Indian Journal of Radio & Space Physics
Vol 43, August - October 2014, pp 284-292

Chemical characterization of particulate matter at Sinhagad, a high altitude station in Pune, India

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Received 27 January 2014; revised 25 August 2014; accepted 5 September 2014

Particulate matter, PM_{10} and $PM_{2.5}$, samples were collected at Sinhagad, a high altitude location, near Pune, India during November 2008 - April 2009. The average concentrations of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ at Sinhagad were 35.8, 14.1 and 21.7 µg m^{-3}, respectively. The average ratio of $PM_{2.5}/PM_{10}$ (0.39) suggested that PM_{10} at study area is dominated by primary particulate emissions by natural activities. In PM_{10}, anions and cations contributed 33% and 67%, whereas in $PM_{2.5}$, these were 43% and 57%. In both PM_{10} and $PM_{2.5}$, contribution of marine components, viz. Na^+, Cl^- and Mg^{2+} were found to be 58% and 49%, respectively reflecting Arabian Sea is the major source of these components. All the ionic components in PM_{10} showed higher concentration in summer, whereas in $PM_{2.5}$, secondary particles (SO_4^{2-}, NO_3^- and NH_4^+) showed higher concentration in winter. The order of neutralization factor was found to be $Ca^{2+} > Mg^{2+} > NH_4^+ > K^+$ for PM_{10}; and $Ca^{2+} > NH_4^+ > Mg^{2+} > K^+$ for $PM_{2.5}$. Source categorization, i.e. marine, crustal and anthropogenic of chemical components of PM has been done. Sea salt and crustal fractions have been calculated by assuming entire Na and Ca from Sea and crust, respectively. The fraction of the chemical component, which remains after deducting the sea and crust fractions, has been considered as of anthropogenic origin. Major contribution observed was from marine sources [$PM_{2.5}$ (43%) and PM_{10} (53%)]; followed by crustal sources [$PM_{2.5}$ (25%) and PM_{10} (30%)]; and then anthropogenic sources [($PM_{2.5}$ (32%) and PM_{10} (17%)].

Keywords: Particulate matter, Chemical composition, Air pollution, Aerosol

PACS Nos: 92.60.Sz; 82.33.Tb

1 Introduction

Particulate air pollution is a complex mixture of small and large particles of varying origin and chemical compositions. Particulate matter (PM) have strong influence in many atmospheric processes, with important environmental effects including changes in visibility, solar radiation transfer (related with global warming), cloud formation, and play a major role in the acidification of clouds, rain and fog[1,2]. The fine airborne particles have high probability of deposition deeper into the respiratory tract and are likely to trigger or exacerbate respiratory diseases. These particles also have higher burdens of toxins, which when absorbed in the body can result in health consequences other than respiratory health effects. Particles, having 2.5-100 µm diameters, usually comprise more of dust from agriculture, construction, road traffic, plant pollens and other natural sources. Smaller particles, with less than 2.5µm diameter, generally come from combustion of fossil fuels. These particles include soot from vehicle exhaust and are often coated with various chemical contaminants or metals. Fine sulphate and nitrate particulates are formed when SO_2 and NO_x condense in the atmosphere. The major neutralizing species in the atmosphere are NH_3 and $CaCO_3$, and their relative dominance over a particular region can be ascertained by comparing their abundance in precipitation[3,4]. The large sources of fine particle are fuel combustion in industries and diesel exhaust from vehicular traffic. Important information on the origin of particulate matter can be obtained through chemical analysis[5-17]. The aerosol composition is highly variable in space and time and depends upon the relative contribution from diverse sources such as sea salts, mineral dust and anthropogenic emissions as well as on the meteorological conditions[18]. Many components can be used as tracers for specific sources, for example sodium is a tracer that is almost exclusively associated with sea salt. The degree of the acidity of deposition depends on neutralization by certain alkaline components, such as NH_4^+, Ca^{2+} [in the form of $CaCO_3$ or $Ca(OH)_2$], Na and Mg. The objective of the present study is to investigate: (i) variation in concentrations of major acidifying (SO_4^{2-} and NO_3^-) and alkaline (Ca^{2+} and NH_4^+) constituents of particulate matter (PM_{10} and $PM_{2.5}$); (ii) their effect on the acidification/neutralization potential of particulate; and (iii) quantification of sources of PM_{10} and $PM_{2.5}$. The data revealed from the present study can be useful for the other studies as this site is free from the local pollution sources.

2 Materials and methods

2.1 Sampling site

Sinhagad (18°21′N and 73°45′E, 1450 amsl) is a hill station on a mountaintop in the Western Ghats mountain range. It is located at about 18 km southwest of Pune (18°32′N, 73°51′E). Its top is flat with an area of about 0.5 km^2. Other mountain peaks of comparable heights surround it. This part of the Western Ghats is covered with vegetation, grass and trees. The only noticeable local source of pollution is wood burning for cooking. A few people live at the summit and some tourists visit the area by foot. The vehicular traffic stops at about 1 km distance from the sampling location and about 100 m below the sampling height. Thus, the site is relatively free from major urban pollution sources. Sampling was carried out in the complex of a Microwave Tower building (about 10 m) owned by Bharat Sanchar Nigam Limited (BSNL), Government of India, at Sinhagad fort. It is a protected site and trespassers or tourists are not allowed to enter. Figure 1 depicts the exact location of Sinhagad site.

2.2 Sample collection

PM_{10} and $PM_{2.5}$ samples were collected from November 2008 to April 2009 at Sinhagad using an air sampler (APM-550 from Envirotech Pvt Ltd, India,). PM_{10} and $PM_{2.5}$ sampler is based on designs standardized by US EPA, Omni-directional air inlet with PM_{10} separation through an impactor followed by $PM_{2.5}$ separation WINS impactor. For sampling of PM_{10} and $PM_{2.5}$, glass (Whatman- GF/A) and PTFE filters were used, respectively. The desiccated filter papers were weighed twice (before and after the sampling) on the micro-balance (AE 163, Metler) and

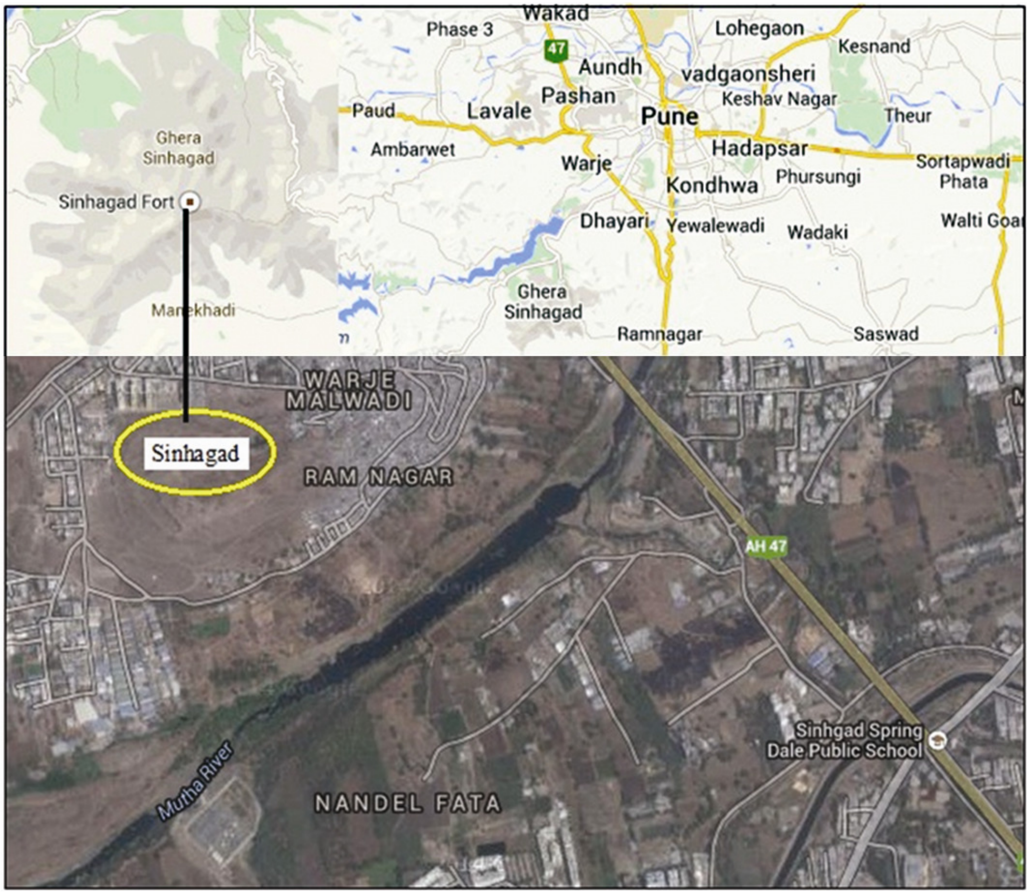

Fig. 1 — Map showing the sampling site

again desiccated for 24 hours. The conditioned and weighed filter papers (PM_{10} and $PM_{2.5}$) were placed in filter holders and taken to the field for sampling to avoid contamination of the filter papers on the way. Before starting the sampling, initial volume and timer readings were noted for PM_{10} and $PM_{2.5}$ in field monitoring sheet. The pre-weighted and coded filter papers were placed in the filter holder of the respective samplers and screwed properly before starting the samplers. Both the PM_{10} and $PM_{2.5}$ samplers were operated for 24 hours sampling period. Before and after each set of sampling, data were entered in the field data sheet in the pre-defined format and concentrations of PM_{10} and $PM_{2.5}$ were calculated gravimetrically. After sampling, filter papers (PM_{10} and $PM_{2.5}$) were removed with forceps and placed in the cassette and the cassette was wrapped with aluminum foil to prevent the degradation of organic compounds due to photo-oxidation and brought back to the laboratory. The weighed filter papers were preserved in freezer for further chemical analysis.

2.3 Chemical analysis

After exposing the filter papers for the required time, papers were extracted for water soluble components by soxhlet extractor. After extraction, the water soluble extracts of PM were analyzed for major anions and cations (Cl^-, SO_4^{2-}, NO_3^-, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+}). Ion chromatograph (DX100) was used for the analysis of Cl^-, SO_4^{2-} and NO_3^-. Also, NH_4^+ was analysed by UV-Vis spectrophotometer (Spectronic -20D). Ammonium (NH_4^+) reacts with phenol and hypochlorite in the presence of a catalyst (sodium nitroprusside) to produce a blue coloured indo-phenol dye (Weatherburn, 1967). After the colour development, the absorption was measured at 625 nm against the blank reagent. The analysis of cations, i.e. Na^+, K^+, Ca^{2+} and Mg^{2+} were done by Atomic Absorption Spectrometer (AAS) (Perkin Elmer - Analyst 100).

2.4 Quality control

2.4.1 Quality control in monitoring

(i) The sampler is designed to work at a constant flow rate of 16.67±0.83 L min^{-1}. Daily flow rate calculations (gas meter reading/timer reading) were being made to make sure that the fluctuations in flow rate are within the range; (ii) Filter in the wins impactor needs to be changed after 72 h of sampling[19] or when the filter gets clogged as per the operator′s judgment. The filter should be immersed in 3- 4 drops of silicon oil at regular intervals as per the need; (iii) Periodic cleaning of the sampler was done to make the sampler dust free so that the dust on the sampler may not be counted with the mass concentration of the sample.

2.4.2 Blank correction

Background contamination was monitored by using field blanks (unexposed filter papers), which were processed with samples. The field blank filters, exposed in the field for few seconds, were collected thrice during the season. Background contamination was eliminated by subtracting the field blank values from concentrations of the samples. Usually, field blank values were very low, typically below or around the detection limits.

2.4.3 Reproducibility test

Reproducibility test demonstrates the stability of the analytical instruments. Analysis of the same standard solution was repeated 10 times on IC, AAS and UV-Vis. spectrophotometer. The relative standard deviation was ranged 0.62-5.8% for all analyzed species which indicates about 95% reproducibility of measured species.

3 Results and Discussion

The average chemical composition of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ at Sinhagad are given in Table 1. The average concentrations of PM_{10} and $PM_{2.5}$ at Sinhagad were found to be 35.8 and 14.1µ gm^{-3}, respectively. The higher PM_{10} values indicate the impact of marine source, especially sea salt from Arabian Sea. The day-to-day change in order of magnitude of PM_{10} and $PM_{2.5}$ concentrations is found to be 7.4 – 58.2 µg m^{-3} for PM_{10} and 7.2–39.9 µg m^{-3} for $PM_{2.5}$. The

Table 1 — Average chemical composition of PM_{10} and $PM_{2.5}$ (neq m^{-3}) at Sinhagad

Species	PM_{10}	$PM_{2.5}$	($PM_{10-2.5}$)
*Mass (µg m^{-3})	35.8 (15.2)	14.1 (8.2)	21.7 (12.8)
Na^+	83.3 (20.8)	34.1 (14.2)	49.3 (23.4)
K^+	17.3 (10.6)	7.96 (2.2)	9.31 (4.2)
Ca^{2+}	72.9 (30.4)	27.2 (10.1)	45.7 (23.2)
Mg^{2+}	36.8 (12.8)	16.2 (4.6)	20.6 (10.7)
NH_4^+	20.2 (10.4)	18.8 (8.8)	1.33 (0.7)
Cl^-	98.1 (40.8)	38.5 (20.1)	59.5 (20.1)
NO_3^-	13.4 (8.8)	10.1 (5.4)	3.4 (1.2)
SO_4^{2-}	32.1 (12.8)	29.0 (15.3)	3.1 (0.5)
Nss SO_4^{2-}	22.7 (12.4)	20.1 (5.8)	2.6 (0.3)

Standard deviation are given in parenthesis difference between the concentrations in PM_{10} and $PM_{2.5}$ (PM_{10}–$PM_{2.5}$) is the actual contribution of the coarse fraction and is found to be 21.7 µg m^{-3}. The mass concentrations of PM_{10}–$PM_{2.5}$ varies, which correspond to the variations in $PM_{2.5}$ and PM_{10}. The average ratio of $PM_{2.5}/PM_{10}$ (0.39) indicates that PM_{10} at study area is dominated by primary particle emissions by natural and man-made activities. Pipal *et al.*[16] have reported very high $PM_{2.5}/PM_{10}$ ratio (0.70) in a rural site in Agra and low at road site location (0.32). Tiwari *et al.*[20] have reported the ratio of $PM_{2.5}/PM_{10}$ over Delhi to be ~0.48, which varied between 0.18 (in June) and 0.86 (in February), suggesting the dominance of coarse mode particles in summer and fine mode particles in winter. Kocak *et al.*[21] have reported the great variability in the ratios of $PM_{2.5}/PM_{10}$ (ranging 0.25 - 0.90) in Turkey. Wang *et al.*[22] reported very high ratio (0.70) between $PM_{2.5}$ and PM_{10} in Shanghai, China due to industrial effect. The observed ratios of the present study are comparable with these reported studies.

3.1 Percentage contribution of PM

The percentage contribution of each chemical species to the total measured ionic components in PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ is shown in Fig. 2. The measured total water soluble ionic fraction contributed approximately 30% to the total $PM_{2.5}$ in which anions and cations account for about 43 and 57%, respectively. On the other hand, approximately 40% of the total PM_{10} was found to contribute as total water-soluble ionic fraction of which anions and cations accounted for about 33 and 67%, respectively. The un-analysed portion of both PM_{10} and $PM_{2.5}$ samples is deemed to consist of carbonaceous aerosols, such as black carbon, organic carbons and other insoluble elements. Contribution of marine components, i.e. Na^+, Cl^- and Mg^{2+} were dominant in both sizes 58% in PM_{10} and 49% in $PM_{2.5}$ at Sinhagad since the air flow during March and April is predominantly westerly and large amount of moisture is brought inland from the Arabian Sea[18] (Fig. 3). This is responsible for the observed high concentration of sea salt ions Cl^-, Na^+ and Mg^{2+}.

Significant contributions of SO_4^{2-}, NO_3^- and NH_4^+ were found in $PM_{2.5}$ after sea salt components, which could perhaps be linked to the anthropogenic emissions primarily from burning of fossil fuel and biomass[23-25] and human activities. On the other hand, considerable amounts of Cl^-, Na^+ along Ca^{2+}, Mg^{2+} and K^+ found in PM_{10} are linked to the natural sources, which are essentially soil derived[25,26].

3.2 Seasonal variation of PM

Figure 4 shows the seasonal variation of chemical composition of PM_{10} and $PM_{2.5}$ at Sinhagad during

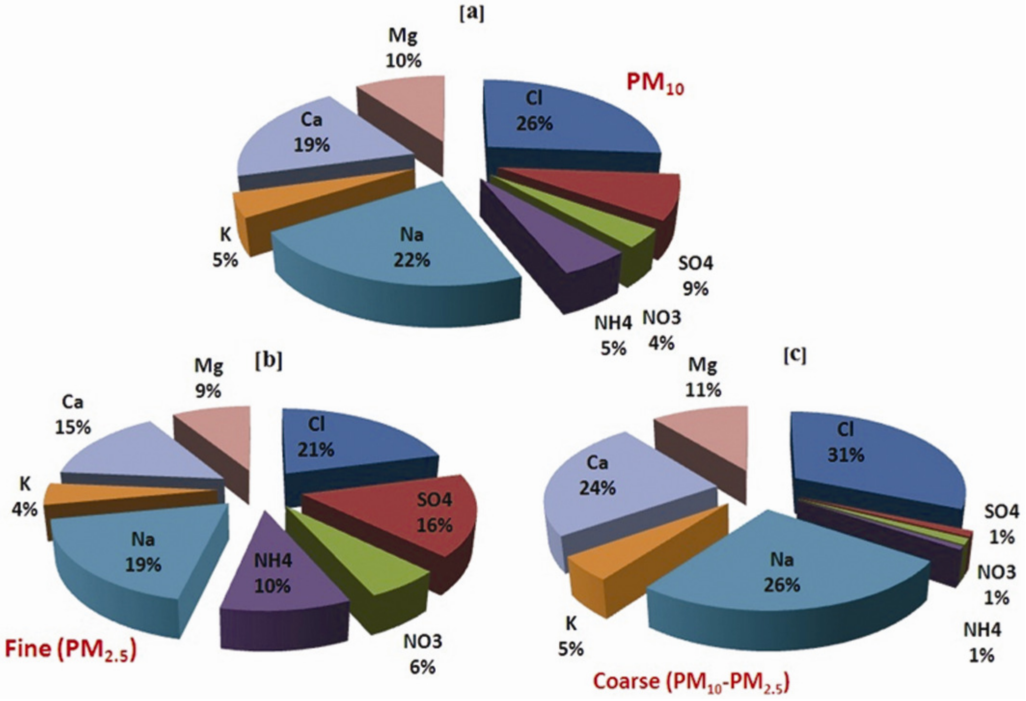

Fig. 2 — Percentage contribution of measured ionic species in $PM_{10,}$ fine and coarse

winter (November to February) and summer (March to April). In PM_{10}, all the ions in summer season showed high concentration because the atmosphere at Sinhagad, generally, remains unstable due to convection. In addition, during summer, particles of local and land origin are continuously lifted up which result in higher concentration of soil-derived elements (Ca^{2+}, Mg^{2+} and K^+). High concentrations of sea salt components (Cl^- and Na^+) may be due to the SW winds coming over the Arabian Sea (Fig. 3).

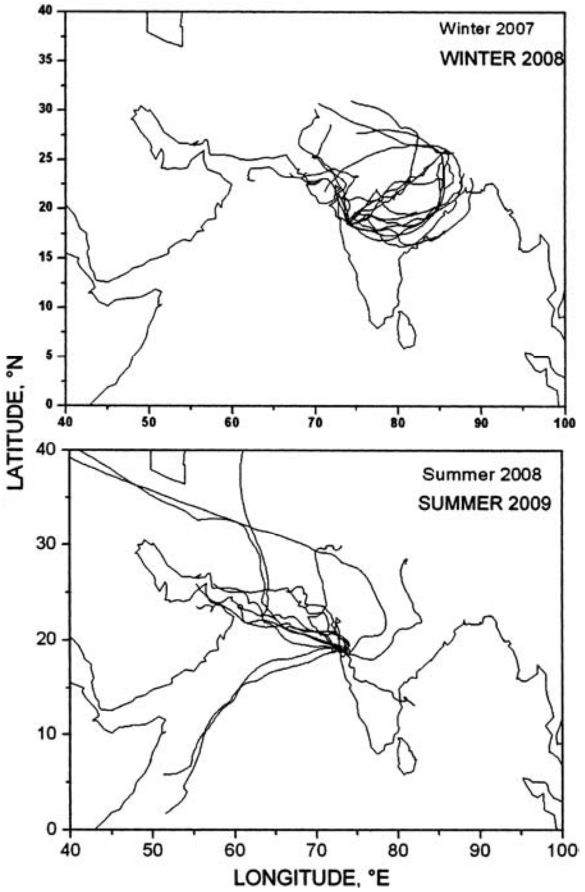

Fig. 3 — Air mass backward trajectory at 1500 m (amsl) for 7 days ending at Sinhagad

The low ionic concentrations during winter can be explained on the basis of the characteristics of lands use and wind pattern. During winter, lands are mostly covered by vegetations and wind comes from continental area, therefore, effect of lifting up of soil oriented components and sea salt are minimal. In $PM_{2.5}$, the concentration of anthropogenic components, like SO_4^{2-}, NO_3^- and NH_4^+, are high in winter as these species are formed through gas to particle conversion process by their respective precursor gases [SO_2, and NO_2 get oxidized when reacted with water vapour in the atmosphere and NH_4^+ is formed from reduction of its precursor ammonia gas (NH_3)]. These precursor gases (SO_2, NO_2 and NH_3) are emitted from various industrial, vehicular and domestic activities[27]. However, the acidic aerosols like SO_4^{2-} and NO_3^- form salts, which when reacted with certain cations like Na^+, Ca^{2+}, K^+ or Mg^{2+}. SO_4^{2-} and NH_4^+ aerosols form compounds like $(NH_4)_2SO_4$ or NH_4HSO_4 which mostly occur in fine fraction of aerosols[28]. The coarse fraction of SO_4^{2-} is in the form of gypsum ($CaSO_4$) or as a reaction product with sea salt aerosols, like Na^+ i.e. $(Na)_2SO_4$ (Refs 29,30). Also, fine fraction of NO_3^- is generally in the form of NH_4NO_3 but its coarse fraction is present in the form of $Ca(NO_3)_2$ or $NaNO_3$ through the reaction of HNO_3 vapours with soil or sea originated elements, respectively[29,30]. Khemani[31] has

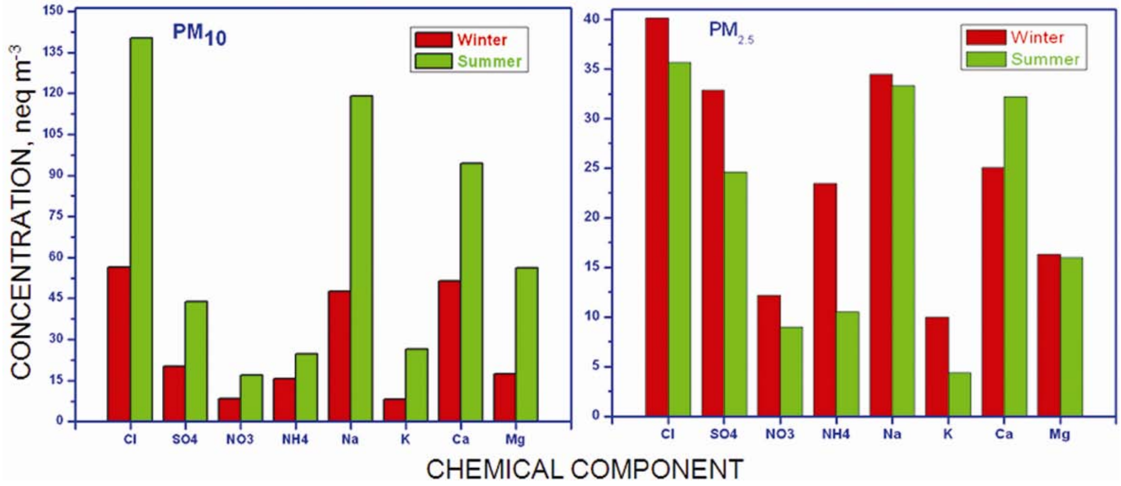

Fig. 4 — Seasonal variation in chemical composition of PM_{10} and $PM_{2.5}$ at Sinhagad

Table.2 — Comparison of ionic concentrations (μg m^{-3}) of atmospheric aerosol particles with other high altitude study

Location	Altitude, m (asl)	Size	Na^+	NO_3^-	Nss SO_4^{2-}	NH_4^+	nssK^+	nss Ca^{2+}	Cl^-	References
Sinhagad, Pune, India	1450	$PM_{2.5}$	0.78	0.62	0.96	0.33	0.31	0.54	1.34	Present study
		PM_{10}	1.91	0.83	1.08	0.36	0.67	1.45	3.43	
Mt Norikura, Japan	2770	Fine fractions (<2.1 μ dia)	0.03	0.66	2.21	0.83	1.25	0.22	-	Osada *et al.*[34]
Mt Norikura, Japan	2770	Fine fractions (<2.1 μ dia)	0.02	0.08	1.25	0.16	-	0.01	0.007	Osada *et al.*[35]
Mt Tateyama, Japan	2450	Fine fractions (<2.1 μ dia)	0.03	0.21	0.56	0.09	0.01	0.03	-	Kido *et al.*[36]
Tibetan Plateau, China	3300	TSP*	0.33	0.21	1.07		0.38	0.35	0.2	Zhao *et al.*[37]
Darjeeling Northeastern Himalayas, India	2194	$PM_{2.5}$	0.66	3.31	3.8	0.88	1.15	0.13	1.21	Chatterjee *et al.*[38]
Nagarkot, Nepal Himalayas	2150	$PM_{2.5}$	0.2	1.2	3.8	1.5	0.62	0.31	0.05	Rengarajan *et al.*[39]
Manora Peak, India	1950	TSP*		0.5			0.52			Goldberg *et al.*[40]
Mt Abu, Western India	1680	TSP*	0.28	0.74		0.37	-	1.7	0.31	Rastogi *et al.*[18]

*TSP = Total suspended particulates

reported presence of $(NH_4)_2SO_4$ as well as $NaNO_3$ at Pune. Coarse form of NO_3^- at Pune is also reported by Momin *et al*[32] and Safai *et al*[33].

Table 2 presents the comparison of different water soluble ionic species with other high altitude stations. It is observed that the concentrations of Na^+, Ca^{2+} and Cl^- are several times higher than all the other hill stations[18,34-40], which indicate the strong influence of sea salt aerosol over Sinhagad. The concentrations of NH_4^+, NO_3^- and K^+ species in PM over Sinhagad were similar with the data reported form other hill stations. The concentration of SO_4^{2-} was less than all other hill stations due to different proximity of sampling station. The present site is nearby the coastal area and high influx of tourists in Sinhagad could be the reason of the higher loading of chemical species.

3.3 Neutralization factors

The acid neutralizing capacity of different cations was estimated by calculating the neutralization factors (NF) with respective to the particular cations. These factors are calculated by considering NO_3^- and SO_4^{2-} as the major acidifying anions; and Ca^{2+}, NH_4^+, Mg^{2+} and K^+ as major acid neutralizing cations in the PM (Ref. 18). The role of Cl^- in acidity as well as that of Na^+ in alkalinity is negligible, since they originate mostly from sea in the form of sea salt. Some of the cations like Ca^{2+} and Mg^{2+} and anions like SO_4^{2-} measured over land and partially originate from sea in the form of neutral salt and partially from the anthropogenic activity. The fractions of these ionic components which originate from sea do not play any role in deciding acidity/alkalinity of the PM. Thus, while defining NF value of a cation, only the non-sea salt (nss) fractions of these components like NssCa, NssMg, NssK and $NssSO_4$ are considered (Table 3).

Table 3 — Non-sea salt (Nss) ratios of different ions

	Nss SO_4^{2-}	Nss K^+	Nss Mg^{2+}	Nss Ca^{2+}
PM_{10}	22.7	15.5	17.9	69.2
$PM_{2.5}$	20.1	7.2	8.5	26.1

Neutralization factors for different cations in PM_{10} and $PM_{2.5}$ collected at Sinhagad are shown in Fig. 5. In both the sizes, Ca^{2+} served as the major neutralizing component for aerosol acidity. The suspended particulate matter, rich in carbonates/bicarbonates of Ca^{2+}, buffers the acidity of aerosol, which is commonly observed under Indian conditions. The order of neutralization was $Ca^{2+} > Mg^{2+} > NH_4^{2+} > K^+$ for PM_{10}; and $Ca^{2+} > NH_4^{2+} > Mg^{2+} > K^+$ for $PM_{2.5}$ at Sinhagad.

3.4 Source categorization of chemical components of PM

Using the computed crustal and sea-salt fractions, PM have been classified into three main categories: (i) crustal or soil originated particulate; (ii) marine or sea salt aerosols; and (iii) anthropogenic or man-made aerosols. Considering that Na is wholly from sea and Ca from crust, non-sea salt (Nss) and non-crustal (Ncr) fractions have been calculated for the measured

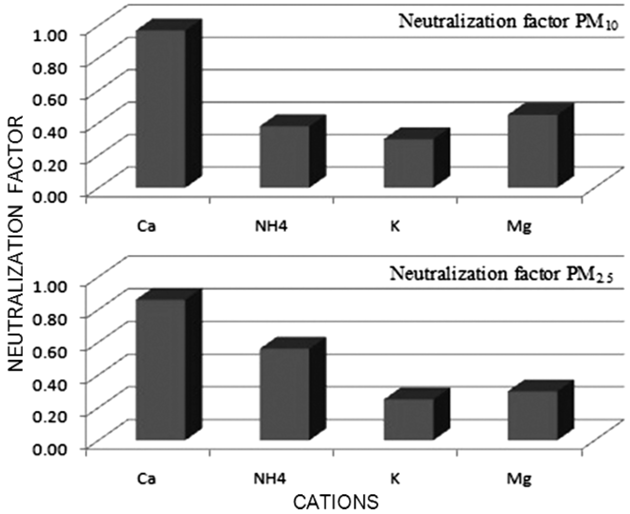

Fig. 5 — Neutralization factors for different cations

chemical components of aerosols. The fraction of the chemical component that remains after deducting sea-salt and crustal fractions is considered from the anthropogenic sources.

3.4.1 Calculation of the non-sea salt fractions

For the assessment of marine contribution towards the formation of different chemical components of aerosols, a widely used method was employed, in which the non-sea salt (Nss) fraction of particular ionic component was calculated, by assuming Na as a reference element for sea source. For example, Nss SO_4^{2-} is computed as:

$$\text{Nss } SO_4^{2-} = [SO_4^{2-}] - \{([SO_4^{2-}]/[Na])_{sea} * [Na]\}$$

where, $([SO_4^{2-}]/[Na])_{sea}$ is the standard ratio of concentration of SO_4^{2-} and Na, which is obtained from sea water composition[41]; and $[SO_4^{2-}]$ and $[Na^+]$ are the concentrations (μeq L^{-1}) in aerosols. Similarly, Nss fractions have been computed for the other chemical components. Annual mean concentrations of non-sea salt ions in PM_{10} and $PM_{2.5}$ are given in Table 3.

Sea salt (ss) fractions for these components computed as:

$$ss = \{([X]/[Na])_{sea} * [Na]\}$$

where, X is the desired component of aerosol for which 'ss' fraction is to be calculated.

Computation of Nss is not only useful for deciding the marine source contribution but is also essential for the examination of acidic or alkaline nature and neutralization potential of certain components of PM.

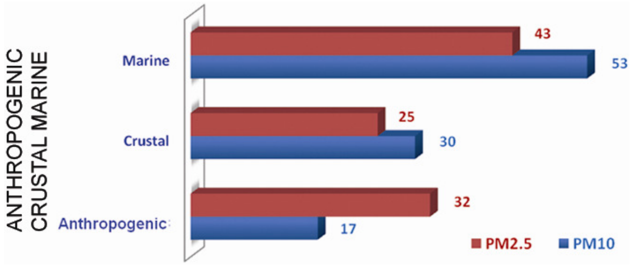

Fig. 6 — Contribution of different sources to the PM composition at Sinhagad

3.4.2 Calculation of the non-crustal fractions

After calculating the sea salt and non-sea salt fractions, crustal fraction for different chemical components have been computed by using Ca^{2+} as a reference element for crustal source. For example, Ncr SO_4 is computed as:

$$\text{Ncr } SO_4 = [NssSO_4] - \{([SO_4]/[Ca])_{crust} * [Ca]\}$$

where, $([SO_4]/[Ca])_{crust}$ is the standard ratio of concentration of SO_4^{2-} and Ca^{2+}, which is obtained from crustal composition[4,41]. Similarly, Ncr fractions have been computed for the other chemical components. Crustal (cr) fractions for these components were computed as:

$$Cr = \{([X]/[Ca])_{crust} * [Ca]\}$$

Thus, the total concentration from crustal source comprised of the sum total of crustal fractions of all the chemical components added to the concentration of Ca^{2+}. It was observed that the crustal fractions of Ca^{2+}, K^+, Mg^{2+} and SO_4^{2-} were in measurable amount. Similarly, the marine source comprised of the sum total of sea salt fractions of all the chemical components added to the concentration of Na^+. Finally, the anthropogenic source comprised of sum total of the non-sea salt and non-crustal fractions of all the above mentioned chemical components and then concentrations of NO_3^- and NH_4^+ added to it (as these components did not show any crustal or marine contribution).

Quantification of the respective contributions of these three sources: crustal, marine and anthropogenic to the total measured chemical composition of aerosols has been depicted in Fig. 6. It was observed that during the sampling period, major contribution was from marine sources (43% for $PM_{2.5}$ and 53% for PM_{10}) followed by crustal source [contributed more in PM_{10} (30%) than in $PM_{2.5}$ (25%)], and anthropogenic source [contributed more in $PM_{2.5}$ (32%) than in PM_{10} 17%)].

4 Conclusions

The present study presents chemical characteristics of aerosols (PM_{10} and $PM_{2.5}$) at Sinhagad, a rural high altitude location, during November 2008 - April 2009. The average concentrations of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ at Sinhagad were found to be 35.8, 14.1 and 21.7 µg m^{-3}, respectively. The average ratio of $PM_{2.5}/PM_{10}$ (0.39) suggested that PM_{10} at study area is dominated by primary particulate emissions by natural and man-made activities. Contribution of anions and cations in $PM_{2.5}$ was about 43% and 57%; while for PM_{10}, it was 33% and 67%, respectively. Contribution of marine components, Na^+ and Cl^- and Mg^{2+} were dominant in both sizes by 58% in PM_{10} and 49% in $PM_{2.5.}$ In $PM_{2.5}$, SO_4^{2-}, NO_3^- and NH_4^+ were also having significant contribution after sea salt, which may due to the anthropogenic emissions primarily from burning of fossil fuel and biomass components and human activities. Considerable amounts of Cl^-, Na^+, Ca^{2+}, Mg^{2+} and K^+ were also found in PM_{10} which may be attributed to natural sources (like soil). In PM_{10}, all the ions exhibited higher concentration in summer because of the unstable atmosphere due to convection. In $PM_{2.5}$, the concentration of anthropogenic components like SO_4^{2-}, NO_3^- and NH_4^+ were higher in winter as these are formed through the gas-to-particle conversion processes. The order of neutralization factor was found to be $Ca^{2+} > Mg^{2+} > NH_4^+ > K^+$ for PM_{10}; and $Ca^{2+} > NH_4^+ > Mg^{2+} > K^+$ for $PM_{2.5}$. It was observed that during the sampling period, major contribution was from marine sources (about 43 % for $PM_{2.5}$ and 53% for PM_{10}) followed by crustal sources ($PM_{2.5}$: 25% and PM_{10}: 30%) and anthropogenic sources ($PM_{2.5}$: 32% and PM_{10}:17%). The archival of composite data sets and the results obtained from these observations will be helpful to assess the impact of anthropogenic sources on aerosol composition and long-range transport of certain pollutants.

Acknowledgement

The authors are grateful to University Grants Commission, New Delhi and Board of College and University Development, Pune for providing the financial assistance. Authors are also thankful to the Head, Department of Chemistry, University of Pune; Director, IITM and Head, PM & A Division, IITM, Pune for providing the necessary infrastructural facilities. Authors are also thankful to BSNL authorities, Pune for their cooperation and providing space at their Microwave Tower station at Sinhagad.

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