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Characteristics and Sources of Water-Soluble Ionic Species Associated with PM_{10} Particles in the Ambient Air of Central India

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Abstract PM_{10} aerosol samples were collected in Durg City, India from July 2009 to June 2010 using an Andersen aerosol sampler and analyzed for eight water-soluble ionic species, namely, Na^+, $NH_4{}^+$, K^+, Mg^{2+}, Ca^{2+}, Cl^-, $NO_3{}^-$ and $SO_4{}^{2-}$ by ion chromatography. The annual average concentration of PM_{10} (253.5 ± 99.4 μg/m^3) was four times higher than the Indian National Ambient Air Quality Standard of 60 μg/m^3 prescribed by the Central Pollution Control Board, India. The three most abundant ions were $SO_4{}^{2-}$, $NO_3{}^-$, and $NH_4{}^+$, with average concentrations of 8.88 ± 4.81, 5.63 ± 2.22, and 5.18 ± 1.76 μg/m^3, respectively, and in turn accounting for 27.1 %, 16.5 %, and 15.5 % of the total water-soluble ions analyzed. Seasonal variation was similar for all secondary ions i.e., $SO_4{}^{2-}$, $NO_3{}^-$, and $NH_4{}^+$, with high concentrations during winter and low concentrations during monsoon. Varimax Rotated Component Matrix principal component analysis identified secondary aerosols, crustal resuspension, and coal and biomass burning as common sources of PM_{10} in Durg City, India.

Keywords Atmospheric aerosols · Water-soluble ions · Seasonal variation · Source identification

The awareness of air pollution has led to numerous studies on the chemical composition of ambient aerosols and the identification of pollution sources (Sun et al. 2004; Klejnowski et al. 2012). Deterioration of urban air quality has now become an increasing and widespread concern both in the developed and developing countries of Asia (Chelani et al. 2010; Singh and Sharma 2011). Rapid industrialization, urbanization and increasing population are polluting the basic resources required to sustain life (Nicolas et al. 2009; Deshmukh et al. 2012a; Pandey et al. 2012). Several studies have revealed that there is a significant correlation between airborne particulate levels and adverse health effects (Stone et al. 2010; Pipalatkar et al. 2012). Medical researchers have defined the respirable fraction as those particles with diameter <10 μm and hence 'PM_{10}' has received worldwide attention for its adverse impacts on human health, visibility, and global climate (Pope 2000; Tsai 2005). The rates of increase of aerosol concentrations in developing countries such as India are higher than those in developed countries all over the world (Mauli et al. 2006; Mohanraj et al. 2011). Indian cities are among the most polluted in the world and most of the time levels of aerosols exceed National Ambient Air Quality Standards (NAAQS) specified by the Central Pollution Control Board (CPCB), India (CPCB 2009) as well as air quality guidelines of the World Health Organization (WHO 2005) (Bhaskar and Mehta 2010; Deshmukh et al. 2012b; Pandey et al. 2012). Several studies have focused on chemical characterization and source identification of aerosols in the cities of India (Mohanraj et al. 2011; Singh and Sharma 2011; Deshmukh et al. 2012a; Pandey et al. 2012) and

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abroad (Wang et al. 2003; Sun et al. 2004; Nicolas et al. 2009; Huang et al. 2010; Guo et al. 2012), but PM_{10} has not been characterized for its emission sources in Durg City, India. Therefore, the present study was carried out to determine the mass levels and seasonal variation of water-soluble ionic species associated with PM_{10} particles in Durg City, India between July 2009 and June 2010. Moreover, water-soluble ionic composition data derived from the study was analyzed using a principal component analysis (PCA) receptor model technique to identify the possible sources contributing to the PM_{10} concentration in Durg City, India.

Durg City (20°23′–22°02′ N and 80°46′–81°58′ E), India, (Fig. 1) is renowned for the existence of one of Asia's largest steel plants, Bhilai Steel Plant (BSP) in Bhilai City, Chhattisgarh, and has a population of ~0.6 million and an average population and vehicular growth rate of 3.24 % and 4.42 % per year, respectively (Deshmukh et al. 2012b). The sampling site was located in front of the Government Science College, in an area encompassed by educational and research institutes, whereas the wider area is crowded with different kinds of industries such as steel plants, cement plants, sponge iron plants and power plants. The closest street with significant road traffic, mainly trucks, buses, cars and 2-stroke automobiles, is about 200 m away from the sampling point. Further details of the sampling site are given elsewhere (Deshmukh et al. 2012b).

Materials and Methods

PM_{10} samples ($n = 120$) were collected with an Andersen aerosol sampler (Model TE 20–800, USA) between July 2009 and June 2010. The sampler was operated on the terrace of a double-storied building at a height of ~15 meters above ground level. Samples were collected on pre-weighed Whatman 41 filters, conditioned at 20°C and 40 % relative humidity for ~24 h, at a flow rate of 28.3 L/min. Each sampling was run for 24 h. Samples were collected on every third day throughout the year covering all four seasons; monsoon (July–September), winter (October–January),

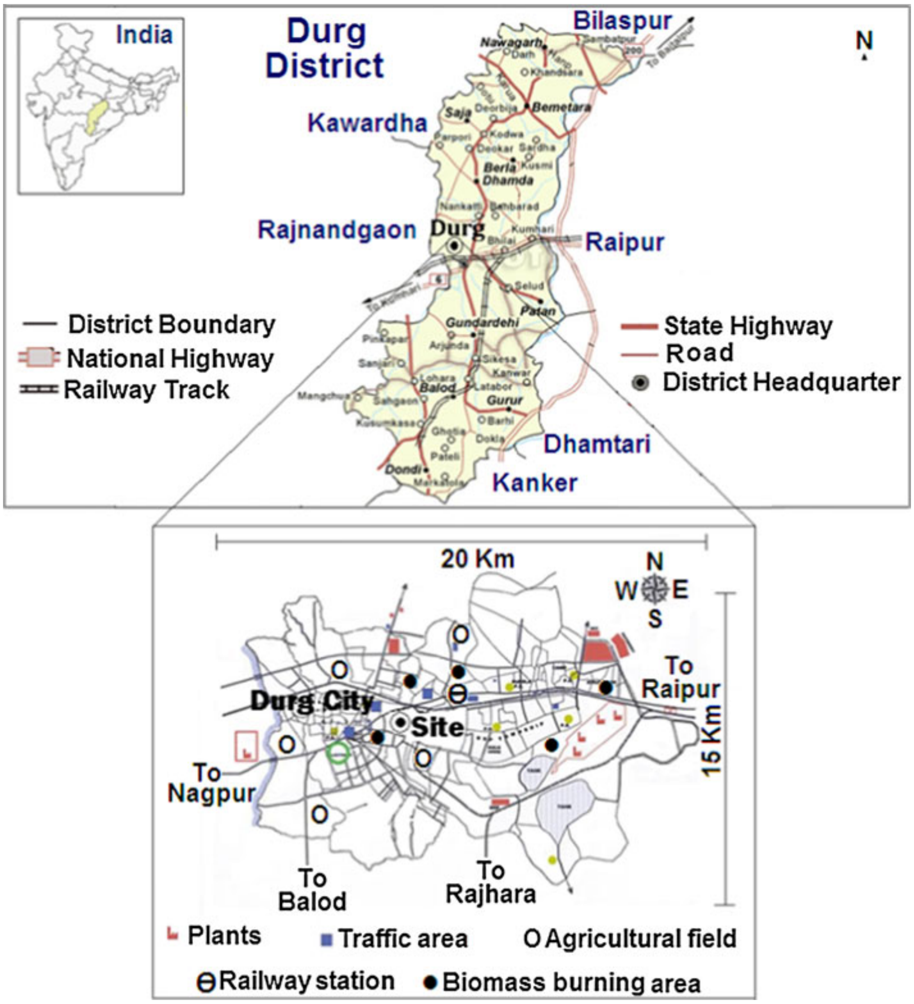

Fig. 1 Location of sampling site in Durg City, India

spring (February–March) and summer (April–June). Field blanks ($n = 24$) were also collected to quantify and subtract artifacts due to adsorption of gas-phase organic components onto the filters. After the collection of aerosol samples, filters were again conditioned at 20°C temperature and a relative humidity of 40 % for ~24 h and then weighed three times using a Sartorius electronic balance (Model CP225D) with a reading precision of 10 µg.

One quarter of each filter sample was used in the extraction. Samples were extracted into 10 mL of ultrapure water (specific resistance: 18 MΩ cm) by using an ultrasonic bath (UT–105 S, Sharp, Japan) and a shaker (EYELA Multi Shaker, Rikakikoi Co., Tokyo, Japan) for 45 min. The extracted solution was filtered through a microporous membrane (pore size, 0.45 µm; diameter, 25 mm), and filtrate was stored in a refrigerator at ~4°C until chemical analysis. An ion chromatograph (Dionex, DX–120, USA) was used to measure Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} in the aqueous extract. Analysis of water-soluble ions was performed using separation columns Ionpac AS 12A and CS 16A, respectively, for anions and cations. Eluents of 2.7 mM Na_2CO_3 and 0.3 mM $NaHCO_3$ (Wako, Japan) were prepared and used for the detection of anions with a pump flow rate of 1.5 mL/min. A 2.6 mM methane sulphonic acid (CH_3SO_3H) solution was used as an eluent for cation analysis with a pump flow rate of 1 mL/min.

The method detection limits (MDLs) were calculated for Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} from 1 ppm laboratory prepared calibration standards. The obtained MDLs were 10.3 µg/L for Na^+, 7.9 µg/L for NH_4^+, 4.2 µg/L for K^+, 18.4 µg/L for Mg^{2+}, 7.4 µg/L for Ca^{2+}, 6.9 µg/L for Cl^-, 7.8 µg/L for NO_3^- and 4.8 µg/L for SO_4^{2-}. The precisions estimated from the standard deviation of repeated measurements of standards and samples were found to be 3.28 %, 1.36 %, 2.64 %, 4.89 %, 2.44 %, 2.04 %, 2.58 % and 1.54 % for Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}, respectively. Recovery results were 103.3 % for Na^+, 99.7 % for K^+, 96.9 % for NH_4^+, 100.1 % for Mg^{2+}, 102.6 % for Ca^{2+}, 101.4 % for Cl^-, 98.5 % for NO_3^- and 100.3 % for SO_4^{2-}. Blank filters were subjected to the same extraction and preparation procedures as for the regular particulate sample filters. The overall mean concentrations of field blank samples collected over the year were 0.03 µg/m^3 for Na^+, 0.06 µg/m^3 for NH_4^+, 0.01 µg/m^3 for K^+ and Mg^{2+}, 0.08 µg/m^3 for Ca^{2+}, 0.04 µg/m^3 for Cl^-, 0.00 µg/m^3 for NO_3^- and 0.14 µg/m^3 for SO_4^{2-}. All the reported particulate and water-soluble ion concentrations were corrected to field blanks.

Meteorological data for the entire study period was obtained from the Department of Agrometeorology, Indira Gandhi Agricultural University, Raipur, Chhattisgarh. January was a cold winter month with a mean temperature of 16.2°C, and May contained the maximum mean summer temperature of 33.6°C. The relative humidity varied from a minimum of 11 % in April to a maximum of 96 % in August. The study region receives heavy rainfall during monsoon but very little during summer and winter. The prevailing winds were south-west with wind speed varying from 1.0 m/s (January) to 12.7 m/s (July). The annual mean wind speed was 4.8 m/s. Wind rose plots (Fig. 2) generated using a WRPLOT View (Lakes Environmental, Canada) show that winter and spring are relatively calm seasons (71.4 % and 66.3 %) compared with summer (17.9 %) and monsoon (16.8 %).

Results and Discussion

The annual average concentration of PM_{10} was 253.5 ± 99.4 µg/m^3 which is more than 5, 12 and 4 times higher than USEPA (50 µg/m^3; USEPA 2002), WHO (20 µg/m^3; WHO 2005), and NAAQS of India (60 µg/m^3; CPCB 2009) annual guidelines, respectively. The 24-h mean concentration of PM_{10} in Durg City, India, exceeded USEPA (150 µg/m^3; USEPA 2002), WHO (50 µg/m^3; WHO 2005) and NAAQS of India (100 µg/m^3; CPCB 2009) 24-h guidelines 95%, 100 % and 97 % of the time, respectively. The concentration of PM_{10} was found to be highest during winter followed by spring and lowest during monsoon season. Further details of the mass concentration and seasonal variation of PM_{10} found in Durg City, India, is given elsewhere (Deshmukh et al. 2012b).

Total water-soluble inorganic ions (TWSII) ranged from 2.66 to 70.64 µg/m^3 and averaged 26.87 µg/m^3 (Table 1). The percentage distribution of individual water-soluble inorganic ions to PM_{10} mass and TWSII are shown in Fig. 3. The analyzed TWSII amounted to, on average, 10.1 % (1.1 %–21.4 %) of PM_{10} mass, 6.1 % (0.2 %–18.5 %) anions and 3.9 % (0.9 %–11.5 %) cations. Of all anions analyzed, SO_4^{2-} was the dominant component followed by NO_3^- and Cl^-. The concentration of SO_4^{2-} and NO_3^- varied from 0.22–28.68 µg/m^3 and 0.10–22.84 µg/m^3 with an average of 8.88 and 5.63 µg/m^3, respectively. On average, SO_4^{2-} and NO_3^- accounted for 3.1 % and 1.9 % of PM_{10} mass and 27.1 % and 15.3 % of TWSII analyzed, respectively. The major sources of SO_4^{2-} and NO_3^- in the atmosphere are the oxidation of their gaseous precursors, SO_2 and NO_x, emitted from various anthropogenic activities (Guo et al. 2012). With respect to cations, NH_4^+ was the dominant component followed by Ca^{2+} and Na^+. The concentration of NH_4^+ and Ca^{2+} varied from 0.45–8.35 µg/m^3 and 1.00–5.99 µg/m^3 with an average of 5.18 and 2.53 µg/m^3, respectively. NH_4^+ and Ca^{2+} amounted to approximately 1.3 % and 1.1 % of the PM_{10} mass, and 15.5 % and 16.5 % of TWSII analyzed. The

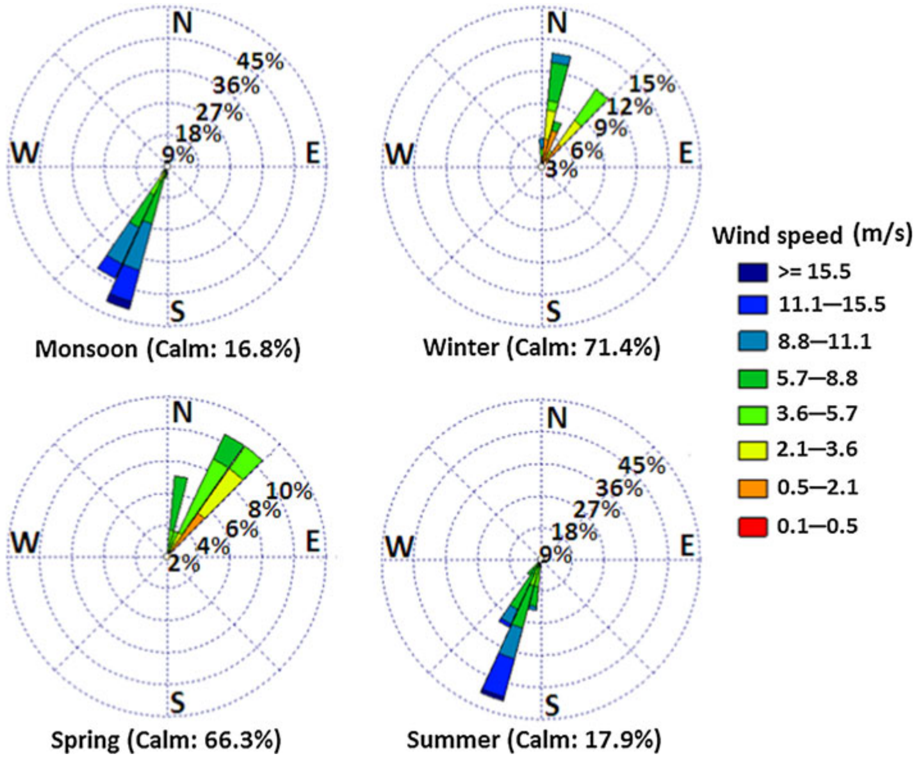

Fig. 2 Wind rose plots for monsoon, winter, spring and summer seasons during the study period in Durg City, India

Table 1 Annual mean, standard deviation, minimum, maximum and coefficient of variation of water-soluble ion concentration (μg/m^3) between July 2009 and June 2010 in Durg City, India

Species	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Total ions	Total cations	Total anions
Mean	1.75	5.18	0.87	0.80	2.53	3.23	5.63	8.88	26.87	9.14	17.73
Standard deviation	0.98	1.76	0.59	0.64	1.08	2.99	2.22	4.81	4.81	4.57	8.81
Minimum	0.27	0.45	0.11	0.10	1.0	0.03	0.10	0.22	2.66	2.09	0.41
Maximum	8.56	8.35	2.23	4.44	5.99	9.78	22.84	28.68	70.64	22.61	61.17
Coefficient of variation	0.56	0.33	0.67	0.80	0.42	0.92	0.39	0.54	0.12	0.50	0.50

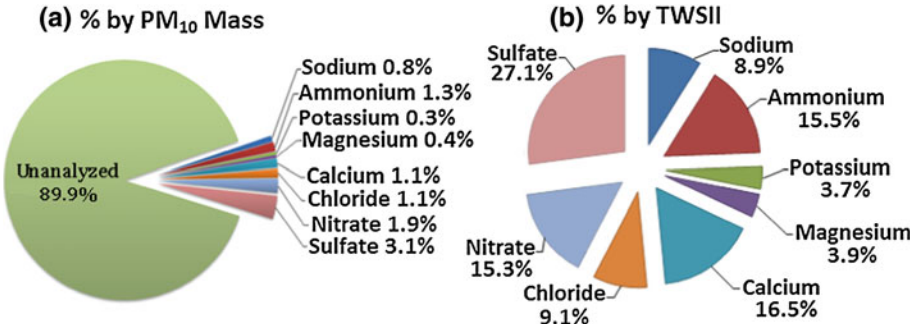

Fig. 3 Percentage distribution of water-soluble inorganic species in overall PM$_{10}$ mass

annual average concentrations of Cl^-, Na^+, K^+ and Mg^{2+} were 3.23, 1.75, 0.87 and 0.80 μg/m^3, respectively, and these four ions combined to account for 2.7 % of the PM$_{10}$ mass and 25.6 % of the TWSII analyzed.

A comparison of the mass concentrations of water-soluble ions associated with PM$_{10}$ found in Durg City, India, was made with those reported at different sites across the world and results are shown in Table 2. Concentrations of the major ions NO_3^- and SO_4^{2-} in Durg City, India, were lower than those found in Delhi and Agra, India and Nanjing and Beijing in China but higher than those reported for Tirupati in India, Southern Spain, Taipei in

Table 2 Average mass concentration (μg/m^3) of water-soluble ions in PM_{10} aerosols at different sites across the world

Area and place	Study period	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	References
Durg, India	Jul 2009–Jun 2010	1.75	5.18	0.87	0.80	2.53	3.23	5.63	8.88	This study
Raipur, India	Jul 2009–Jun 2010	1.76	2.92	0.76	0.84	3.02	3.46	5.52	9.98	Deshmukh et al. (2012a)
Delhi, India	Sep 2005–Jan 2006	5.76	6.06	4.11	1.30	6.82	8.23	15.13	16.74	Chelani et al. (2010)
Mumbai, India	Apr 2007–Mar 2008	3.16	5.69	2.19	2.88	5.67	3.31	2.38	12.68	Gupta et al. (2012)
Agra, India	Mar 2007–Feb 2008	3.97	9.14	4.36	1.71	7.23	2.95	12.90	11.32	Singh and Sharma (2011)
Tirupati, India	Oct 2001–Sep 2002	3.37	0.19	0.42	0.19	1.47	1.06	1.05	5.72	Mauli et al. (2006)
Beijing, China	2003–2004	1.60	11.9	1.74	2.04	9.05	2.69	21.1	24.8	Sun et al. (2004)
Nanjing, China	Feb 2001–Dec 2001	3.77	10.81	3.41	0.38	4.58	1.51	9.19	18.13	Wang et al. (2003)
Southern Spain	Dec 2004–Nov 2005	0.99	1.10	0.28	0.16	2.29	0.60	3.76	4.36	Nicolas et al. (2009)
Taipei, Taiwan	May 2011–Nov 2011	0.65	1.49	0.22	0.20	0.37	0.54	1.32	6.30	Gugamsetty et al. (2012)
Ishikawa, Japan	Jan 2001–Dec 2003	1.86	1.31	0.12	0.23	0.25	2.77	1.77	4.49	Guo et al. (2012)

Taiwan and Ishikawa in Japan. The concentration of Na^+ in Durg City, India, was similar to that reported for Raipur City, India. It was lower than that found in other Indian cities, namely Delhi, Mumbai, Agra and Tirupati, and in Nanjing in China and Ishikawa in Japan, but higher than that reported in Southern Spain and Taipei, Taiwan. The concentration of NH_4^+ in Durg City, India, was lower than that found in Delhi, Mumbai, and Agra in India and Beijing and Nanjing in China, but higher than that found in Raipur, and Tirupati in India, Southern Spain, Taipei in Taiwan and Ishikawa in Japan. The concentration of K^+ in Durg City, India, was lower than that found in Indian cities such as Delhi, Mumbai and Agra and Chinese cities such as Beijing and Nanjing but higher than that reported for Tirupati in India, Southern Spain, Taipei in Taiwan and Ishikawa in Japan. Finally, concentrations of Mg^{2+}, Ca^{2+} and Cl^- in Durg City, India were lower than those reported for Raipur, Delhi and Mumbai City in India and higher than those found in Southern Spain, Taipei in Taiwan and Ishikawa in Japan.

The seasonal concentrations and monthly variations of water-soluble inorganic ions are given in Table 3 and Fig. 4, respectively. The concentrations of major water-soluble ions such as SO_4^{2-}, NO_3^- and NH_4^+ were highest during winter followed by spring and lowest during monsoon. Higher concentrations during winter can be attributed to increased anthropogenic activities; vehicular traffic, fossil fuel burning and industrial activities. NO_3^- is generally formed from the oxidation of NO_x and its subsequent reaction with NH_3, which leads to formation of ammonium nitrate (NH_4NO_3) particles (Wang et al. 2006), and most NO_3^- and SO_4^{2-} exist as ammonium nitrate (NH_4NO_3) and ammonium sulphate [$(NH_4)_2SO_4$] (Han et al. 2008). The lower temperature during winter favors the shift from gas phase of nitric acid to the particle phase of nitrate, which could have led to the high concentration of NO_3^- during winter season. Conversely, lower concentration of NO_3^- during summer may be due to the volatilization of NH_4NO_3, which increases with increase in temperature and decrease in relative humidity (Mauli et al. 2006). The

Table 3 Seasonal statistics of PM_{10} and water-soluble ions concentration (μg/m^3) in Durg City, India

Species	Monsoon ($n = 30$) Mean ± SD[a] (Min., Max.)	Winter ($n = 40$) Mean ± SD[a] (Min., Max.)	Spring ($n = 20$) Mean ± SD[a] (Min., Max.)	Summer ($n = 30$) Mean ± SD[a] (Min., Max.)
Na^+	0.46 ± 0.14 (0.27, 0.70)	0.76 ± 0.19 (0.44, 1.11)	4.44 ± 1.93 (2.56, 8.56)	2.55 ± 1.23 (1.13, 5.49)
NH_4^+	2.22 ± 0.78 (0.45, 2.62)	7.50 ± 1.39 (2.82, 8.35)	3.83 ± 1.06 (2.34, 5.30)	2.96 ± 1.48 (1.12, 5.86)
K^+	0.35 ± 0.28 (0.11, 0.78)	1.44 ± 0.40 (0.89, 2.23)	1.12 ± 0.30 (0.59, 1.46)	0.49 ± 0.30 (0.11, 1.06)
Mg^{2+}	0.12 ± 0.04 (0.10, 0.16)	0.16 ± 0.03 (0.10, 0.29)	2.11 ± 1.07 (1.16, 4.44)	1.46 ± 0.78 (0.56, 3.46)
Ca^{2+}	1.61 ± 0.73 (1.00, 3.47)	2.36 ± 0.53 (1.64, 3.61)	3.70 ± 1.28 (1.88, 5.99)	2.90 ± 0.95 (1.28, 4.45)
Cl^-	0.43 ± 0.26 (0.04, 1.38)	6.86 ± 1.95 (3.74, 9.78)	2.48 ± 0.69 (1.54, 3.97)	1.67 ± 1.28 (0.03, 3.22)
NO_3^-	0.65 ± 0.47 (0.10, 1.99)	12.97 ± 5.48 (4.82, 22.84)	3.30 ± 0.44 (2.65, 4.03)	2.37 ± 1.71 (0.55, 4.29)
SO_4^{2-}	1.67 ± 0.98 (0.28, 4.98)	17.77 ± 6.03 (8.64, 28.68)	8.05 ± 2.51 (5.00, 12.78)	4.78 ± 3.57 (0.22, 10.26)
TWSII[b]	6.51 ± 4.86 (2.66, 14.88)	46.83 ± 12.78 (26.55, 70.64)	29.02 ± 6.46 (20.69, 38.43)	19.20 ± 7.90 (7.38, 32.14)

[a] Standard deviation

[b] Total water-soluble inorganic ions

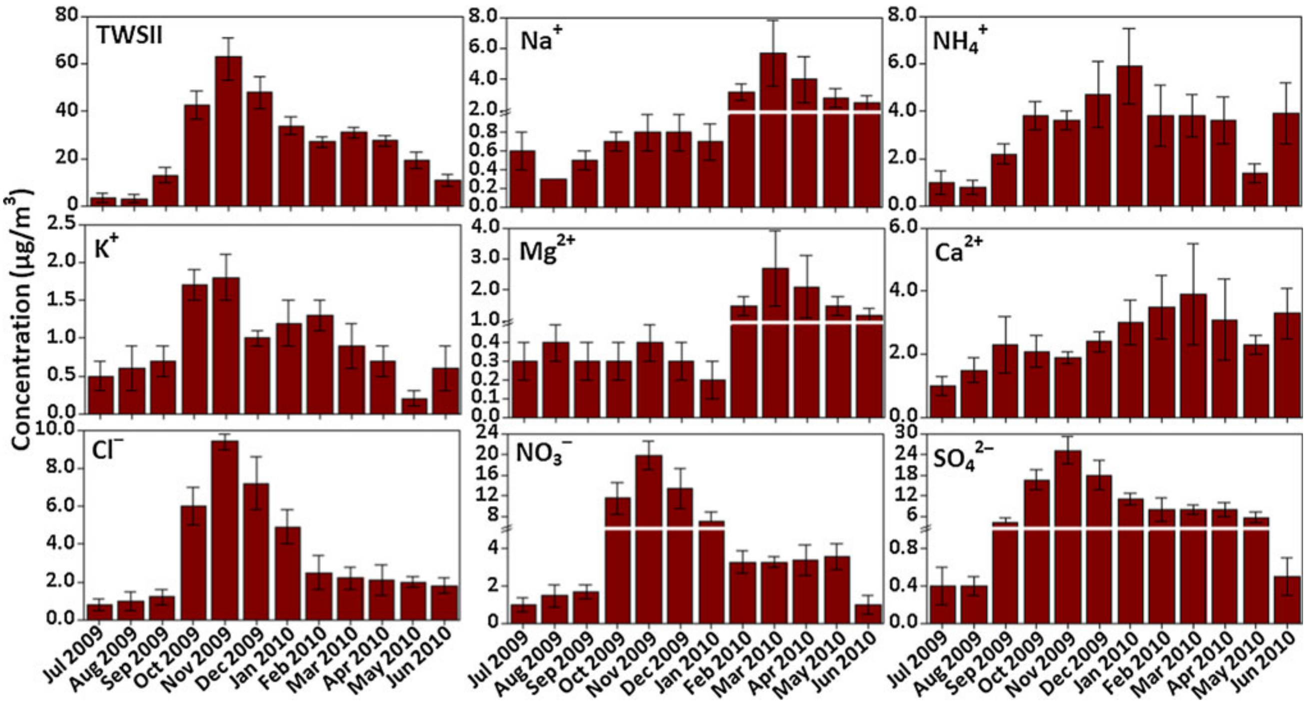

Fig. 4 Monthly variation of PM_{10} and water-soluble ions concentrations from July 2009 to June 2010 in Durg City

concentration of K^+ and Cl^- were higher during winter, and this is primarily related to increased biomass burning during the winter period. Potassium is a tracer of biomass burning (Deshmukh et al. 2012a) and the major fuel used for cooking and heating activities in this region is biomass. Furthermore, in Durg City, Cl^- may originate from neutralization of atmospheric NH_3 by HCl that is mainly derived from coal burning and other burning processes (Deshmukh et al. 2012a). Unlike the other ions, Ca^{2+}, Mg^{2+} and Na^+ were distinctly higher in spring and summer than in winter and spring seasons. The spring peak of Ca^{2+}, Mg^{2+} and Na^+ is most reasonably attributed to the high concentrations of fugitive dust in spring (Chelani et al. 2010). Lower concentrations of water-soluble ions observed during monsoon can be attributed to the washout of aerosol-containing water-soluble ions by precipitation.

In this study, principal component analysis (PCA) plus Varimax rotation of components (PCA + Varimax) was used to identify the sources of analyzed water-soluble ionic species using statistical software (SPSS 16.0). As shown in Table 4, three air quality principal components (PCs) with eigenvalues exceeding 1.0 were found indicating that these PCs have a significant influence on the air quality in Durg City, India. The first PC showed high loading of NH_4^+, NO_3^-, and SO_4^{2-} with a total variance of 58.4 %. These ions are secondary in origin and formed by chemical transformations in the atmosphere of ammonium sulfate and ammonium nitrate (Sun et al. 2004). The major sources of SO_4^{2-} and NO_3^- in the atmosphere are the oxidation of their gaseous precursors SO_2 and NO_x, emitted from various anthropogenic activities (Wang et al. 2006). The second PC showed high loading of Na^+, Mg^{2+} and Ca^{2+} with a total variance of 22.2 %. Theses ions are typically associated with crustal materials in windblown dust, resuspended dust, and dust from paved and unpaved roads (Nicolas et al. 2009). The third PC showed high loading of K^+ and Cl^- with a total variance of 12.2 %. K^+ is a tracer of biomass burning which includes domestic burning of firewood and agricultural residues (Singh and Sharma 2011). This PC represents a combination of coal and biomass burning.

Table 4 Varimax rotated PCA loadings for water-soluble ions of PM_{10} particles in Durg City, India

Variables	PM_{10}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Eigenvalue	% total variance
PC1	0.69	0.13	0.91	0.12	−0.19	0.29	0.15	0.93	0.86	8.41	58.4
PC2	0.35	0.74	−0.18	−0.02	0.89	0.80	0.29	0.08	0.27	4.20	22.2
PC3	0.22	0.24	0.02	0.82	0.19	0.21	0.95	0.12	0.14	2.44	12.2

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