Anthropogenic fine aerosols dominate over the Pune region, Southwest India

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Abstract

The major water-soluble ions, organic carbon (OC), elemental carbon (EC) and mass concentration of fine- ($\text{PM}_{2.5}$) and coarse-mode ($\text{PM}_{10-2.5}$) aerosols were measured at Pune during January–December 2016. The mass closure approach was used by comparing the sum of the masses of the individual chemical species to the gravimetric PM obtained by weighting the filter samples. The 1 year mean total mass concentration of fine and coarse mode was $40 \mu\text{g m}^{-3}$ (64%) and $23 \mu\text{g m}^{-3}$ (36%), respectively. The $\text{PM}_{2.5}/\text{PM}_{10}$ ratio was 0.64 ± 0.9 indicating an abundance of fine-mode particles over Pune during the study period. A principal component analysis identified three components, where the one with highest explanatory power (59%) displayed clear impact of anthropogenic sources on the measured mass concentration of a majority of the compounds. The strong linear relationships between EC, OC, nss-SO_4^{2-} and nss-K^+ suggest a predominance of a common primary source, with a contribution from biofuel as well as biomass burning sources. Keeping the strong correlation and sources of individual chemical species as the base, it was noticed that (1) major contributors to fine- and coarse-mode particles over the Pune regions are carbonaceous aerosols and secondary inorganic aerosols (non-sea-salt SO_4^{2-} , NO_3^- , and NH_4^+), (2) anthropogenic aerosols contribute mostly to the fine-mode, and (3) meteorological parameters play an important role in controlling levels of fine- and coarse-mode particles. Taken together, the study clearly indicates the dominance of anthropogenic sources during the entire year with more significance in the winter season.

1 Introduction

The rapid growth in emerging economies over the last decades has led to dangerous levels of air pollution throughout Asia (Grandey and Cheng 2016; Cohen et al. 2017; Lelieveld et al. 2018). As a result, air pollution levels in many South Asian cities are above the World Health Organization guideline values (WHO 2018). The human population of South Asia is about one-fifth of the world population (Im et al. 2017). In addition to contributing to a positive radiative forcing (ranging ~ 0.2 to 1.2 W/m^2), these atmospheric

components also have multiple secondary effects, including surface dimming (affecting agricultural crop yields), glacier melt and shifts of the storm frequency and the monsoon system (Haywood and Boucher 2000; Bollasina et al. 2011; Atkinson et al. 2014; Boucher et al. 2013; Cohen et al. 2017; WHO 2018).

Airborne particulate matter (PM) is usually divided and nominated by its aerodynamic diameter, and the most widely monitored particles are fine- ($\text{PM}_{2.5}$; particle aerodynamic diameter $< 2.5 \mu\text{m}$) and coarse-mode particles ($\text{PM}_{10-2.5}$; particles aerodynamic diameter between $2.5 \mu\text{m}$ and $10 \mu\text{m}$). Fine particles, for instance, are of greatest interest with respect to health effects (Gent et al. 2009; Atkinson et al. 2014; WHO 2018) and other environmental impacts such as visibility degradation, radiative forcing and climate change (Boucher et al. 2013). Monitoring of both fine- and coarse-size particles is useful in controlling risk to human beings and for developing efficient PM control strategies.

The fine particles originating from anthropogenic and natural sources are released directly from the sources or are formed in the atmosphere largely through a photochemical reaction from precursor gases (Lee et al. 2014). Thus,

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they are composed mainly of sulfate, ammonium, organic compounds, elemental carbon and metals. Coarse-mode particles rise in the air with a wide range of processes such as mechanical grinding, windblown dust, sea spray, non-exhaust vehicle emissions, agriculture, construction and quarrying activities which produce these particles (Charron and Harrison 2005; Kassomenos et al. 2012). Concentration and distribution of carbon aerosols and water-soluble inorganic species (WSIS) associated with fine- and coarse-mode particles can provide indications of source contributions, chemical transformations in the atmosphere and potential adverse effects.

Tackling the sources of air pollution requires systematic collection of air quality data and a scientific approach to air quality management. The present paper describes year-round data sets on fine- and coarse-mode aerosol mass with major organic and inorganic chemical constituents collected in Pune, India. A size-fractionated air filter sampling allows the identification of the (1) size distribution of water-soluble and -insoluble chemical species, (2) contribution of each an individual component to the coarse and fine mode, and (3) impact of meteorological processes on the observed concentration levels in PM with the help of air mass back trajectories and local meteorological parameters.

2 Materials and methods

2.1 PM sampling site

Pune (18°32'N, 73°51'E, 559 m amsl) is one of the rapidly developing cities in South Asia. It is located in the Deccan Plateau, about 100 km east of Konkan coast adjoining the Arabian Sea and at a distance of about 160 km from Mumbai, the largest and economically most important city in the India. Today, Pune is India's eighth largest urban agglomeration with 5 million inhabitants (Butsch et al. 2017). The city boasts of having a maximum density of vehicles in the country, and vehicle population is equal to the city population (The Pune Municipal Corporation report).

Aerosol filter samples were collected at the rooftop of the Indian Institute of Tropical Meteorology (IITM), at about 15 m height above the ground in Pashan, an urban site at about 6 km to the northwest of the main city of Pune. Sampling of PM₁₀ and PM_{2.5} was carried out using a dust sampler (NETEL India Pvt Ltd). Sample collection duration was set ~24 h with a flow rate of 16.7 LPM. A total of 99 samples were collected using pre-weighed and pre-combusted 47 mm-diameter quartz filters. All possible precautions were taken at every step in the collection of samples to minimize any contamination. Based on the season and air mass back

Fig. 1 Clustered 6-hourly, 7-day air mass back trajectories' (AMBTs) mean for Pune (total $n=396$). The cluster means lines colored on the map indicate three different source regions: Indian Subcontinent (red), Arabian Peninsula (purple) and Indian Ocean (blue)

trajectory cluster, we classified the samples into three groups as shown in Fig. 1.

2.2 Gravimetric and chemical analysis

Filter papers were weighed thrice before and after sampling using an electronic balance with a detection limit of 0.01 mg after stabilizing in a desiccator at a constant temperature ($20 \pm 2^\circ\text{C}$) and relative humidity ($40\% \pm 3\%$) for 24 h. This insures that all filter samples were brought to the same degree of dryness before being weighed. Handling and loading of filter papers on the equipment were made using hand gloves and cleaned forceps. After collection of the samples, the filter papers were packed in polyethylene ziplock bags. Afterward, the samples were extracted with precaution for the chemical analyses. Field blanks were taken frequently (every month, $n=11$) and analyzed using procedures similar to those adopted for samples and necessary correction factors were taken into consideration while the computation of ionic concentrations.

For chemical analysis, filters were extracted with ultrapure water (40 ml) using ultrasonic bath and were analyzed for major water-soluble ions. The anions (F^- , Cl^- , NO_3^- , SO_4^{2-}) were analyzed by using ion chromatograph (IC100, Dionex Co. Ltd.) and the cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}) were determined by using atomic absorption spectrophotometer (Perkin-Elmer A-Analyst, 400) with an air-acetylene flame. The error in analytical reproducibility for the major elements ranges from 0 to 5%. In addition, the institute (IITM) has been participating in the international inter-comparison studies, i.e., EANET's Inter-laboratory

comparison projects and WMO's Laboratory Inter-comparison studies (LIS). The results from these comparison studies showed good agreement, i.e., the variation is about $\pm 10\%$. The detailed description of the extraction procedure and the detection limits for IC and AAS analysis are given in Budhavant et al. (2016). The NH_4^+ ion was measured by colorimetric method (Weatherburn 1967) using a double beam spectrophotometer (Spectronic 20D, Milton Roy Company, USA).

The OC and EC contents were measured using an OC/EC Analyzer of Sunset Laboratory, USA (Model 4G), by selecting the National Institute for Occupational Safety and Health (NIOSH) 5040 method based on thermal optical transmittance (TOT) (Birch and Cary 1996). The analyzer was calibrated periodically for quality control purposes by using a blank punch of pre-heated quartz fiber filter and standard sucrose solutions. The average relative standard deviation of triplicate analysis was 5% for EC and 7% for OC. The average blank concentrations from the quartz filters for OC was 0.26 ± 0.10 ($\mu\text{g}/\text{cm}^2$) and $\text{EC} < 0.01$ $\mu\text{g}/\text{cm}^2$. Details on this technique and related uncertainties have been discussed in other studies (Safai et al. 2013, 2014).

2.3 Meteorology

The study region is under the influence of the monsoon system, leading to dynamic changes in meteorological conditions from season to season (Fig. 1). The NOAA HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory (Stein et al. 2016) model was used to calculate 7-day air mass back trajectories (the interval of 6 h) at an arrival height of 600 m over the sampling site.

Based on cluster analysis of the HYSPLIT, air mass back trajectory files yielded that the air parcels sampled over Pune during January–December 2016 were grouped into three classes (Fig. 1). These originated from the Indian Subcontinent (hereafter referred to as group 1, red color), the Arabian Peninsula (group 2, purple) and the Indian Ocean (group 3, blue). About 38% each of the received air parcels at Pune were classified as group 1 and of Africa and Arabia origin, and about 42% with the remaining 20% occurrences were of Indian Ocean origin. In monsoon, the predominant winds are from the oceanic regions (i.e., the Arabian Sea and Indian Ocean) with very limited contact with anthropogenic sources. In wintertime, air is transported mainly from the Indo-Gangetic Plain (IGP), parts of Central India and Bay of Bengal to the sampling site. By combining mass size distribution spectra of aerosols and meteorology, we can better understand the sources and transformation of the measured aerosol components.

Transport on local scales (< 10 km) surrounding the site is better represented by surface wind observations at the site (Fig. 2). The local meteorological conditions (wind speed

Fig. 2 Distribution of wind speed and wind direction measured at Pune during the sampling period

and direction, temperature, relative humidity and rainfall) are obtained from the Indian Meteorological Department (IMD) at the Pashan site (within 1 km of sampling station). The daily mean relative humidity was 68% and varied in the range 23–100%. The daily mean temperature was 25 °C and varied in the range 10–33 °C. The average wind speed during the sampling period was observed to be 3.3 m/s (Fig. 2).

2.4 Principal component analysis (PCA)

Source assessment for different chemical species in PM_{10} samples was carried out based on principal component analysis (PCA). PCA is a mathematical procedure that transforms a number of correlated variables into a number of uncorrelated variables called principal components. PCA has been an extensively used tool to reduce the complexity of atmospheric data and has been applied in several studies on aerosol chemical composition (Safai et al. 2010; Budhavant et al. 2011; Martinsson et al. 2017). PCA with VARIMAX rotation was performed by using the software SPSS (Statistical Package for the Social Sciences). A VARIMAX is a method in which uncorrelated factors are sought by a rotation that maximizes the variance of the factor loadings. Only the factors corresponding to eigenvalue greater than or equal to one were considered.

3 Result and discussion

3.1 Mass closure

The mass closure approach was studied by comparing the sum of the masses of the individually identified chemical

Fig. 3 Mass closure of particulate matter in coarse mode ($PM_{10-2.5}$) and fine mode ($PM_{2.5}$) measured at Pune during January–December 2016

species to the gravimetric PM obtained by weighing the filter samples (Fig. 3). The mass of chemical components was calculated by adding all measured water-soluble ions, EC and organic matter (OM). To estimate the contribution of OM to particulate mass concentration, a conversion factor 1.6 ± 0.2 for converting OC to OM was suggested for urban aerosol (Turpin and Lim 2001).

To see the effect of marine influence on the fine and coarse aerosol composition, sea-salt ratios were calculated using Na^+ as reference (Keene et al. 1986). The non-sea-salt (nss) fraction of SO_4^{2-} , K^+ and Ca^{2+} were calculated by subtracting the sea-salt fractions from their measured concentrations. Further, the chemical components were divided into four classes: carbonaceous aerosols (EC + OM), secondary inorganic aerosols (nss- SO_4^{2-} , NO_3^- , and NH_4^+) (SIA), sea salt and dust. The remaining unaccounted $PM_{2.5}$ (30%) and PM_{10} (35%) probably resulted from unmeasured carbonates, trace elements and aerosol-bound water. The contribution of particle-bound water can constitute up to 20–35% of the annual mean PM_{10} and $PM_{2.5}$ concentrations (Tsyro 2005).

3.2 Particulate mass

Particle mass and size are the fundamental properties of an aerosol. In addition, the chemical composition of atmospheric aerosols contributes to the diverse processes that continuously determine how the ever-changing aerosol population evolves. Figure 4 depicts the time series plot of PM mass for the study period at Pune. The average concentrations of $PM_{2.5}$ and PM_{10} aerosol were $40.2 \pm 14.3 \mu g m^{-3}$ (mean $\pm$ standard deviation) and $63.0 \pm 25.1 \mu g m^{-3}$,

respectively, during the year 2016. The estimated mass of coarse fraction ($22.8 \pm 13.9 \mu g m^{-3}$) was made by subtracting $PM_{2.5}$ from PM_{10} (Table 1). These observed values of PM are slightly higher than the annual standard stipulated by the Indian National Ambient Air Quality Standard (NAAQS) ($40 \mu g m^{-3}$ for $PM_{2.5}$ and $60 \mu g m^{-3}$ for PM_{10}) and comparatively more than the threshold limit set by the World Health Organization (WHO) ($25 \mu g m^{-3}$ for $PM_{2.5}$ and $50 \mu g m^{-3}$ for PM_{10}). The PM mass concentrations showed strong seasonal variation (Figs. 3, 4), with elevated concentrations during the winter ($PM_{2.5}$ $49.4 \mu g m^{-3}$, PM_{10} $81.1 \mu g m^{-3}$) season followed by summer ($PM_{2.5}$ $40.5 \mu g m^{-3}$, PM_{10} $61.5 \mu g m^{-3}$), and low concentration during the monsoon ($PM_{2.5}$ $26.5 \mu g m^{-3}$, PM_{10} $40.4 \mu g m^{-3}$). Variations in mass concentrations of $PM_{2.5}$ and PM_{10} were observed due to seasonal variation of different emission sources and differences in local meteorological conditions at the sampling site.

To understand the contributions of $PM_{2.5}$ – PM_{10} , the $PM_{2.5}/PM_{10}$ ratio is plotted in Fig. 4. This ratio can provide crucial information relating to the particle origin, formation process and effects on human health (Speranza et al. 2014). Higher ratios of $PM_{2.5}/PM_{10}$ attribute particle pollution to anthropogenic sources and smaller ratios indicate considerable involvement of coarse particles, which might be related to natural sources, e.g., dust storm (Sugimoto et al. 2016) and sea salt (Budhavant et al. 2011, 2015a). The $PM_{2.5}/PM_{10}$ ratio is often used to characterize the underlying atmospheric processes within the local environment (Yu and Wang 2010; Chu et al. 2015). In the present study, the $PM_{2.5}/PM_{10}$ ratio was 0.64 ± 0.09 with the highest monthly average ratio of 0.71 ± 0.09 (October) and the lowest ratio of

Fig. 4 Daily average variations of fine- ($\text{PM}_{2.5}$, white area) and coarse-mode aerosols ($\text{PM}_{10-2.5}$, gray color) and $\text{PM}_{2.5}/\text{PM}_{10}$ (blue color) ratio measured at Pune during January–December 2016

Table 1 Comparison of aerosol mass and concentrations of carbonaceous aerosols ($\mu\text{g}/\text{m}^3$) with other sites in the world

Location	$\text{PM}_{2.5}/\text{PM}_{10}$ (mass)	$\text{PM}_{2.5}$		PM_{10}		References
		OC	EC	OC	EC	
Pune, India	0.64	9.71	3.11	10.7	3.35	Present study
Ahmedabad, India	0.33	18.3	3.0	29.8	4.4	Rengarajan et al. (2011)
Tianjin, China	0.53	14.5	4.3	32.6	6.1	Li et al. (2012)
Karachi, Pakistan	0.17	9.7	1.9	21.8	4.7	Shahid et al. (2016)
Beijing, China	0.59	13.7	7.4	20.9	9.4	Zhang et al. (2008)
Beijing, China	0.58	19.2	7.0	23.5	8.5	Chan et al. (2005)
Lhasa, China	0.63	3.27	2.24	4.74	2.31	Li et al. (2016)
Tianjin, China	0.56	16.9	5.7	30.8	8.9	Gu et al. (2010)
Beijing, China	0.57	13.7	7.4	21	9.4	Wang et al. (2014)
Mahabubnagar, India	0.78	8.65	1.52	14.05	2.12	Bisht et al. (2015)

0.50 ± 0.04 (January). However, the ratio is high during post-monsoon (Oct–Nov) and pre-monsoon (Mar–April) months, clearly indicating larger fine particle fractions in PM_{10} at this location. High fractions of fine-mode particles are attributed to low-level inversion and favorable meteorological conditions for the accumulation of pollutants in the lower atmosphere. These fine-mode aerosols are mainly emitted from various anthropogenic sources such as biomass burning in open fields, domestic fuel in rural settings, emissions from brick kilns, fossil fuel burning and vehicular exhausts. The observed $\text{PM}_{2.5}/\text{PM}_{10}$ ratio of 0.64 in the present study was comparable with that of other cities in Asia such as Beijing (0.59, Zhang et al. 2008), Lhasa (0.63, Li et al. 2016), Tianjin (0.56, Gu et al. 2010) and Mahabubnagar (0.78, Bisht et al. 2015).

3.3 Organic and elemental carbon

The distribution and properties of the carbon aerosol fraction are important as they have a proportionally much larger impact on the aerosol absorption of solar radiation. The total carbonaceous fraction of aerosols is typically divided into organic carbon (OC) and elemental carbon (EC). EC normally comprises 3–10% of the total aerosol mass in South Asia (e.g., Lelieveld et al. 2001; Budhavant et al. 2015b; Gawhane et al. 2017). The mean concentration of EC was found to be $3.11 \pm 2.38 \mu\text{g}/\text{m}^3$ in $\text{PM}_{2.5}$ and $3.35 \pm 2.49 \mu\text{g}/\text{m}^3$ in PM_{10} (Table 1). EC is a primary pollutant emitted from numerous combustion-related sources (Bond et al. 2013). Since EC undergoes minimal chemical transformation, it serves as a good tracer for anthropogenic aerosols (Budhavant et al. 2018). The EC concentration reported in this study is nearly the same as 6 years (2005–2010) mean BC ($3.58 \pm 1.55 \mu\text{g}/\text{m}^3$, measured with an aethalometer at

880 nm) reported from the station (Safai et al. 2013) with maximum concentration during winter and minimum during the monsoon season in each year.

Organic carbon (OC) usually comprises the largest carbon-containing fraction of ambient aerosols. In addition to incomplete combustion processes, OC is also emitted from biogenic sources. The concentrations of OC in Pune showed a similar seasonal pattern to EC (Fig. 5). A significant correlation between OC and EC ($R^2=0.87$ for $PM_{2.5}$ and $R^2=0.90$ for PM_{10}) was observed during the study period, suggesting their common emission sources (Fig. 5). The mean EC and OC concentrations measured in $PM_{2.5}$ and PM_{10} in Pune were compared with that reported in some other locations in South Asia (Table 1). The total loading of the carbonaceous aerosol may be estimated as total carbon (TC=EC+OC). The monthly average highest concentration of TC was observed in December, $26.10 \pm 5.78 \mu\text{g}/\text{m}^3$, whereas the lowest values were found in the month of July, $5.01 \pm 0.97 \mu\text{g}/\text{m}^3$ for PM_{10} .

3.4 Water-soluble inorganic composition

A summary of the WSIS concentrations associated with fine- and coarse-mode particle was given in Table 2. The ion balance expressed by the sum of the concentration (in equivalent unit) of cation to anion is a good indicator to study the acidity of the environment (Dentener et al. 1996; Hillamo et al. 1998; Kerminen et al. 2001). The coarse-mode particulate cation-to-anion ratios in most samples were close to unity or higher (avg. 1.23 ± 0.29), but the fine-mode particulate cation-to-anion ratios were close to unity or lower (avg. 0.92 ± 0.15). The anion deficiency in

coarse particles is thought to have been caused by undetected carbonate ion which is originated from construction activities and road dust (Chebbi and Carlier 1996; Wall et al. 1988; Clarke and Karani 1992; Dentener et al. 1996), whereas the missing cation contribution in fine particles was most likely the hydrogen ion (Hillamo et al. 1998; Kerminen et al. 2001), showing that the coarse particles in most samples were neutral or alkaline and the fine particles in most samples were neutral or acidic. The alkaline coarse particles would provide reactive sites for absorbing acidic gases in the atmosphere and therefore speed up their scavenging processes. However, the acidic nature of fine particles could add extra adverse health effects (Wang and Shooter 2002).

The most dominant ion was SO_4^{2-} with an average $3.95 \mu\text{g}/\text{m}^3$ (17%), followed by Cl^- (9%), Na^+ (6%), NO_3^- (5%) and Ca^{2+} (3%) of the total measured composition of fine particulate matter. K^+ (1.9%), NH_4^+ (1.5%) and Mg^{2+} (1.4%) had minor contributions (Fig. 6), whereas Cl^- was the most dominant ion in coarse-mode particles with an average value of $2.89 \mu\text{g}/\text{m}^3$ (26%), followed by SO_4^{2-} (22%), Na^+ (15%), Ca^{2+} (10%) and NO_3^- (7%). The dominance of nss- SO_4^{2-} , EC and OC to the fine fractions indicates a large contribution from anthropogenic sources for $PM_{2.5}$, whereas marine aerosol and suspended dust contributed more for coarse particles (Fig. 3). Potassium was equally distributed between both size ranges. Sulfate, nitrate and ammonium were considered secondary inorganic species that were formed in the atmosphere from gas-phase precursors (Stone et al. 2010). The same kind of results have been reported by earlier studies (Kocak et al. 2007; Wang and Shooter 2002; Kothai et al. 2011; Choung et al. 2016;

Fig. 5 Time series of organic carbon (OC), elemental carbon (EC) concentrations and OC/EC ratio in $PM_{2.5}$ (red color line) and PM_{10} (black color line) in Pune during January–December 2016

Table 2 Comparison of aerosol mass and inorganic composition ($\mu\text{g}/\text{m}^3$) at different sites in the world

Site	Coarse (PM _{10-2.5})							Fine (PM _{2.5})							References
	Mass	Mg ²⁺	K ⁺	Ca ²⁺	NH ₄ ⁺	NO ₃ ⁻	SO ₄ ²⁻	Mass	Mg ²⁺	K ⁺	Ca ²⁺	NH ₄ ⁺	NO ₃ ⁻	SO ₄ ²⁻	
Pune, India	23.0	0.41	0.44	1.08	0.21	0.74	2.47	40	0.32	0.43	0.60	0.34	1.06	3.95	Present Study
Alberta, Canada	7.6	0.07	0.09	0.39	-	-	-	8.59	0.01	0.04	0.05	-	-	-	
Klang Valley, Malaysia	22.5	0.23	0.41	0.98	-	-	-	26.9	0.12	0.39	0.13	-	-	-	Rahman et al. (2011)
Vasi, India	71.0	0.68	0.43	2.40	-	-	-	42.0	0.22	0.28	0.31	-	-	-	Kothai et al. (2011)
Raipur, India	189	2.61	4.65	16.4	0.38	3.84	10.0	125	1.69	5.90	10.2	8.76	8.16	46.5	Verma et al. (2010)
Ashdod, Israel	39.6	0.43	0.43	4.20	-	-	-	21.2	0.09	0.20	0.46	-	-	-	Mamane et al. (2008)
Rio de Janeiro, Brazil	26.0	0.49	0.25	0.26	0.32	1.39	1.12	17.0	0.05	0.24	0.05	0.94	0.40	2.56	Mariani and de Mello (2007)
Beirut, Lebanon	44.5	0.14	0.07	2.98	0.18	1.74	1.72	26.2	0.03	0.08	0.52	0.77	0.18	4.27	Kouyoumdjian and Saliba (2005)
Auckland, New Zealand	9.9	0.16	0.06	0.10	-	0.19	0.38	8.23	0.05	0.05	0.03	0.08	0.23	0.50	Wang and Shooter (2005)
Bobadela, Lisbon	16.0	0.11	0.04	0.43	0.02	1.00	0.48	24.0	0.05	0.13	0.38	1.30	1.30	3.90	Almeida et al. (2005)
Monagrega EbroBasin, Spain	17.7	0.14	-	0.99	0.88	1.1	3.85	12.8	0.04	-	0.27	1.1	0.2	3.61	Alastuey et al. (2004)
Nanjing, China	316	0.38	3.81	4.58	10.8	9.19	18.1	225	6.14	3.29	1.72	9.49	7.46	16.3	Wang et al. (2003)
Barcelona, Spain	49.6	0.28	0.42	2.26	2.67	5.78	6.66	33.4	0.07	0.23	0.52	3.2	4.21	5.57	Rodriguez et al. (2002)

Karydis et al. 2010). All the chemical components showed low concentrations during the monsoon season, mainly due to the washout effect of rains and due to prevailing SW winds that arrive from the Indian Ocean and Arabian Sea bringing marine air masses. The present results were also compared with other studies reported from India and other part of the world (Table 3).

3.5 Source identification

Principal component analysis allowed us to deduce three significant sources affecting the observational site that accounted for 91% of the variance (Table 3). The first factor accounts for 59% of the total variance and has a strong positive correlation (Table 4) and contributions from EC, OC, NO_3^- , NH_4^+ , SO_4^{2-} , K^+ , and Mg^{2+} that suggests the mixed impact of anthropogenic activities such as fossil fuel burning from industries and vehicles, as well as other biomass burning activities. The second factor accounts for 21%, includes loadings for Na^+ and Cl^- and shows that the dominance of marine components originated from the sea regions. However, the third factor (12%) consisted of Ca^{2+} and indicated that the source was suspended soil and road dust, mainly in the coarse fraction sources. The next step was identification of the possible contribution of these sources of fine- and coarse-mode aerosols.

3.5.1 Carbonaceous aerosols

The mass ratio of OC to EC reflects multiple processes in the atmosphere, e.g., the OC/EC ratio is typically higher from biomass combustion than from fossil source emissions (Cachier et al. 1996; Budhavant et al. 2015b), it is elevated by biogenic SOA contributions (Chow et al. 1993, 1996; Saarikoski et al. 2008) and it is affected by atmospheric processing (aging) of organic chemicals (Kroll et al. 2011). In the present study, the measured average OC/EC ratios were 4.17 ± 2.31 for $\text{PM}_{2.5}$ and 4.26 ± 2.16 for PM_{10} . The OC/EC ratios ranged from 1.8 to 10.9 for $\text{PM}_{2.5}$ and 1.95 to 11.1 for PM_{10} (Fig. 5). The OC/EC ratios are usually in the range of 2–3 in urban cities, whereas higher ratios were reported for wood combustion, residential heating, forest fires and road dust (Schauer et al. 2001; Zhang et al. 2007; Feng et al. 2009; Achilleos et al. 2016). The co-variability of OC and EC together with their significant linear relationship and their near-exclusive partitioning to the fine $\text{PM}_{2.5}$ fraction suggests that the ambient concentration levels of both EC and OC were controlled largely by primary emissions and atmospheric dispersion.

Higher concentration of OC and EC were observed during winter months (Fig. 5), which may be due to more biomass burning along with some meteorological factors (low temperature and wind speed). Other processes that can

Fig. 6 Organic carbon (OC), elemental carbon (EC) and ionic concentrations in fine- (light gray) and coarse-mode (dark gray with lines) aerosols measured at Pune during January–December 2016

Table 3 Principal component analysis (PCA) factor loadings for different chemical components of PM₁₀

Chemical component	Factor loadings component		
	1	2	3
	Anthropogenic	Sea	Soil
EC	0.93		
OC	0.95		
NO ₃ [−]	0.94		
NH ₄ ⁺	0.84		
SO ₄ ^{2−}	0.95		
Na ⁺		0.99	
Cl [−]		0.98	
K ⁺	0.92		
Ca ²⁺			0.97
Mg ²⁺	0.65		
Eigen value	5.85	2.07	1.15
% of variance	58.5	20.7	11.5
Cumulative %	58.5	70.1	90.6

contribute to cold-season OC levels are the adsorption of semi-volatile OCs onto existing solid particles and the dissolution of soluble gases that can undergo reactions in particles (Pandis et al. 1992; Xu et al. 2012; Pipal and Satsangi 2015). The lower OC and EC values in monsoon months are mainly associated with the wet removal of aerosols by precipitation. The major source contributions of carbonaceous aerosols reported from South Asia in cities like Ahmedabad (Rengarajan et al. 2011), Mahabubnagar (Bisht et al. 2015), and Karachi (Shahid et al. 2016) are from biomass burning. On the other hand, fossil fuel combustion was expected to be the major source of EC and OC in mega cities such as Delhi (Saxena et al. 2017), Lahore (Alam et al. 2014) and Dhaka (Begum et al. 2012). A year-round study using source-diagnostic ¹⁴C-EC shows that both fossil and biomass combustion processes are about equally responsible for the emission of EC over South Asia (Gustafsson et al. 2009; Budhavant et al. 2015c).

Table 4 Matrix of correlation coefficients for the components in PM₁₀

	EC	OC	NO ₃ [−]	NH ₄ ⁺	SO ₄ ^{2−}	Na ⁺	Cl [−]	K ⁺	Ca ²⁺	Mg ²⁺
EC	1.00	0.95	0.83	0.78	0.84	0.17	0.10	0.82	−0.13	0.54
OC	0.95	1.00	0.86	0.76	0.86	0.19	0.12	0.85	−0.10	0.51
NO ₃ [−]	0.83	0.86	1.00	0.82	0.91	−0.01	−0.04	0.82	−0.02	0.43
NH ₄ ⁺	0.78	0.76	0.82	1.00	0.76	0.07	−0.02	0.71	−0.26	0.41
SO ₄ ^{2−}	0.84	0.86	0.91	0.76	1.00	0.15	0.14	0.90	0.10	0.60
Na ⁺	0.17	0.19	−0.01	0.07	0.15	1.00	0.96	0.13	0.19	0.45
Cl [−]	0.10	0.12	−0.04	−0.02	0.14	0.96	1.00	0.10	0.27	0.40
K ⁺	0.82	0.85	0.82	0.71	0.90	0.13	0.10	1.00	0.03	0.61
Ca ²⁺	−0.13	−0.10	−0.02	−0.26	0.10	0.19	0.27	0.03	1.00	0.18
Mg ²⁺	0.54	0.51	0.43	0.41	0.60	0.45	0.40	0.61	0.18	1.00

Correlations higher than 0.6 are highlighted in bold

3.5.2 Secondary inorganic aerosol

Secondary inorganic aerosol (SIA) is the second major component of fine (11%) and coarse (8%) particles in Pune. The annual SIA concentration in Pune was $5.02 \mu\text{g}/\text{m}^3$ in fine particles, with strong seasonal variation (Fig. 3): highest in winter ($7.15 \mu\text{g}/\text{m}^3$) and lowest in monsoon ($1.95 \mu\text{g}/\text{m}^3$), whereas the annual SIA concentration in PM_{10} was $8.03 \mu\text{g}/\text{m}^3$, highest in winter ($14.9 \mu\text{g}/\text{m}^3$) and lowest in monsoon ($2.26 \mu\text{g}/\text{m}^3$). Atmospheric secondary SO_4^{2-} and NO_3^- aerosols result primarily from the oxidation of sulfur dioxide and nitrogen oxides. The mass ratio of $\text{NO}_3^-/\text{SO}_4^{2-}$ could be used as an indicator of the relative importance of mobile and stationary sources of sulfur and nitrogen in the atmosphere (He et al. 2017). The mass ratios of $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ higher than unity indicated that the particles were mainly from mobile sources (vehicular activity). Conversely, the mass ratios of $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ lower than unity suggested that the sources of particles came mainly from stationary sources (industrial activities). In this study, the average ratio of $\text{NO}_3^-/\text{SO}_4^{2-}$ was 0.28 ± 0.12 and 0.44 ± 0.33 in fine and coarse particles, indicating a greater contribution of SO_4^{2-} that shows the dominance of stationary sources compared to vehicular activity. Earlier studies reported a high average $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio in cities such as Bobadela (Almeida et al. 2005), Rio de Janeiro (Mariani and de Mello 2007) and Beirut (Kouyoumdjian and Saliba 2005) (Table 2).

The strong positive correlation between NO_3^- and SO_4^{2-} in the coarse- ($R^2=0.81$) and fine-mode ($R^2=0.65$) particles indicates same anthropogenic origin over the Pune region. A positive correlation between NO_3^- and SO_4^{2-} concentrations in the atmosphere has often been found (Harrison and Plo 1983; Wakamatsu et al. 1996; Pathak et al. 2009). However, strong negative correlation between SO_4^{2-} and NO_3^- has also been reported on clear days, hazy days, foggy days, and respirable particulate air pollution days (Kong et al. 2014).

3.5.3 Sea salt and crustal aerosols

The percentage contributions from different sources to the chemical ions in PM were calculated by using the crustal and marine fractions. Na^+ was assumed to have originated fully from marine source and nss-Ca^{2+} was from crustal source. The nitrogenous ionic constituents such as NO_3^- and NH_4^+ were contributed primarily by anthropogenic sources and the contribution of natural sources was almost nil.

Sea salt ($\text{Na}^+ + \text{ss-Cl}^- + \text{ss-Mg}^{2+} + \text{ss-Ca}^{2+} + \text{ss-K}^+$) contributed substantially to coarse particle mass, accounting for 17% of the total mass. Sea salt-associated ions (mainly Na^+ , Cl^-) illustrate strong internal correlations ($R^2=0.93$) and nearly zero intercept in PM_{10} , denoting that they primarily

originate from sea salt. The sea-salt aerosols had the highest concentrations in monsoon (25%) under the influence of air mass originating from over the Arabian Sea before arriving at the sampling site. A crustal source was observed to be the dominant source for Ca^{2+} aerosols in the Indian Subcontinent (Kulshrestha et al. 1998; Safai et al. 2010). In this study, soil/dust factor contributed the least, accounting for less than 2% $\text{PM}_{2.5}$ mass and 5% of total PM_{10} mass. The contribution of dust was found to be higher in the monsoon (6% in PM_{10} and 3% in $\text{PM}_{2.5}$), followed by winter (5% in PM_{10} and 3% in $\text{PM}_{2.5}$) and summer (4% in PM_{10} and 2% in $\text{PM}_{2.5}$).

4 Conclusions

The primary strengths of this study are the simultaneous collection (a year-round) of the fine- ($\text{PM}_{2.5}$) and coarse-mode ($\text{PM}_{10-2.5}$) particles at Pune, India. The observed values of PM are slightly higher than the annual standard stipulated by Indian National Ambient Air Quality Standard, and comparatively more than the threshold limit set by the World Health Organization. The mass balance of the determined components indicated that anthropogenic aerosols were the largest contributor to PM mass in Pune with a share of 55% of the fine-mode and 51% of the coarse-mode mass. The second largest contributor is the sea salt (11% of $\text{PM}_{2.5}$ and 16% of $\text{PM}_{10-2.5}$), followed by a minor contribution from dust aerosols. The 1-year averaged ratio of $\text{NO}_3^-/\text{SO}_4^{2-}$ was 0.28 ± 0.12 and 0.44 ± 0.33 in fine and coarse mode, indicating a greater contribution of SO_4^{2-} , which shows the dominance of stationary sources over vehicular emissions. Taken together, this study will be supportive to establish the baseline of fine and coarse particulate matter for policymakers and researchers.

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