Environ Monit Assess (2010) 168:195–203
DOI 10.1007/s10661-009-1103-6

Seasonal air quality profile of inorganic ionic composition of PM_{10} near Taj Mahal in Agra, India

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Received: 1 August 2008 / Accepted: 21 July 2009 / Published online: 11 August 2009

Abstract Atmospheric aerosols and their impacts on the environment particularly on human health is an issue of significant public and governmental concern. Though studies on air quality related to total suspended particulate matter have done by various authors in India, yet respirable suspended particulate matter (PM_{10}) is not characterized so far particularly in a historical and world heritage city like Agra. This study presents seasonal variation in mass levels of PM_{10} and its ionic composition. PM_{10} samples were collected in the proximity of Taj Mahal and subjected to chemical analysis using ion chromatography technique. The preliminary findings reveal that the 24-h average of PM_{10} mass level varies from 115 to 233, 155 to 321, and 33 to 178 µg/m^3, respectively, in summer, winter, and rainy seasons indicating critical pollution situation. These values are very much higher than the National Ambient Air Quality Standards of 75 µg/m^3 (prescribed by Central Pollution Control Board, India) in both of summer and winter seasons whereas quite near the permissible limits in rainy season. The equivalent ratios of NH_4^+ to nonsea salt SO_4^{2-} and NO_3^- and $\sum$Cations to $\sum$Anios were found to be greater than unity indicating high source strength of ammonia and alkaline nature of aerosols. The study suggests the need for continuous and long-term systematical sampling and detailed physiochemical analysis of PM_{10} and also to know the characteristics of PM in background areas for better understanding of the emission sources.

Keywords Atmospheric aerosols · TSPM · PM_{10} · Respirable suspended particulate matter · Equivalent ratio

Introduction

Atmospheric aerosols have been found to play an important role in the human health problem (Dockery et al. 1993; Maynard and Maynard 2002), visibility degradation (Malm et al. 1994), cloud formation (Ramanathan et al. 2001), and

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scattering and absorption of solar radiation or climate change (Penner and Novakov 1996; Penner et al. 2004). Urban particles come from a multitude of sources, anthropogenic to biogenic; they are very diverse in chemical composition and structure (Salma et al. 2001; Chonga et al. 2002; Johnson et al. 2006; Han et al. 2005). These aerosols contain a major proportion of inorganic ionic constituents and consist of mainly Na^+, NH_4^+, Ca^{2+}, Mg^{2+}, K^+, NO_3^-, SO_4^{2-}, F^-, and Cl^-. Determining their composition is essential to understanding their properties and reactivity and hence their environmental effects. In addition to their emissions, both climatology (temperature, humidity, radiation, wind speed, and precipitation) and geography (topography, soil conditions, and proximity of arid zones or the coast) effect on ambient aerosol characteristics in a given region (Anttila and Salmi 2006).

Though ambient air quality in respect to total suspended particulate matter, SO_2, and NO_2 is being monitored on a regular basis in India since 1967 by governmental agencies like National Environmental Engineering Research Institute and Central Pollution Control Board (CPCB), yet respirable suspended particulate matter (PM_{10}—particles smaller then 10 in aerodynamic diameter) has been added recently by the end of 2000 (World Bank 2001; TERI 2001). In India, ambient air quality data are available for few cities like Varanasi (Pandey et al. 1992), Agra (Kulshresta et al. 1992; Khare et al. 1996), Hyderabad (Sastry et al. 2004), Pune (Gadgil and Jadhav 2004), Paonta Sahib (Gupta et al. 1998), Delhi (Singh et al. 1998), Shilong (Katiyar et al. 2002), and Mumbai (Sharma and Patil 1992); however, data of fine particles are very scare. Sufficient data on PM_{10} are now available worldwide that emphasize on characteristics and sources (Kyotani and Iwatsuki 2002; Shi et al. 2003; Dan et al. 2004; Samburova et al. 2005; Johnson et al. 2006), particle size distribution (Chonga et al. 2002; Han et al. 2005), and spatial and temporal patterns (Gomiscek et al.

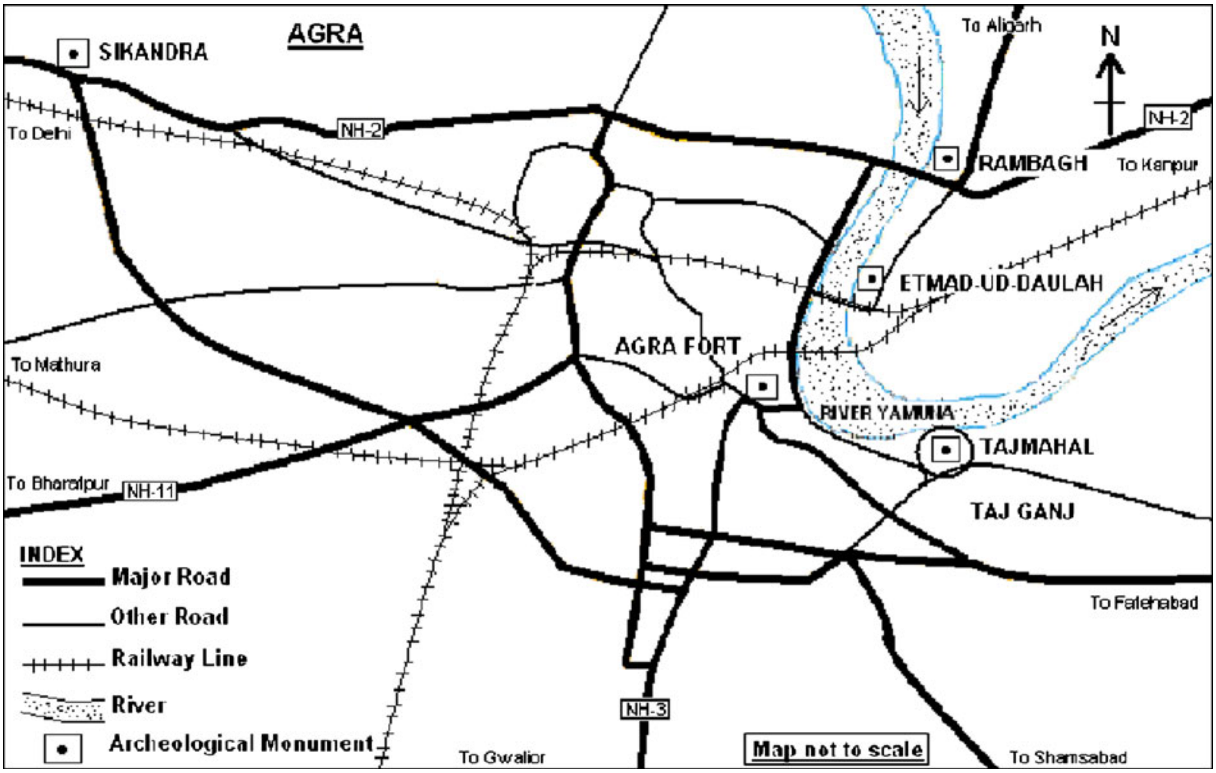

Fig. 1 Location of study site in Agra (shown in *circle*)

2004; Anttila and Salmi 2006). Some studies reported the emissions of fine particles caused by natural episodes (Niemi et al. 2004, 2005; Wang et al. 2006). Nowadays, in developed countries, attention has been paid to fine and ultrafine particles. In the present study, particular attention has been paid to evaluate seasonal variations in mass levels of PM_{10} and inorganic ionic composition and to assess the influences of meteorological conditions. Such data can be useful to asses the trend in pollution emission for control measures and urban planning.

Materials and methods

Study site

The city of Agra (27°12′ N latitude and 78°17′ E longitude), well known as the city of Taj Mahal, one of the wonders of the world, is located in the state of Utter Pradesh in northern India. Two third of its peripheral boundaries are surrounded by Thar Desert of Rajasthan. The climate is tropical, semiarid, and semi-urban region with calcareous sodic, sandy, and dusty soils. The year is divisible into three distinct seasons: summer (March to June), rainy or monsoon (July to October), and winter (November to February) with an annual average of temperature 26°C, relative humidity (RH) 58%, rainfall of about 650 mm (90% received in rainy season), and the prevailing wind direction is NW. Taj Mahal is situated in the eastern part of the city on the bank of river Yamuna (Fig. 1).

Ambient sampling

Sampling was performed from January to December 2007 in the close proximity of Taj Mahal in Taj Ganj area at a residential roof top having a vertical height of about 10 m and a horizontal distance of about 100 m from the Taj Mahal boundary. PM_{10} samples were taken using an Envirotech (New Delhi, India) APM 460 high-volume sampler having a nominal particle size cutoff with diameter of 10 mm, twice in a month at fix-day intervals throughout the year covering all three seasons. Each PM_{10} sample was collected on 20.3 × 25.4 cm Whatman GF/A glass fiber filters having a pore size of 1.6 mm (Whatman International Ltd, England) for 8 h at a nominal flow rate of 1 m^3/min (CPCB 2003). The flow rates through the manometer were monitored once per hour. The sampler flow meter was calibrated once a month. Glass fiber filters were conditioned in desiccators containing silica gel for 24 h before and after sample collection. PM_{10} mass concentrations were determined gravimetrically using precalibrated electronic balance of 10 μg precision. Field blanks were used to ensure accuracy of weights. Meteorological parameters for air temperature, relative humidity, and wind speed were taken using portable weather systems, simultaneously on hourly basis during the sampling hours.

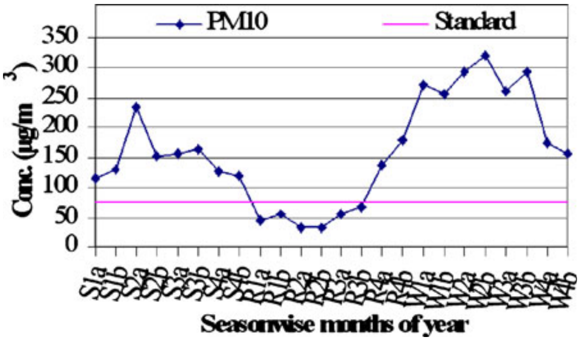

Fig. 2 Seasonal variations in PM_{10} mass levels

Analysis of inorganic ions

For further analysis of inorganic ions viz. F^-, Cl^-, SO_4^{2-}, NO_3^-, Na^+, K^+, NH_4^+, Ca^{2+}, and Mg^{2+},

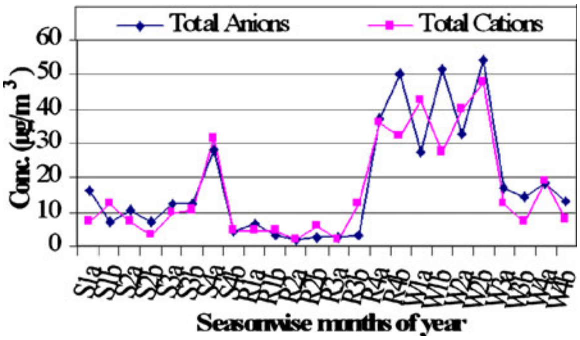

Fig. 3 Seasonal variations in cations and anions mass levels

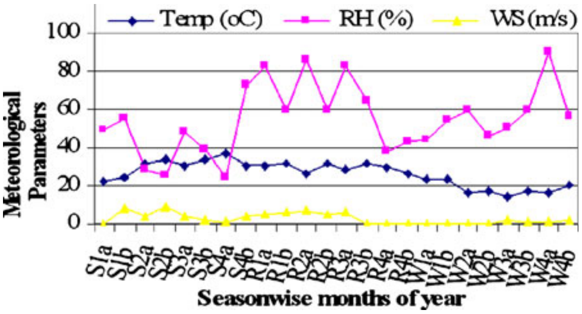

Fig. 4 Seasonal variations in meteorological parameters (*Temp* temperature, *RH* relative humidity, *WS* wind speed, *S* summer, *R* rainy, *W* winter season. *1, 2, 3, and 4* Respective months in a season. *a*, *b* Number of observations in a month)

the filter papers including blanks were extracted in 25 ml deionized water using ultrasonicator for an hour, kept overnight, and filtered through Whatman 42 filter. The target ions were analyzed by ion chromatograph with cation exchange column, Ionpac CS12, and anion exchange column, Ionpac AS4 A-SC with conductivity detector. The eluent used for anion was 1.8 mM Na_2CO_3/1.7 mM $NaHCO_3$ and for cation 18 mM methanesulfonic acid with the regenerant flow rate of 1 ml/min. The concentrations of chemical species were calculated by subtracting the values of blank filters from exposed filters.

Results and discussion

Seasonal variations in PM_{10} and ionic composition

Seasonal variations in mass concentrations of PM_{10}, inorganic cations and anions, and meteorological parameters of respective days as 24-h averages are shown, respectively, in Figs. 2, 3, and 4. As can be seen from Fig. 2, very higher elevated mass levels of PM_{10} were observed during summer and winter seasons (115–233 and 155–321 μg/m^3, respectively) which exceeded the National Ambient Air Quality Standards of 75 μg/m^3, proposed

Table 1 Relative concentrations of chemical components in PM_{10} (micrograms per cubic meter)

Sampling date	PM_{10}	F^-	Cl^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Wind direction
5 January 2008	262	1.90	2.80	7.90	4.31	2.65	2.30	2.90	4.20	0.56	SE
19 January 2008	295	0.41	3.85	0.63	9.60	0.56	0.40	5.29	0.45	0.20	WNW
9 February 2008	176	1.08	4.20	9.01	3.80	2.10	9.10	3.24	4.20	0.50	SW
23 February 2008	155	0.33	2.83	5.60	4.52	2.66	1.10	3.84	0.04	0.23	WNW
9 March 2007	115	6.91	2.40	4.12	2.70	1.34	0.70	2.16	2.30	0.35	ESE
23 March 2007	131	0.17	0.86	1.91	4.00	0.17	9.70	1.42	0.04	0.79	S
6 April 2007	233	1.26	2.10	2.92	4.10	1.40	2.21	1.95	1.70	0.16	WSW
20 April 2007	152	0.32	2.18	1.70	3.14	0.89	1.00	1.26	0.10	0.30	SSW
4 May 2007	158	2.01	3.30	5.04	2.08	2.26	2.20	1.90	3.00	0.68	SSW
18 May 2007	165	0.38	2.94	2.36	6.80	0.48	6.40	2.35	0.06	0.87	WSW
1 June 2007	128	0.81	6.60	14.72	5.82	12.70	4.50	6.12	4.10	4.00	WNW
15 June 2007	121	0.34	1.65	0.09	2.62	0.77	1.60	0.64	0.69	0.56	WSW
6 July 2007	45	1.17	2.60	1.71	0.79	1.25	0.50	1.26	1.10	0.15	SW
20 July 2007	54	0.11	1.06	0.12	1.91	1.80	1.00	0.61	0.92	0.12	ESE
3 August 2007	34	0.54	0.50	0.73	0.29	0.35	0.70	0.36	0.46	0.10	SW
17 August 2007	33	0.09	1.01	0.15	1.46	2.28	0.30	2.08	0.90	0.09	W
7 September 2007	57	0.45	0.60	0.96	0.49	0.43	0.40	0.56	0.60	0.19	W
21 September 2007	68	0.19	1.16	0.31	1.42	3.21	5.80	3.19	0.08	0.04	SW
5 October 2007	138	1.08	11.60	17.98	6.52	10.44	15.40	4.75	4.60	0.60	SSW
19 October 2007	178	2.09	10.52	21.20	16.22	4.88	18.70	1.20	6.27	0.66	NW
2 November 2007	270	1.16	12.40	8.30	5.70	8.06	11.70	14.80	7.20	0.40	NW
16 November 2007	256	1.70	16.30	23.74	9.62	9.24	3.60	5.70	7.80	0.96	SSW
7 December 2007	295	0.36	14.20	9.90	8.20	8.20	9.20	15.20	6.20	1.20	ESE
21 December 2007	321	1.62	20.92	25.23	6.52	11.43	3.40	23.22	8.12	1.74	SE

Table 2 Correlation matrix of PM_{10} aerosols

Spearman's rank correlation	PM_{10}	F^-	Cl^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Temp (°C)	RH (%)	WS (m/s)
PM_{10}	1.00	0.72[a]	0.72[a]	0.72[a]	0.65[a]	0.49[a]	0.83[a]	0.73[a]	0.50[a]	0.35	−0.33	−0.46[b]	−0.32
F^-		1.00	0.87[a]	0.77[a]	0.72[a]	0.61[a]	0.71[a]	0.61[a]	0.63[a]	0.52[a]	−0.09	−0.58[b]	−0.47[b]
Cl^-			1.00	0.67[a]	0.63[a]	0.58[a]	0.84[a]	0.78[a]	0.40	0.48[a]	−0.22	−0.51[b]	−0.28
SO_4^{2-}				1.00	0.88[a]	0.54[a]	0.71[a]	0.60[a]	0.61[a]	0.46[a]	0.06	−0.80[b]	−0.42[b]
NO_3^-					1.00	0.48[a]	0.71[a]	0.61[a]	0.49[a]	0.33	−0.13	−0.70[b]	−0.60[b]
Na^+						1.00	0.55[a]	0.56[a]	0.06	0.00	−0.05	−0.44[b]	−0.17
NH_4^+							1.00	0.88[a]	0.26	0.24	−0.30	−0.50[b]	−0.28
K^+								1.00	0.08	0.22	−0.38	−0.41[b]	−0.31
Ca^{2+}									1.00	0.48[a]	0.13	−0.49[b]	−0.31
Mg^{2+}										1.00	0.14	−0.37	−0.09
Temp (°C)											1.00	−0.31	0.46[a]
RH (%)												1.00	0.18
WS (m/s)													1.00

Significant level 0.05

Temp temperature, *RH* relative humidity, *WS* wind speed

[a] Significant positive correlation

[b] Significant negative correlation

by CPCB, India, whereas PM_{10} levels were observed near permissible limits in rainy season (33–178 µg/m^3) which was due to wash down of pollution by monsoon precipitations. Overall, the PM_{10} mass concentration ranged from 33–321 µg/m^3 with an annual average of 160 µg/m^3. The relative composition of inorganic ions in PM_{10} as 24-h average concentrations is depicted in Table 1.

Correlation between data sets

The aim of the statistical analysis was to gain some of preliminary understanding how closely the mass levels of PM_{10} and its ions are related to the influences of meteorology and their emission sources. For this purpose, the tests were used and correlation results are depicted in Table 2.

As expected from other studies (Simpson 1992; Pandey et al. 1992; Katiyar et al. 2002), a significant negative correlation between wind speed and mass level data of PM_{10} and ions shows the inverse relationship between the data and therefore indicates the predominance of local sources. In this case, strong winds flush out the pollution load whereas low winds allow the pollution levels to rise while weak or negligible correlations indicative the more complex picture with advections bringing pollution into the region from other sources. A strong negative correlation of RH with PM_{10} and ionic data sets also shows inverse relationship (Katiyar et al. 2002); thus, in case of higher RH, the pollutants get settled down or washed out by precipitations particularly in rainy season. Air temperature also shows negative correlations with the data sets. A significant positive correlation of ions with PM_{10} shows their higher associations and sources of origin are same in nature.

The average concentrations of anions and cations and the percentage ionic composition in PM_{10} are given in Figs. 5 and 6, respectively. The ionic composition accounted for about 17% in PM_{10} in ambient air. The ratio of $\sum$Cations and $\sum$Anios milliequivalents per cubic meter was

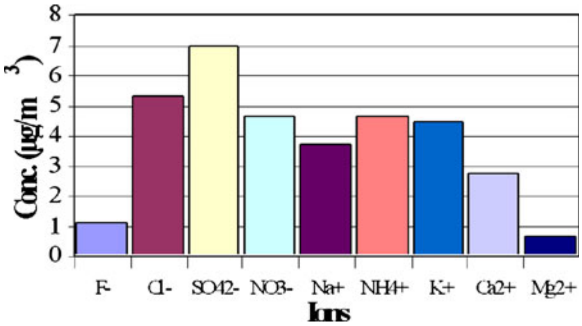

Fig. 5 Average ionic concentrations (micrograms per cubic meter)

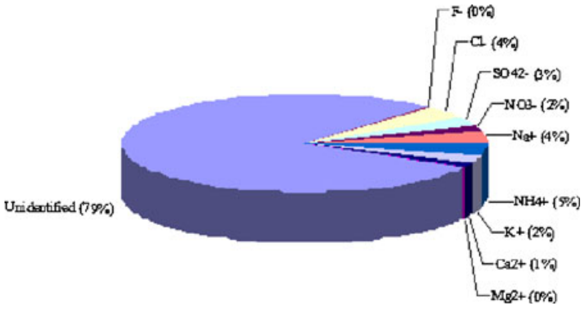

Fig. 6 Percentage of inorganic ionic constituents of PM_{10}

1.68, and the correlation between $\sum$Cations and $\sum$Anios milliequivalents per cubic meter showed a very good correlation ($r^2 = 0.63$). The minimum, maximum, and standard deviation values are presented in Table 3.

Among the anionic components, sulfate followed by chloride and nitrate was the major and fluoride was the minor component whereas in cationic components, ammonium followed by potassium and sodium was major and calcium and magnesium were minor components. The possible sources of sulfate include fuel combustion, vehicles, and industries. Sea spray may also be source for sulfate. The ionic constituents such as SO_4^{2-}, K^+, Ca^{2+}, and Mg^{2+} are derived from either marine or nonmarine origins, such as anthropogenic and soil emissions. Hence, it is necessary to discriminate sea salt (ss), SO_4^{2-}, K^+, Na^+, and Ca^{2+} from nonsea salt (nss) contribution.

The nonmarine component was evaluated from the Na^+ concentration on an assumption that all the sodium ions originated from sea. The marine ss and nonmarine nss contribution and their percentage to the total ionic content in PM_{10} were calculated using ratio like SO_4^{2-}/Na^+, K^+/Na^+, and Ca^{2+}/Na^+ in bulk sea water using the formulae (Kumar et al. 2006).

$$\text{nss}X = \text{Total } X_{\text{Measured}} - \text{ss}X$$

$$\text{where, ss}X = [\text{Na}]_{\text{Measured}} \times [X/\text{Na}]_{\text{Sea}}.$$

X is an ionic component in aerosol and X/Na is the standard sea water ratio. The nss contribution of SO_4^{2-}, K^+, Ca^{2+}, and Mg^{2+} to PM_{10} was over 88.70% suggesting an anthropogenic origin or natural origin like soil as shown in Table 4.

Potassium ions are commonly found in plants as a constituent of chlorophyll. nss-K^+ contributed 96.43% of the total K^+ indicating biomass burning as a source. Equivalent ratio of Cl^-/ Na^+ was found to be in 0.938 which is less than the ratio found in sea water indicating marine sources.

Neutralization ratio

Ammonia emitted in atmosphere by natural decay and decomposition, and it is rapidly converted to NH_4^+ aerosols at a rate of 30% h^{-1}. These ammonium ions react with acidic gases like sulfuric and nitric acid in the air forming ammonium salts. The neutralization ratio (NR), the ratio of the concentration of NH_4^+ (in nanoequivalents per cubic meter) to the sum of the concentrations of nss-SO_4^{2-} and NO_3^- (in nanoequivalents per cubic meter), was calculated to assess the acidity of atmospheric aerosols (Balasubramanian et al. 2003). A neutralization ratio of unity suggests the presence of nss-sulfate and nitrate as their ammonium salts. The mass of NH_4^+ ions as aerosols is taken to be the amount needed to neutralize the acidity, while NR values lower than unity indicate presence of acidic aerosols. Neutralization ratios greater than unity probably arise from the presence of NH_4Cl. In the present study, the average

Table 3 Statistics of ions in PM_{10} (micrograms per cubic meter)

	F^-	Cl^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}
Mean	1.10	5.36	6.93	4.69	3.73	4.66	4.42	2.71	0.64
Minimum	0.09	0.50	0.09	0.29	0.17	0.30	0.36	0.04	0.04
Maximum	6.91	20.92	25.23	16.22	12.70	18.70	23.22	8.12	4.00
Stdev	1.39	5.73	7.93	3.66	3.95	5.13	5.58	2.75	0.82
Count	24	24	24	24	24	24	24	24	24

Table 4 Contributions of nss and ss ions to PM_{10}

Ion	nss		ss	
	Concentration (μg/m^3)	% in PM_{10}	Concentration (μg/m^3)	% in PM_{10}
SO_4^{2-}	0.125	86.64	0.019	13.36
K^+	0.109	96.43	0.004	3.57
Ca^{2+}	0.128	94.65	0.007	5.35
Mg^{2+}	0.035	66.46	0.018	33.54

equivalent ratio of NH_4^+ to the sum of nss-SO_4^{2-} and NO_3^- (in nanoequivalents per cubic meter) was 1.18 that suggests high source strength of NH_3 to neutralize not only sulfuric acid completely but also HNO_3 almost completely. The higher ammonium concentrations were attributed due to close vicinity of the river Yamuna and agricultural activities in this river belt and the presence of wastewater treatment plant in a circumference of about 3 km from the site.

A high loading for crustal elements (nssMg and nss-Ca) is attributed to a number of reclamation and construction activities of infrastructural developments in the city, whereas the presence of nitrate, nss-sulfate, nss-K, and ammonium is indicative of combustion sources. Nss-K is a well-known tracer of biomass burning. Na and Cl are possibly indication of the presence of sea salts; however, Na may also be contributed due to sodic soils in the region. The contribution of the above pollution sources to the observed mass concentration of aerosols in Agra appears to be reasonable, considering the geographical location and the type of human-induced activities taking place in and around the city.

Conclusions

PM_{10} measurements were carried out for 1 year from January to December 2007 in a semi-urban city of Agra and results showed that 24-h average values of PM_{10} vary from 33 to 321 μg/m^3. Very higher elevated levels were observed in summer and winter seasons indicating critical pollution situation, whereas low levels in rainy season indicating the influence of monsoon precipitations washing down the pollutants. Except the emission sources, meteorological and geographical conditions play an important role in air quality evaluation. Characterization for anions and cations revealed that the water soluble inorganic ionic composition of PM_{10} accounts for about 17%. The equivalent ratios of NH_4^+ to nss-SO_4^{2-} and NO_3^- (1.18) and $\sum$Cations to $\sum$Anions (1.68) were greater than unity, indicating higher source strength of NH_3 and alkaline nature of atmospheric aerosols. The nss contribution of SO_4^{2-}, Ca^{2+}, K^+, and Mg^{2+} was over 89.17%, suggesting an anthropogenic and/or natural origin like soils as dominant sources than sea salts, whereas Nss-K is a well-known tracer of biomass burning that contributes 96.43% to K^+ emissions. The study suggests the need for continuous and long-term systematical sampling and detailed physiochemical analysis of PM_{10} and also to know characteristics of PM in background areas for better understanding the emission sources.

Acknowledgements The authors gratefully acknowledge the contributions of Dr. Rajeev Upadhaya, UPPCB, Mathura and Dr. Amit Bansiwal, NEERI, Nagpur for providing instrumental support with IC measurements. We would like to thank Mr. Fasiur Rehman for sharing his valuable knowledge in this study.

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