

# Winter Particulate Pollution over Raipur, India

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**Abstract:** Particulate air pollution during the winter season in the urban regions of India is severe due to substantial fuel and mineral combustion in adverse climatic conditions. In this work, chemical characteristics and sources of coarse particulate matter (PM<sub>10</sub>) and particulates associated chemicals during winter period of years 2006–2013 in the polluted city of Raipur, Chhattisgarh, India, are reported. The ambient air coarse particulate (PM<sub>10</sub>) concentration during the winter period of 2006–2007 ranged from 221 to 760  $\mu\text{g m}^{-3}$ . The major fraction of the PM<sub>10</sub> was composed of organic carbon, elemental carbon, iron, calcium, and sulfate. Their concentrations were remarkably reduced in the rainy season due to high wind speeds (around 10  $\text{kmh}^{-1}$ ) and removal with rain. The concentration variations and sources of PM and associated chemical species (i.e., carbons, ions, and metals) in the ambient air are discussed. DOI: [10.1061/\(ASCE\)HZ.2153-5515.0000444](https://doi.org/10.1061/(ASCE)HZ.2153-5515.0000444). © 2019 American Society of Civil Engineers.

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## Introduction

Particulate matter (PM) is composed of a wide variety of airborne materials (i.e., dust, smoke, and soot, among others), which are directly emitted into the air or result from the transformation of gaseous pollutants due to natural and anthropogenic sources (WHO 2006). PM has been reported as criteria pollutants by the USEPA (USEPA 2010). PM ( $\leq 10 \mu\text{m}$ ) is of most concern for its effects on human health, i.e., chronic lung disease and asthma, lung cancer, heart attacks, exacerbation of chronic obstructive pulmonary disease (COPD), premature death, preterm birth, and low birth weight, among other (Ferrante et al. 2015). Some metals interact with enzymes, cell components, or DNA, for example, to cause adverse

health effects (Fortoul et al. 2015). The optically active components (e.g., elemental and organic carbon and sulfate) of the particulates affect precipitation and cloud-cover events (Bell and Holloway 2007). High particulate concentrations in ambient air have caused synoptic weather in several parts of the world (Awad and Mashat 2016; Ding et al. 2016; Han et al. 2015; Sati and Mohan 2014; Guttikunda et al. 2013). The distribution, composition, and origins of particulates in ambient air of several regions have been reported (Zeb et al. 2018; Ding et al. 2016; Pan et al. 2015; Xu et al. 2015; Tao et al. 2014; Satsangi et al. 2013; Singh et al. 2013; Yadav and Satsangi 2013; Khillare and Sarkar 2012; Kulshrestha et al. 2009; Lakhani et al. 2008; Gupta et al. 2007; Yttri et al. 2007; Begum et al. 2006; Mouli et al. 2006; Oanh et al. 2006; Gupta and Kumar 2006; Duan et al. 2005; Sillanpaa et al. 2005). The aerosol dynamics (i.e., distribution, composition, sources, and impacts) in developing countries like India are complicated due to scarce aerosol data, high emissions, and severe health hazards. In this work, concentration variations, composition, and sources of the chemical species associated with ambient particulates in the most polluted city, Raipur, during the winter season of years 2006–2013 are described.

## Materials and Methods

### Study Area

The capital city of the Indian state of Chhattisgarh, Raipur (21°24'N; 81°63' E) is surrounded by coal- and mineral-based industries and was therefore selected for the proposed investigation. The Urla industrial area is spread over around 300 ha in the north-east direction, with installation of wide range of various industries. Another industrial area, Silrara, is spread over around 900 ha in the east direction and is home to industries such as sponge iron units and ferroalloy plants, among others. Similarly, many cement plants are located in the east direction of the city within an approximate 70-km radius. The largest steel plant (Bhilai) is located around 20 km away in the northwest direction from Raipur. The Borai, Durg industrial area (around 192 ha) is located approximately 40 km away from Raipur in the north–south direction. At least 300 rice mills are running in neighboring cities such as Durg,

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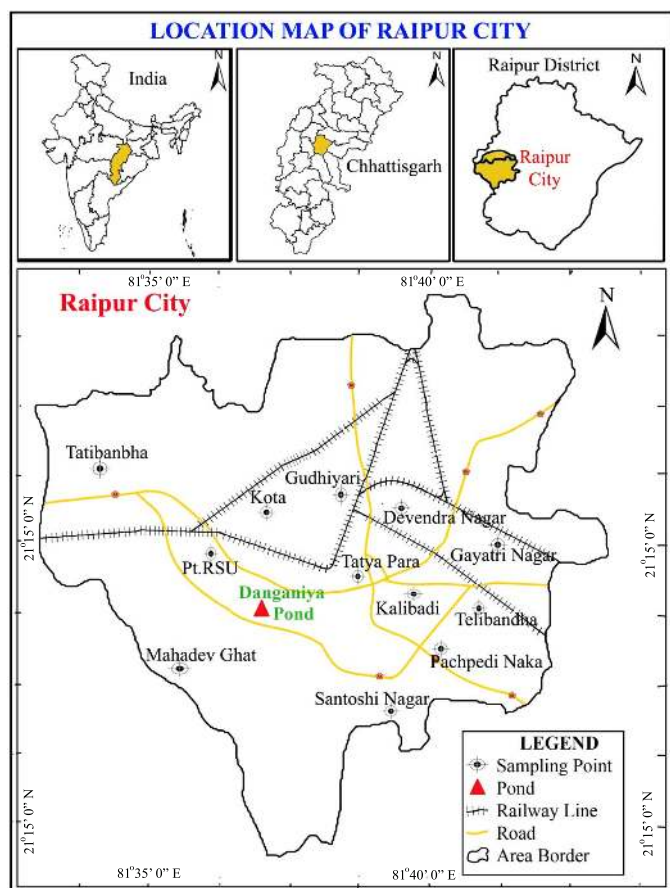
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Rajnandgaon, Tilda, Bhatapara, Arang, Rajim Nawapara, and Dhamtari, among others. A large industrial city, Korba, where several coal mines and thermal power plants operate, lies in the northeast direction around 150 km away from the city.

### Sampling of Particulate

The PM<sub>10</sub> sample was collected by using portable air sampler University of California (UC Davis) over a 47-mm (Whatmann QMA, Darmstadt, Germany) quartz filter at flow rate of 19.4 L min<sup>-1</sup>. Similarly, the suspended particulate matter (SPM) sample was collected by use of a high-volume air sampler (flow rate of 1,062.5 L min<sup>-1</sup>) on the quartz filter (Whatmann QMA, 515.6 cm<sup>-2</sup>). They were heated beforehand (600°C for 6 h) to reduce blank values. The samplers were installed on the second floor of building (above a height of around 10 m from the ground) to minimize dust originating from ground-level activities. The filter was loaded in the sampler and run to collect particulates over 12 h (6:00 a.m.–6:00 p.m.). In similar ways, one collection blank was prepared. After collection, the dismantled filter was kept in petri dish and transferred to the laboratory. Similarly, a total of 18 PM<sub>10</sub> and SPM samples were collected from 15 sites of the city in the winter period of (2006–2007, Fig. 1). For temporal studies, 15 PM<sub>10</sub> samples from three locations i.e., Kota, Kalibadi, and Sankara, were collected in the month of January during years 2008–2013. All loaded filters were kept in a petri dish and refrigerated at -4°C until the analysis.



**Fig. 1.** Map of India with location of the sampling sites in Raipur City and major possible particle sources.

### Analysis of Particulate Carbons

The thermal method was used for analysis of the organic (OC) and elemental carbon (EC) as prescribed in the literature (VDI German 1996). All carbons were converted into CO<sub>2</sub> by subsequent measurement with a nondispersive infrared (IR) detector. The sum total concentration of OC and EC was considered as total carbon (TC) in this work. The OC and EC analyses were carried out in triplicate. Their relative standard deviation (RSD) values were observed within  $\pm 3.0\%$ .

### Analysis of Elements

The proton-induced X-ray emission (PIXE) technique was used for measurement of the metal content of the filter. The Istituto Nazionale di Fisica Nucleare Tandatron (INFN, Florence, Italy) accelerator 3-MV facility in Florence, Italy, was used as external beam for the excitation of the elements (Calzolai et al. 2006). The RSD values for the triplicate analysis of the elements fell within  $\pm 5\%$ .

### Analysis of Water-Soluble Ions

The ion content of PM was leached out with deionized water (15 mL, 0.054  $\mu\text{S cm}^{-1}$ ) by sonicating at 60°C for 1 day (around 24 h). An ion chromatograph (Dionex DX120, Sunnyvale, California) equipped with anion and cation separation columns and a conductivity detector was used for analysis of the ions. An aliquot of the filtered sample (200  $\mu\text{L}$ ) was injected into the chromatography for quantifying the ions. The bicarbonate (9 mM) and methane sulfonic (20 mM) eluents were employed as leaching agents for the anions and cations, respectively. The RSD values for triplicate analysis of ions were observed between  $\pm 5\%$  and 10%.

### Quality Assurance and Quality Control

The precision of the portable air sampler was determined by using a 2,300-sequential speciation air sampler [Partisol model (Thermo Fisher Scientific Air Quality Instruments, Franklin, Massachusetts)]. Similarly, a total of six samples (2  $\times$  3) by both samplers were collected. The masses of the PM were determined by a Mettler M3 electronic (Columbus, Ohio) balance after blank correction. The RSD values ( $n = 3$ ) for PM<sub>10</sub> collections with the portable and Partisol samplers fell in between  $\pm 2.9\%$  and 2.2%, respectively. The carbon analyzer was calibrated with known quantities of propane gases and CO<sub>2</sub> for analysis of the samples. The sensitivity of the PIXE technique for studied elements was standardized by using the reference aerosol filter sample. For ion chromatographic analysis, the calibration curves were prepared by injecting different concentrations of the standard solutions (Emanuel Merck, Darmstadt, Germany) for determination of ions. The precision of the analysis in the proposed work was expressed in terms of standard deviation ( $\pm\text{SDT}$ ).

## Results and Discussion

### Meteorology

The meteorology remarkably influences the PM concentration in ambient air. The values of temperature ( $T$ ), relative humidity (RH), wind speed (WS), and sunshine (SS) during the period December 2006–February 2007 were recorded in the range of 16.7°C–23.4°C, 52%–70%, 1.5–5.9 km h<sup>-1</sup>, and 5.3–9.4 h day<sup>-1</sup> with an average value of 20°C, 59%, 2.7 km h<sup>-1</sup>, and 8.2 h day<sup>-1</sup>, respectively. The lowest wind speed (around 1.0 km h<sup>-1</sup>) was

**Table 1.** Particulate concentration in winter 2006–2007 at various locations in Raipur, India

S. No.	Sampling date	Sampling site	PM		Concentration in air ( $\mu\text{g m}^{-3}$ )
			Type	Mode	
1	December 12, 2006	Devendra Nagar (DN)	CA	PM <sub>10</sub>	696
2	January 3, 2007	Gudiyari (G)	CA	PM <sub>10</sub>	650
3	January 4, 2007	Telebandha (TL)	CA	PM <sub>10</sub>	321
4	January 6, 2007	Gayatri Nagar (GN)	RA	PM <sub>10</sub>	265
5	January 8, 2007	Santoshi Nagar (SN)	RA	PM <sub>10</sub>	577
6	January 9, 2007	Kalibadi (KB)	CA	PM <sub>10</sub>	462
7	January 10, 2007	Pachperi Naka (PN)	CA	PM <sub>10</sub>	308
8	January 12, 2007	Dangania (D)	RA	PM <sub>10</sub>	221
9	January 14, 2007	Mahadevghat Road (MR)	RA	PM <sub>10</sub>	510
10	January 15, 2007	RS University (RSU)	RA	PM <sub>10</sub>	295
11	January 16, 2007	Tatibandh (TB)	RA	PM <sub>10</sub>	304
12	January 19, 2007	Kota (K)	RA	PM <sub>10</sub>	353
13	January 21, 2007	Tatyapara (TP)	CA	PM <sub>10</sub>	420
14	February 7, 2007	Tara Industrial area (TI)	IA	PM <sub>10</sub>	384
15	February 8, 2007	Sankara (SK)	IA	PM <sub>10</sub>	760
16	February 5, 2007	Sankera (SK)	IA	SPM	1,266
17	February 5, 2007	Siltera (SL)	IA	SPM	1,150
18	February 7, 2007	Tara Industrial area (TI)	IA	SPM	1,577

Note: CA = commercial area; IA = industrial area; and RA = residential area.

observed during months of December and January. Thereafter, the wind speed steadily increased at rate of around  $1.5 \text{ km h}^{-1}$ , reaching maxima (round 10 km) in the months of June and July. After July, the wind speed again steadily decreased to its minima in the month of December. Due to the adverse climatic conditions during the months of December and January, air pollutants could not export to the outside environment and therefore increased several times higher than the mean values, causing several climatic and health hazards.

### Distribution of Particulate Matter

High PM content in the ambient air is responsible for the origin of complex environmental and health issues in urban areas. The PM concentration in the air during period from December 2006 to February 2007 is summarized in Table 1. Wide PM<sub>10</sub> ( $n = 15$ ) and SPM ( $n = 3$ ) concentration variations in the air were observed: 221–760 and 1,150–1,577  $\mu\text{g m}^{-3}$  with an average value of  $435 \pm 168$  and  $1,331 \pm 221 \mu\text{g m}^{-3}$ , respectively. The maximum PM<sub>10</sub> mass concentration in the air was seen in the industrial (i.e., Sankara) and commercial areas (i.e., Devendra Nagar and Gudhiyari). In contrast, a significant high mass concentration of PM<sub>10</sub> in sites such as Santoshi Nagar and Mahadev ghat were also observed, but these were expected due to transport of effluents from the Bhilai Steel Plant. The PM<sub>10</sub>/SPM average ratio was observed to be 0.33. The wind speed of the air mass varied around  $1.0$ – $10 \text{ km h}^{-1}$  with the minimum and maximum value in December and July, respectively. The concentration of PM<sub>10</sub> had a good negative correlation ( $r = -0.86$ ) with the wind speed of the air mass. The minimum PM<sub>10</sub> concentration in the air during July was marked due to the high wind speed ( $\geq 10 \text{ km h}^{-1}$ ) and the PM<sub>10</sub> removal with rain. The average air-quality standard for urban areas proposed by the USEPA was used as the air-quality criteria. The PM<sub>10</sub> concentration in Raipur City during the winter period was found to be around eightfold greater than the permissible limit of  $50 \mu\text{g m}^{-3}$  (USEPA 2004). Severe ambient particulate pollution in Raipur City was identified, which was much higher than values observed in other parts of the country (Gupta et al. 2007; Begum et al. 2006; Gupta and Kumar 2006; Oanh et al. 2006).

### Distribution of Carbon

The major fractions of the aerosols are composed of elemental carbon [EC or black carbon (BC)] and organic carbon. Among them, EC is an optically active component of the PM, being the second culprit for climate change after CO<sub>2</sub>. In addition, they also adsorbed heavy metals and organic compounds. The OC were composed of a variety of organic compounds of climatic and environmental interests. The concentration of OC<sub>10</sub>, EC<sub>10</sub>, TC<sub>10</sub>, OC<sub>SPM</sub>, EC<sub>SPM</sub>, and TC<sub>SPM</sub> in the air in the range of 14–109, 11–114, 25–219, 20–35, 41–101, and 64–136  $\mu\text{g m}^{-3}$ , respectively (Table 2). The OC<sub>10</sub>/OC<sub>SPM</sub> and EC<sub>10</sub>/EC<sub>SPM</sub> average ratios were found to be 1.37 and 0.60, respectively. Tremendously high concentrations of EC were observed in the industrial (i.e., Sankara) and commercial areas (i.e., Devendra Nagar and Gudhiyari), as expected due to emissions from industrial sources. The EC concentration with the PM<sub>10</sub> had fair correlation ( $r = 0.68$ ). However, the maximum OC concentration was observed in the commercial areas, i.e., Devendra Nagar and Gudhiyari, which may be due to vehicle emissions. The OC concentration with the PM<sub>10</sub> had fair correlation ( $r^2 = 0.84$ ) with the PM<sub>10</sub>. However, the concentration of OC and EC with the wind speed of air mass had a fair negative correlation ( $r = -0.81$  to  $-0.85$  to  $0.83$ ) with their lowest concentrations in the month of July, similar to the PM<sub>10</sub> scenario. The average ratios of OC<sub>10</sub>/PM<sub>10</sub> and EC<sub>10</sub>/PM<sub>10</sub> were observed to be 0.08 and 0.09, respectively. The OC<sub>10</sub>/EC<sub>10</sub> ratio was between 0.28 and 1.74, with an average value of  $0.99 \pm 0.36$ . The concentrations of the EC and OC seem to be much higher than the values observed in other locations of the country and world (Gupta et al. 2017; Costa et al. 2016; Ding et al. 2016; Xu et al. 2015; Yttri et al. 2007; Duan et al. 2005; Sillanpaa et al. 2005; Latha and Badrinath 2003).

### Distribution of Major Elements

The major fraction of the aerosols was composed of crustal elements. The ambient air concentrations of Al, P, Ti, Cr, Mn, Fe, Cu, Zn, and Zr (associated with PM<sub>10</sub>) varied as 6.5–20.3, 0.2–0.4, 1.0–2.9, 0.1–0.2, 0.5–2.4, 14–78, 0–0.2, 0–1.3, and 0–0.3  $\mu\text{g m}^{-3}$ , respectively (Table 3).

**Table 2.** Mass concentration of organic carbon (OC), black carbon (BC), and total carbon (TC)

S. No.	Concentration in air ( $\mu\text{g m}^{-3}$ )			Concentration in PM (%)		
	OC	EC	TC	OC	EC	TC
1	109	110	219	15.7	15.8	31.5
2	107	96	203	16.5	14.8	31.3
3	28	25	53	8.7	7.8	16.5
4	27	32	59	10.2	12.1	22.3
5	33	19	52	5.7	3.3	9.0
6	38	30	68	8.2	6.5	14.7
7	26	19	45	8.4	6.2	14.6
8	14	11	25	6.3	5.0	11.3
9	26	28	54	5.1	5.5	10.6
10	20	22	42	6.8	7.5	14.3
11	15	23	38	4.9	7.6	12.5
12	15	16	31	4.2	4.5	8.7
13	25	26	51	6.0	6.2	12.2
14	18	39	57	4.7	10.2	14.9
15	32	114	146	4.2	15.0	19.2
Mean PM <sub>10</sub>	36	41	76	7.7	8.5	16.2
±SD	30	35	61	3.8	4.1	7.1
16	23	41	64	1.8	3.2	5
17	20	63	83	1.7	5.5	7.2
18	35	101	136	2.2	6.4	8.6
Mean SPM	26	68	94	1.9	5.0	6.9
±SD	8	30	37	0.3	1.7	1.8

Note: SD = standard deviation.

**Table 3.** Mass concentration of trace elements in ambient air ( $\mu\text{g m}^{-3}$ )

S. No.	Al	P	Ti	Cr	Mn	Fe	Cu	Zn	Zr
1	13.0	0.2	2.0	0.1	2.4	28	0.20	1.2	0.2
2	10.2	0.4	1.6	0.1	1.3	26	0.10	0.3	0.1
3	7.3	0.2	1.2	0.1	1.0	16	0.01	0.3	0.2
4	6.5	0.3	1.0	0.1	0.5	16	0.01	0.2	0.0
5	15.6	0.3	2.5	0.1	1.2	39	0.01	0.3	0.3
6	11.3	0.4	1.6	0.1	1.0	25	0.05	0.2	0.2
7	6.8	0.2	1.2	0.1	0.5	14	0.01	0.2	0.3
8	6.5	0.3	1.0	0.1	0.8	15	0.02	0.2	0.2
9	12.9	0.3	1.9	0.2	2.2	38	0.07	0.6	0.2
10	8.4	0.2	1.2	0.1	1.1	20	0.04	0.2	0.3
11	9.7	0.4	1.5	0.1	1.1	19	0.01	0.3	0.3
12	10.2	0.3	1.6	0.2	0.8	24	0.04	0.3	0.3
13	13.7	0.3	1.9	0.1	0.9	26	0.04	0.0	0.3
14	10.6	0.4	2.0	0.1	0.6	28	0.04	0.2	0.3
15	20.3	0.3	2.9	0.2	2.2	78	0.04	1.3	0.2
Mean PM <sub>10</sub>	10.9	0.3	1.7	0.1	1.2	27.5	0.05	0.4	0.2
±SD	3.8	0.1	0.5	0.0	0.6	15.9	0.05	0.4	0.1
16	20.7	0.1	2.8	0.2	2.7	82	1.80	0.2	0.2
17	15.5	0.2	2.4	0.2	0.6	54	0.10	0.6	0.1
18	23.6	0.3	3.9	0.3	2.7	87	1.10	0.1	0.2
Mean SPM	19.9	0.2	3.0	0.2	2.0	74.3	1.0	0.3	0.2
±SD	4.1	0.1	0.8	0.1	1.2	17.8	0.9	0.3	0.1

The concentration of examined elements in the SPM varied as 15.5–23.6, 0.1–0.3, 2.4–3.9, 0.2–0.3, 0.6–2.7, 54–87, 0.1–1.8, 0.1–0.6, and 0.1–0.2  $\mu\text{g m}^{-3}$ , respectively (Table 3). Among them, the concentration of elements Al, Ti, Mn, Fe, Cu, and Zn with PM<sub>10</sub> had a fair correlation ( $r = 0.63 - 0.83$ ), unlike P, Cr, and Zr. However, their concentrations with wind speed had a negative partial correlation ( $r = -0.45$  to  $-0.47$ ). The concentration ratio of the corresponding elements (associated with SPM and PM<sub>10</sub>) was found to be 1.9, 0.7, 1.8, 2.0, 1.7, 2.7, 20, 0.8, and 1.0, respectively.

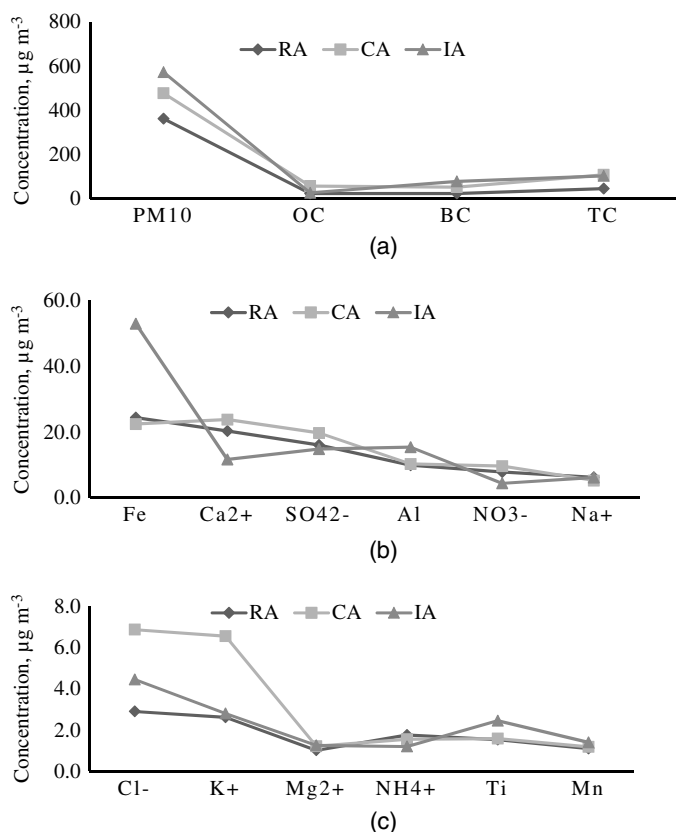
**Table 4.** Mass concentration of water aerosol soluble ions in ambient air ( $\mu\text{g m}^{-3}$ )

S. No.	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	NH <sub>4</sub> <sup>+</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>
1	3.8	15.9	1.5	26.6	1.9	38.4	14.3	12.0
2	4.2	9.3	1.5	29.4	0.8	17.7	13.5	13.9
3	5.3	2.9	1.0	21.1	1.7	14.9	8.6	3.9
4	3.0	2.4	0.7	16.4	0.8	8.5	7.6	3.1
5	10.8	2.3	1.1	31.0	0.2	11.5	5.8	3.3
6	6.2	4.7	1.2	25.7	2.9	23.2	10.5	4.2
7	6.2	3.7	1.0	20.4	1.2	12.9	6.0	3.2
8	3.8	1.7	0.6	11.8	1.2	11.0	3.5	1.0
9	5.9	3.5	1.3	30.2	4.0	28.6	13.9	2.2
10	5.9	4.0	1.3	17.6	4.6	25.7	11.5	5.1
11	6.5	1.8	0.8	12.6	0.8	12.1	6.2	2.9
12	8.1	2.6	1.3	22.9	0.7	15.5	6.7	2.7
13	6.3	2.8	1.1	19.9	0.8	11.5	5.4	4.0
14	8.0	2.4	1.2	11.9	0.9	12.7	4.6	3.4
15	4.4	3.2	1.3	11.5	1.5	17.0	4.3	5.5
Mean PM <sub>10</sub>	5.9	4.2	1.1	20.6	1.6	17.4	8.2	4.7
±SD	2.0	3.7	0.3	6.9	1.3	8.2	3.7	3.5
16	1.5	1.5	0.6	9.0	1.3	12.7	3.3	0.8
17	1.8	0.8	0.7	10.5	0.2	8.7	1.9	0.6
18	2.2	2.9	1.3	14.0	0.8	18.7	6.1	1.4
Mean SPM	1.8	1.7	0.9	11.2	0.8	13.4	3.8	0.9
±SD	0.4	1.1	0.4	2.6	0.6	5.0	2.1	0.4

Among them, the crustal elements Al, Ti, Cr, Mn, Fe, and Cu were moderately to strongly enriched in the SPM modes. The maximum concentration of the elements was seen in the Tara and Sankara industrial areas, probably due to emissions from industrial sources. Significantly high metal concentrations in the ambient air of Raipur City were halted, which were observed to be much higher than values detected in other parts of the country and world, probably due to the operation of several coal- and mineral-based industries in the study area (Pan et al. 2015; Yadav and Satsangi 2013; Khillare and Sarkar 2012; Lakhani et al. 2008; Mouli et al. 2006).

### Distribution of Water-Soluble Ions

Ions in the ambient air were contributed by both local and distant sources. Remarkably high concentrations of ions (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup>) associated with PM<sub>10</sub> were identified. These ions were water-soluble and affected the quality of the rain and surface water. The concentrations of the aforementioned investigated ions varied in the range of 1.0–3.9, 3.5–14.3, 8.5–38.4, 0.2–4.6, 3.0–10.8, 1.7–15.9, 0.6–1.5, and 11.5–31.0  $\mu\text{g m}^{-3}$ , respectively (Table 4). Similarly, concentrations of examined ions associated with SPM were in the range of 0.6–1.4, 1.9–6.1, 8.7–18.7, 0.2–1.3, 1.5–2.2, 0.8–2.9, 0.6–1.3, and 9.0–14.0  $\mu\text{g m}^{-3}$ , respectively. All ions except Na<sup>+</sup> and NH<sub>4</sub><sup>+</sup> had a poor correlation with the PM<sub>10</sub> concentration ( $r = 0.36 - 0.73$ ). Their concentration with wind speed had a partial negative correlation ( $r \approx -0.45$ ), with the minimum values in July, unlike Na<sup>+</sup> and NH<sub>4</sub><sup>+</sup> because they were contributed by other sources. PM<sub>10</sub>/SPM concentration ratios of 5.2, 2.2, 1.3, 2.0, 3.3, 2.5, 1.2, and 1.8 for Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup> were observed. All ions were moderately to significantly enriched in the coarse modes. The  $\sum$  PM<sub>10</sub> WSI<sub>8</sub> concentrations varied from 16.9 to 309  $\mu\text{g m}^{-3}$  with an average value of  $119 \pm 108 \mu\text{g m}^{-3}$ . Significant higher loading of the ions in the air of commercial and residential locations (i.e., Devendra Nagar, Gudhiyari, Santoshi Nagar, and Mahadev Ghat) were observed. The concentration of SO<sub>4</sub><sup>2-</sup> with the NO<sub>3</sub><sup>-</sup> in the PM<sub>10</sub> had a fair correlation ( $r = 0.81$ ), which may



**Fig. 2.** Spatial variation of PM and associated species. RA = residential area; CA = commercial area; IA = industrial area; OC = organic carbon; BC = black/elemental carbon; and TC = total carbon.

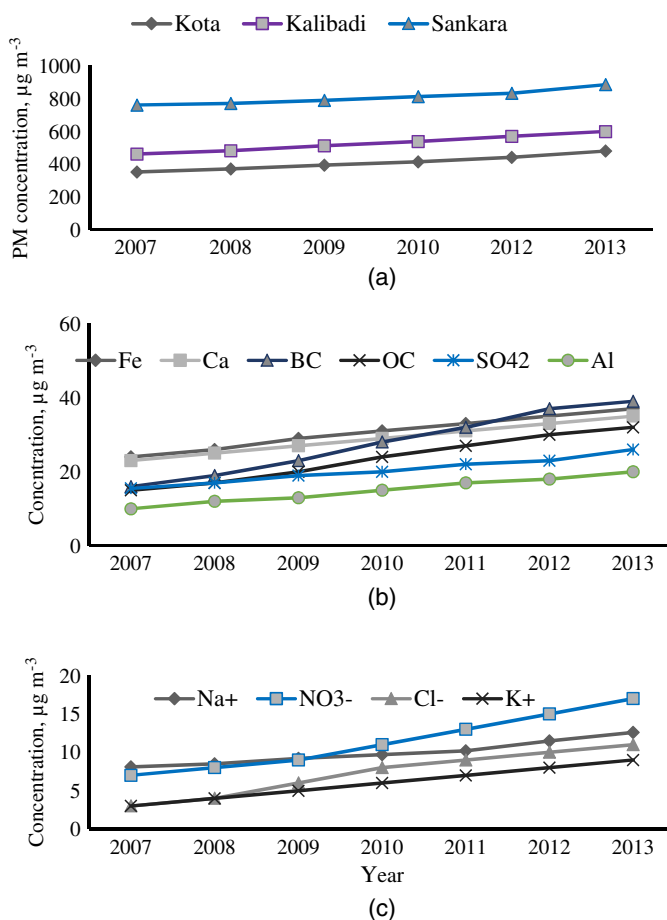
be due to their formation by similar processes. The  $\sum$  anion/ $\sum$  cation average equivalent ratio for the PM<sub>10</sub> and SPM was 0.40 and 0.45, respectively, indicating the alkaline nature of the air. The relative abundance of 7%, 13%, 27%, 3%, 9%, 7%, 2%, and 32% for Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup> in the PM<sub>10</sub> was identified. The major fraction (59%) of the PM was contributed by ions SO<sub>4</sub><sup>2-</sup> and Ca<sup>2+</sup>. Similar concentration ranges for these ions (associated with the coarse particulates) as those recorded for the ambient air of other parts of the country and China were observed (Tao et al. 2014; Satsangi et al. 2013; Singh et al. 2013; Kulshrestha et al. 2009).

### Spatial Distribution

Concentrations of the chemical species related to PM in the air are affected by the meteorology and geography of the area. Higher concentrations of carbon and metals were observed in the industrial areas, probably due to coal and mineral combustion emissions (Fig. 2). In contrast, the highest concentrations of species OC, Ca, K, Cu, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> were seen in the commercial areas, probably due to contributions from vehicular emissions and atmospheric inputs. However, the higher concentration of two species, Na and NH<sub>4</sub><sup>+</sup>, was marked in the residential area, which may be due to input from municipal wastes.

### Temporal Variations

The concentrations of PM and associated species in the air are related to the frequency and strength of emission inventories. The vast industrialization and urbanization of Raipur City is occurring due to the easily available of natural resource materials,



**Fig. 3.** Temporal variation of PM and associated elements. OC = organic carbon; and BC = black/elemental carbon.

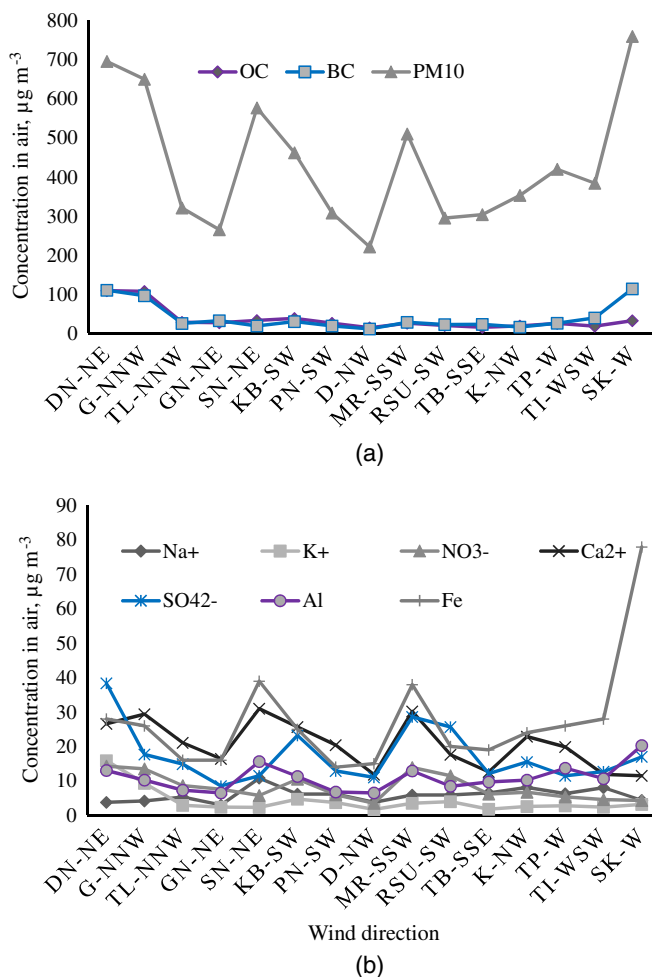
i.e., coal, biomass, and minerals. The concentrations of PM and associated species were found to increase during 2006–2013, as expected due to increases in the quantum of population, industry, vehicles, and construction work, as shown in Fig. 3.

### Composition of Aerosols

The urban aerosols are composed of carbons, metals, ions, and organics, among others. The fraction of OC, EC (BC), Fe, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, Al, NO<sub>3</sub><sup>-</sup>, Na<sup>+</sup>, Cl<sup>-</sup>, K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Ti, Mg, Mn, P, Zn, Zr, Cr, and Cu in the PM<sub>10</sub> ( $n = 15$ ) was found to fall in the ranges of 4.2%–16.5%, 3.3%–15.8%, 4.0%–10.3%, 1.5%–6.2%, 2.0%–8.7%, 1.6%–3.3%, 0.6%–3.9%, 0.6%–2.3%, 0.1%–2.1%, 0.4%–2.3%, 0.03%–1.6%, 0.3%–0.5%, 0.2%–0.4%, 0.2%–0.4%, 0.03%–0.14%, 0.01%–0.17%, 0.01%–0.10%, 0.01%–0.06%, and 0.01%–0.03% with average values of 7.7%, 8.5%, 6.2%, 5.1%, 4.2%, 2.6%, 2.0%, 1.5%, 1.0%, 0.9%, 0.4%, 0.4%, 0.3%, 0.3%, 0.08%, 0.08%, 0.06%, 0.03%, and 0.01%, respectively. However, in the SPM, the fraction of all species except Cu was found in the lower orders. The total fraction of the carbons, metals, and water-soluble ions in the PM<sub>10</sub> and SPM estimated was found to be 41.4% and 22.1%, respectively. The uncertainty observed in the PM<sub>10</sub> and SPM was 58.6% and 77.9%, respectively.

### Sources

Local industries, including Bhilai Steel Plant, fully encircle Raipur City. The highest concentrations of PM<sub>10</sub>, EC, K<sup>+</sup>, Fe, and Al were



**Fig. 4.** Influence of wind direction in distribution of PM and associated species. NE = northeast; NNW = north–northeast; NE = north east; SW = southwest; NW = northwest; SSW = south–southwest; SSE = south–southeast; NW = northwest; W = west; and WSW = west–southwest.

found when the air masses come from the northeast direction due to influence of local industries (Fig. 4). The significant concentration of OC and Cl<sup>-</sup> was seen when the air masses come from the north–northwest direction due to industrial influence of the Bhilai Steel Plant. The remarkable high mass concentration of the secondary aerosol species, i.e., NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, was seen when the air masses flowed from the south–southwest direction due to thermal power plants running in Nagpur and Chandrapur areas. The highest concentration of Ca<sup>2+</sup> was assumed to be due to running of several rice mills in the south–southwest direction of the city.

The widely used principle component analysis-multilinear regression analysis (PCA-MLRA) was used to apportion the emission sources contributing to the ambient particulates in Raipur (USEPA 1997; John et al. 1994). Four factors (Table 5) were identified as per criteria of cumulative percentage variance >80% and eigenvalue of >2 (Stevens 1996). The Varimax rotated factor analysis showed four possible different sources for the OC, BC, metallic elements (ME), and water soluble ions (WSI). Factor 1 comprised of anthropogenic sources with influence of traffic for OC, BC, K<sup>+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and Cl<sup>-</sup> with a variance of 37%. A substantial amount of coal (>10 Mt year<sup>-1</sup>) was burned in Raipur City and surrounding areas by various industries. This means that the pollutants originated by the fuel combustion as well

**Table 5.** Factor loadings (varimax normalized)

Species	Factor 1	Factor 2	Factor 3	Factor 4
OC	0.95	0.06	-0.02	0.06
BC	0.70	0.41	-0.41	-0.17
Na <sup>+</sup>	-0.20	0.13	0.86	0.12
K <sup>+</sup>	0.89	0.04	0.02	0.28
Mg <sup>2+</sup>	0.62	0.30	0.30	0.35
Ca <sup>2+</sup>	0.43	0.02	0.57	0.44
NH <sub>4</sub> <sup>+</sup>	-0.03	0.03	0.04	0.87
SO <sub>4</sub> <sup>2-</sup>	0.54	0.19	-0.03	0.74
NO <sub>3</sub> <sup>-</sup>	0.61	-0.10	0.24	0.67
Cl <sup>-</sup>	0.90	0.07	0.21	0.08
Al	0.08	0.94	0.13	-0.03
P	0.02	0.11	0.47	-0.51
Ti	0.08	0.93	0.15	-0.10
Cr	-0.03	0.62	-0.42	0.31
Mn	0.27	0.69	-0.43	0.32
Fe	0.06	0.93	0.09	-0.06
Cu	-0.11	0.09	-0.81	0.06
Zn	0.43	0.68	-0.12	0.07
Variance	5.42	4.33	2.77	2.69
Total	0.28	0.23	0.15	0.14

as blown dust. Factor 2 included mineral sources for Al, Ti, Cr, Mn, Fe, and Zn with a variance of 19%. This pattern of metals indicated the originated from the roasting of the raw materials and blown dust. Factor 3 traced a consistent source for Na<sup>+</sup> and Ca<sup>2+</sup> with a variance of 14%. Mainly three sources, i.e., roasting of pyrite and dolomite, fossil fuel combustion, and blown dust, contributed to a continuous presence of Na<sup>+</sup> and Ca<sup>2+</sup> in the coarse particulates. Factor 4 predicted secondary sources for specie NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup> with a variation of 11%.

## Conclusion

The severe particulate air pollution (more than eightfold higher than the prescribed value), especially in the period December to January in Raipur City, India, was observed to be due to adverse climatic weather. The particulates were composed of high fractions of BC (9.4%), OC (11.2%), metals (9.2%), and water-soluble ions (26.2%). Anthropogenic activities and atmospheric transformations were expected to be major sources of PM and associated species in the air. Wind speed was observed as one major meteorological factor responsible for the halting of winter particulate pollution.

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