± 0.11	0.3 ± 0.1	–	0.41 ± 0.09	
	Monsoon	0.5 ± 0.12	0.45 ± 0.2	0.32 ± 0.14	0.4 ± 0.09	0.42 ± 0.13	
	Post-monsoon	0.51 ± 0.08	0.6 ± 0.14	0.35 ± 0.16	0.5 ± 0.1	0.5 ± 0.1	
$PM_{2.5}/NO_x$	Winter	1.18 ± 0.72	1.5 ± 0.64	1.1 ± 0.8	4.6 ± 2.9	1.5 ± 0.7	4.4 ± 2.1
	Summer	1.3 ± 0.8	1.1 ± 0.5	0.7 ± 0.4	4.5 ± 3.5	1 ± 0.49	3.9 ± 3.3
	Monsoon	1 ± 0.7	0.96 ± 0.5	0.71 ± 0.14	3 ± 2.5	0.83 ± 0.5	2.5 ± 1.5
	Post-monsoon	0.9 ± 0.5	1.5 ± 1	0.8 ± 0.7	3 ± 2.7	1.4 ± 1.1	5.3 ± 3.1
$PM_{2.5}/CO$	Winter	75 ± 25	69 ± 22	102 ± 52	–	122 ± 52	–
	Summer	76 ± 42	13 ± 12	37 ± 30	154 ± 77	100 ± 52	
	Monsoon	48 ± 24	28 ± 21	49 ± 13	118 ± 80	57 ± 37	
	Post-monsoon	58 ± 25	91 ± 32	90 ± 31	157 ± 80	79 ± 41	
$PM_{2.5}/SO_2$	Winter	6.4 ± 2.6	9.4 ± 4.2	10.7 ± 6.1	18.4 ± 12	11.9 ± 7	–
	Summer	3.9 ± 2.8	5.8 ± 3.5	4.6 ± 3	13 ± 9	3.9 ± 2	
	Monsoon	5.9 ± 4.6	12 ± 7	7 ± 4.9	13 ± 10	5.7 ± 4.5	
	Post-monsoon	7.3 ± 6.5	16 ± 8	12 ± 8	15 ± 11	8.7 ± 6.4	
NO_x/CO	Winter	75 ± 24	59 ± 27	134 ± 61	68 ± 40	88 ± 40	–
	Summer	72 ± 45	–	–	56 ± 31	101 ± 38	
	Monsoon	65 ± 41	–	–	50 ± 32	80 ± 51	
	Post-monsoon	73 ± 24	52 ± 36	122 ± 42	82 ± 48	69 ± 36	
NO_x/SO_2	Winter	6.8 ± 3.6	8.6 ± 6.3	14 ± 9.2	5.9 ± 4.9	8.6 ± 4.7	–
	Summer	4.2 ± 3.4	–	8.3 ± 5.8	5.1 ± 3.2	4.4 ± 2.7	
	Monsoon	10.2 ± 11.5	–	12.1 ± 7.9	5.0 ± 2.7	7.6 ± 6.3	
	Post-monsoon	9.6 ± 6.6	9.5 ± 5.8	18.2 ± 8.5	7.0 ± 6.4	7.0 ± 5.7	
NCR		FRB	GRN	ROH	PCH	NOD	GZB
$PM_{2.5}/PM_{10}$	Winter	–	–	–	–	0.6 ± 0.1	0.65 ± 0.1
	Summer	–	–	–	–	0.28 ± 0.1	0.35 ± 0.1
	Monsoon	–	–	–	–	0.4 ± 0.1	0.3 ± 0.1
	Post-monsoon	–	–	–	–	0.5 ± 0.2	0.5 ± 0.3
$PM_{2.5}/NO_x$	Winter	3.1 ± 2.2	7.9 ± 3.2	10 ± 2.6	3 ± 1.3	2.6 ± 1.6	3.4 ± 1.7
	Summer	2.9 ± 2.6	–	–	1.9 ± 1	1.9 ± 0.7	2.9 ± 1.4
	Monsoon	2.6 ± 1.9	5.4 ± 4.0	–	2.3 ± 1.2	1.5 ± 0.5	1.5 ± 0.7
	Post-monsoon	2.6 ± 1.8	5.8 ± 3.4	11 ± 8	3.3 ± 1.4	1.8 ± 1.2	2.7 ± 1.5
$PM_{2.5}/CO$	Winter	100 ± 63	99 ± 54	99 ± 85	119 ± 67	80 ± 35	82 ± 22
	Summer	80 ± 61	–	195 ± 93	80 ± 53	45 ± 14	69 ± 30
	Monsoon	51 ± 44	111 ± 91	80 ± 49	65 ± 36	29 ± 11	33 ± 14
	Post-monsoon	83 ± 58	93 ± 42	51 ± 22	101 ± 56	60 ± 28	70 ± 27
$PM_{2.5}/SO_2$	Winter	9.3 ± 5.8	20 ± 5.5	–	11 ± 7	8 ± 3.9	5.4 ± 2.1
	Summer	14 ± 9	–	–	6.3 ± 3	2.1 ± 0.8	1.7 ± 0.6
	Monsoon	7 ± 2	11 ± 8	–	9.3 ± 4.2	1.8 ± 0.9	1.3 ± 0.
	Post-monsoon	9.5 ± 6.2	17 ± 6.8	–	10 ± 4.5	4.7 ± 3.2	3.4 ± 3.1
NO_x/CO	Winter	36 ± 23	17 ± 15.2	7.8 ± 3.2	42 ± 21	39 ± 16	27 ± 7.6
	Summer	36 ± 29	47 ± 43	–	47 ± 31	25 ± 9	26 ± 9.4
	Monsoon	21 ± 20	49 ± 41	49 ± 30	30 ± 22	21 ± 8	22 ± 8.2
	Post-monsoon	30 ± 13.7	19 ± 8.7	20 ± 17	33 ± 18	39 ± 16	28 ± 7.6
NO_x/SO_2	Winter	6.2 ± 3.1	2.9 ± 4.1	–	4.0 ± 1.9	4.3 ± 2.8	1.9 ± 1.2
	Summer	5.9 ± 3.8	3.3 ± 2.9	–	3.6 ± 1.6	1.3 ± 0.7	0.7 ± 0.4
	Monsoon	4.3 ± 3.3	5.7 ± 3.1	–	4.5 ± 4.1	1.3 ± 1.1	0.9 ± 0.3
	Post-monsoon	5.1 ± 2.9	5.0 ± 3.6	–	3.3 ± 1.6	3.1 ± 2.2	1.3 ± 0.8

highest being in winter and lowest during summer, also indicating an increase in $PM_{2.5}$ concentrations and/or reduction in PM_{10} and NO_x concentrations over the studied duration.

3.5. Bivariate polar plot and k-mean clustering of PM

The role of wind (speed and direction) on $PM_{2.5}$ and PM_{10} concentrations are examined via the bivariate polar plots (Figs. 8 and 9) for all the sites across Delhi and its NCR region. The meteorological data were obtained from each station concurrently with the PM data. The features of the polar plot were quite similar for all sites, i.e., the highest

$PM_{2.5}$ concentrations occur under very low wind speed conditions < 3 m s^{-1} and show little directional dependence. These high concentrations of $PM_{2.5}$ under stagnant atmospheric condition indicates that the local sources dominated $PM_{2.5}$ concentrations under such conditions in each region such as road transport emissions, and domestic heating. Low wind speed (calm condition) helps in the build-up and accumulation of $PM_{2.5}$ emitted by traffic around monitoring sites, resulting in high $PM_{2.5}$ concentrations. At higher wind speeds, lower $PM_{2.5}$ concentrations were recorded for all wind directions, consistent with a significant impact of local sources (e.g., traffic, domestic heating, biomass burning, and construction activities) on $PM_{2.5}$ concentrations
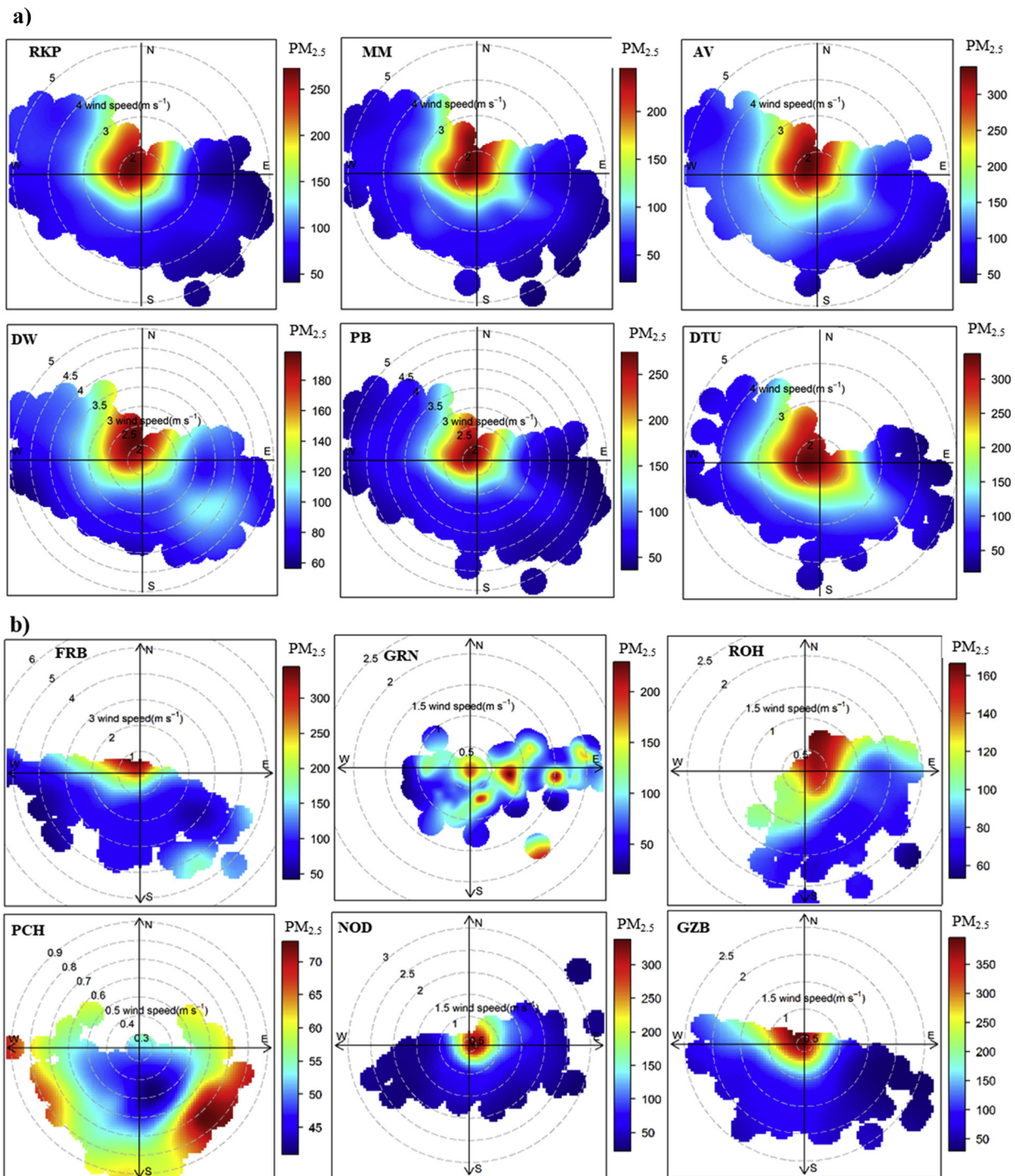
Fig. 8. Bivariate Polar Plots of $PM_{2.5}$ (µg m^{-3}) for six sites in Delhi (a), and NCR (b). The centre of each plot represents a wind speed of zero, which increases radially outward. The concentrations are shown by the colour scale. The IGI airport wind speed and direction data were used for all Delhi sites, but the wind speed and direction data were used separately for each of the NCR sites.

rather than a regional pollution source. The PCH site is the sole location showing major influences of long-range transport of $PM_{2.5}$ (Fig. 8). Fig. 9 shows a bivariate polar plot for the PM_{10} data for eight sites (only those sites which have monitored PM_{10} data) over the period. Fig. 9 reveals that high concentrations of PM_{10} were mostly associated with low wind speed conditions and when weak winds prevail along the northwest and southeast directions. PM_{10} concentrations were low when high winds are observed, particularly from the northeast and eastern quarters. Resuspension is a probable reason but it would tend to fall and then rise as the wind speed increase. The lower levels at high

winds point to local, rather than regional, sources within Delhi. In fact, the previous studies have revealed that PM_{10} source is dominated by the construction sources, associated with construction activity and dust emissions, within Delhi (Hazarika, Srivastava, & Das, 2017; Pant et al., 2015). Recent works have reported about 79 % of PM_{10} emissions originating from road dust resuspension from road dust emissions within Delhi (Nagpure, Gurjar, Kumar, & Kumar, 2016; Singh, Biswal, Kesarkar, Mor, & Ravindra, 2020). Previous studies have revealed that construction activity derived particles were estimated at about 10 % of the PM_{10} concentrations (CPCB, 2010; Guttikunda & Jawahar, 2012;
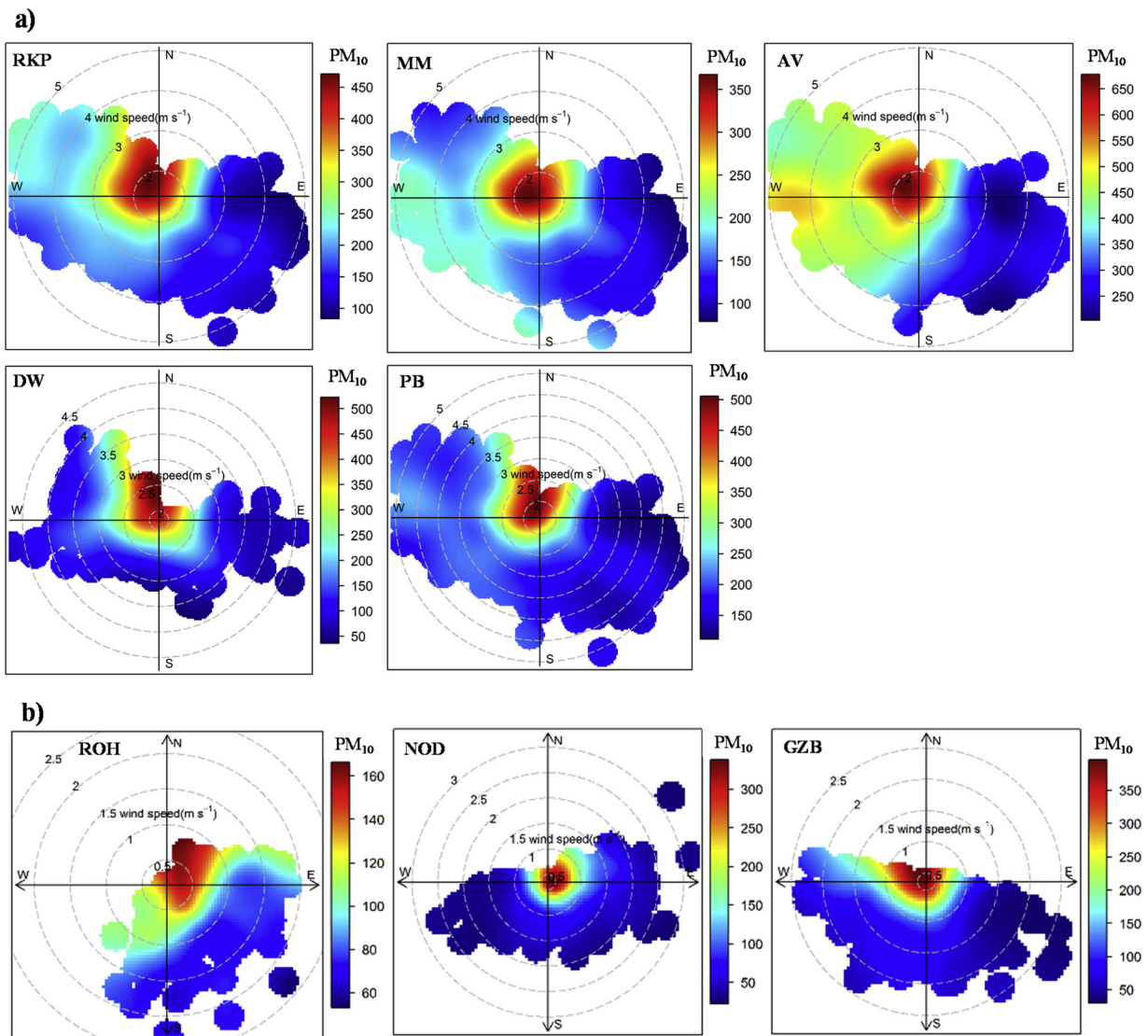
Fig. 9. Bivariate Polar Plots of PM$_{10}$ (µg m^{-3}) for (a) six sites in Delhi (a), and (b) NCR. The centre of each plot represents a wind speed of zero, which increases radially outward. The concentrations are shown by the colour scale. The IGI airport wind speed and direction data were used for all Delhi sites, but the wind speed and direction data collected at individual sites were used for each of NCR sites.

Guttikunda, Goel, & Pant, 2014).

The k-means cluster analysis has been carried out for the PM$_{2.5}$ (for MM, and NOD as an example) for clusters between 1 and 6 (Fig. 10). The method aims to minimize the sum of squared distances between all data set values and the cluster centre. This clustering identifies homogeneous groups by minimising the clustering error defined as the sum of the squared Euclidean distances between each dataset point and the corresponding cluster centre. The number of clusters is determined and shown in Fig. S4 (Lee & Kim, 2018). Moreover, after the post-processing of PM$_{2.5}$ data, it was found that the six cluster solution is appropriate for identifying local and external sources (Fig. 10). In the case the MM site, when the comparison of the polar cluster plot at MM (Fig. 10) with the polar concentrations plot at MM (Fig. 8) shows that cluster 5 mostly represents the local sources (red colour centre in Fig. 8), while cluster 4 probably represents distant sources transported to the site from the west direction. In case of the NOD site, when Fig. 10 (NOD) is compared with the polar plot (Fig. 8, NOD) it is seen that cluster 5 mostly represents the local source, while other clusters, especially cluster 6 and 3 might represent a distant source (or other sources) which come from north-easterly and south-easterly wind direction. Fig. 10 makes it feasible to match the specific wind direction clusters with the original

polar plots, thus allowing sources of PM$_{2.5}$ to be revealed in terms of the cluster. The above finding showed that PM$_{2.5}$ and PM$_{10}$ sources are dominated by the local source emissions across Delhi and its NCR region. This might be related to the regional nature of PM given the similarity between levels at different sites.

4. Summary, conclusions and future outlook

We examined the spatiotemporal characteristics of particulate matter (PM$_{10}$ and PM$_{2.5}$) and trace gases (NO$_X$, O$_3$, SO$_2$ and CO) within a network of 12 air quality monitoring stations across Delhi-NCR for the years 2014–2017. The results allowed the following conclusions:

- The concentrations of air pollutants during winter months were significantly higher than those during summer and monsoon months, with the exception of O$_3$. The annual variation of O$_3$ was opposite to other pollutants with the highest in the summer and the lowest in the winter.
- COD results showed a high homogeneity in PM$_{2.5}$ between monitoring sites within Delhi as opposed to the high spatial divergence between Delhi and NCR monitoring sites, especially PCH and ROH
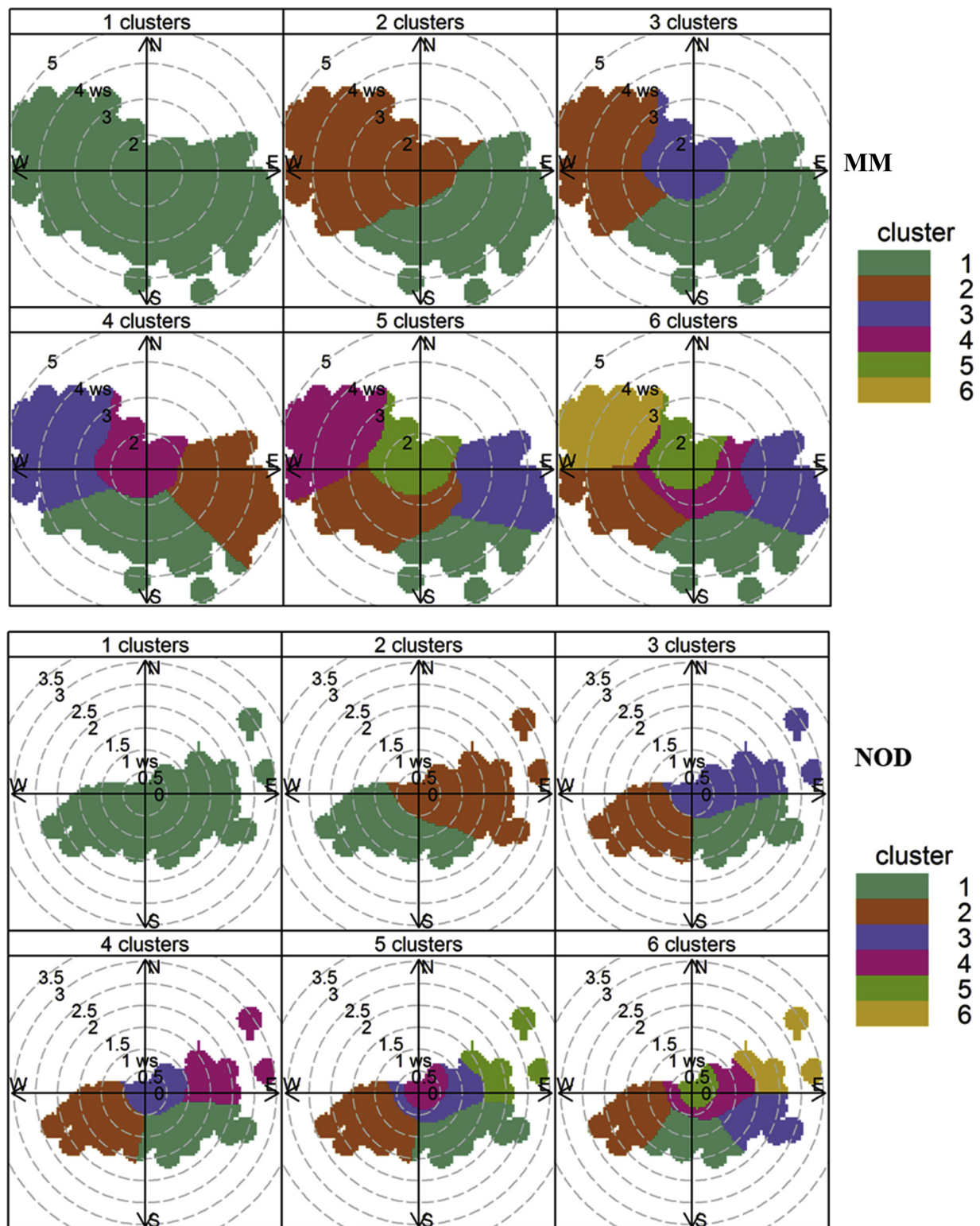
Fig. 10. A six-cluster solution obtained for $PM_{2.5}$ Bivariate polar plot (polar cluster) for two sites (MM, and NOD). The cluster for $PM_{2.5}$ calculated over the entire period. The wind speed and direction data were separately used for each site.

sites. Pearson correlation for daily $PM_{2.5}$ concentrations between the monitoring sites across Delhi –NCR region during the winter periods was higher than those during the summer period.

- The $PM_{2.5}/PM_{10}$ ratio was highest in winter (0.53-0.65), and lowest in summer (0.28-0.44) across the region. The $PM_{2.5}/NOx$ ratio follows the same pattern as $PM_{2.5}/PM_{10}$; the values were highest in winter and lowest in summer and monsoon. The average $PM_{2.5}/CO$

ratio values were found to be quite similar in winter and summer. At RKP and ROH sites, the $PM_{2.5}/CO$ is clearly higher in summer than during the winter period, indicating that the process of the secondary formation across this region also plays an important role in PM concentration.

- The relationships between air pollutants and governing meteorological parameters can be obtained through studying the bivariate

polar plots and clustering of similar features of this relationship. This technique allowed the identification of PM source contributions using relatively simple information.

The bivariate polar and polar cluster plots techniques were found to be useful in visualising the source characteristics of the different pollutants. We found the $PM_{2.5}$ concentrations in Delhi were more influenced by local sources rather than a regional source. The results from this study increase our understanding of the spatiotemporal variation and contribution of other sources to PM across the Delhi-NCR region, which can enable the development of health-related air quality policies in India. The limitations of the data and the locations of the monitoring stations within Delhi and its NCR do not in isolation allow a full understanding of the inflow and outflow of pollution and the quantification of the percentage contribution of the remotely transported $PM_{2.5}$ to the total $PM_{2.5}$ in Delhi. To obtain a holistic picture of pollutant sources across Delhi and its NCR, a more detailed data set is needed to fill a significant gap in the publicly available data. This points towards better upkeep of the existing monitoring stations, quality control of the data, and new stations covering the sub-urban areas surrounding Delhi, alongside more comprehensive measurements covering (for example) PM composition. Moreover, the majority of sites in the Delhi-NCR showed appreciably high pollutant concentrations, meaning that these despite being located outside of Delhi they may not necessarily be considered to be urban background stations. These findings also point to special considerations when setting up new monitoring stations in upwind and downwind background regions of Delhi and its NCR, to allow generation of evidence on apportionment of local versus remotely driven poor air quality in Delhi, especially during episodic conditions such as during winters (Kumar et al., 2017) or crop burning periods. The current configuration of the network does not permit the evaluation of long-range transport between Delhi and the NCR (and the vice-versa). The datasets acquired from representative local background locations, unlike the current sites which are dominated by local sources, would also allow for the deployment of techniques such as Lenschow analyses (Lenschow et al., 2001) to estimate local source contribution within Delhi via traffic and urban increments. Finally, studies into pollutant transport can be supported by specialised tools such as the Weather Research and Forecasting model with chemistry (WRF-Chem; Chen et al., 2019), Trajectory Statistical Methods (TSMs; Diémoz et al., 2019), concentration weighted trajectory (CWT; Mehmood et al., 2019), and Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT; Stein et al., 2015) over the city to understand the trajectory of regional plumes travelling towards Delhi as well as visualise and identify the transport of pollutants from outside sources of Delhi and vice-versa.

Acknowledgements

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.scs.2019.102003.

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