

The effect of meteorological conditions on aerosol size distribution in Istanbul

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Abstract Ambient aerosols were sampled by a high-volume cascade impactor in Istanbul, through May 2012 and November 2014. Seventy-eight size-segregated samples were gathered within the period at six different stages. The particles exhibited tri-modal distribution. The peak at $<0.49 \mu\text{m}$ was the most dominant among the others. The average mass median diameter was $1.3 \mu\text{m}$. The average total suspended particulate concentration was $75 \mu\text{g m}^{-3}$, and PM_{10} , PM_4 , $\text{PM}_{2.5}$, and PM_1 concentrations, derived from log-probability plots, were 62.5, 52.9, 46.9, and $34.2 \mu\text{g m}^{-3}$, respectively. Particle concentrations related to meteorological conditions through Pearson's correlation coefficient. The Pearson's correlation coefficient was poor in describing the association between coarse particles and meteorological conditions due to the increased urban effect, short-range transportation of marine aerosols, and long-range transportation. Particles >7.2 and $7.2\text{--}3 \mu\text{m}$ had a strong relation, indicating same sources. Increased relative humidity enriched $0.95\text{--}1.5\text{-}\mu\text{m}$ particle fraction in winter. Particles between 0.49 and $3 \mu\text{m}$ were inversely related to ambient temperature. Dilution effect of the wind was significant for $\text{PM}_{1.5}$. Wind acted as a source for larger particles by carrying them from other source regions. Multiple linear regression was applied to particulate matter fractions in order to model the concentrations of each fraction related to meteorological data. In the model, the particle fractions of $1.5\text{--}0.95$ and $0.95\text{--}0.49 \mu\text{m}$ exhibited the highest prediction performance.

Keywords Particle size distribution · Respirable fraction · Thoracic fraction · Pearson's correlation coefficient · MLR · PSCF

Introduction

Principal air pollutants can be classified as gasses and particulate matter (PM). Either of these pollutants adversely affects human health. However, PM has additional drawbacks such as the impact on the earth's radiation balance (Yin and Harrison 2008). That means PM plays a role in global climate change. This effect occurs through three mechanisms: (i) scattering and absorption of solar radiation; (ii) scattering, absorption, and emission of thermal radiation; and (iii) acting of particles as cloud condensation nuclei and ice condensation nuclei (Lohmann and Feichter 2005). Among the other pollutants, PM is related with the most important effects of air pollution (Maté et al. 2010). Several epidemiological studies revealed that PM is the cause of respiratory, cardiovascular, allergic, and lung cancer diseases (Bernstein et al. 2004; Künzli et al. 2000). Six to 14 times of more chronic bronchitis incidence was reported in Delhi than other cities in India, where PM air pollution is a serious problem (Perrino et al. 2011). PM is reported to lead an increase in mortality (Dockery et al. 1993; Pope et al. 2002). Some studies relate $\text{PM}_{2.5}$ exposure and increased mortality (Halonen et al. 2009; Jerrett et al. 2005; Yorifuji et al. 2005). An increase of 1.1% in hospital admissions was reported for $10 \mu\text{g m}^{-3}$ increase in $\text{PM}_{2.5}$ concentrations in China (Duan et al. 2016). Maté et al. (2010) expressed that continuous exposure to $\text{PM}_{2.5}$ increases the risk of respiratory and cardiovascular diseases and tumor growth within the respiratory tract. It was also stated that infants, children, and elderly people are more prone to the effects of air pollution. Apart from respiratory system effects,

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the autonomic nervous system is being affected from $PM_{2.5}$, and these particles cause cardiovascular diseases (Zanobetti et al. 2004). Additionally, Hoek et al. (2001) linked $PM_{2.5}$ with susceptibility to cardiac arrhythmias. These climatic and human health effects related to PM depend on their properties such as concentration, size, and their chemical composition (IPCC 2001; Kaufman et al. 2002). Therefore, monitoring such properties of PM is mandatory in order to understand their effects, sources, formation, removal, and transformation mechanisms (Duarte et al. 2008). In this respect, the most important data of PM is its size distribution which supplies data on the sources, the source processes, and the transformation processes during atmospheric transport (Salma et al. 2005).

Beyond of all, some definitions have been made for more precise evaluation of the particulate matter. Inhalable fraction is the total airborne particles, which are inhaled through the nose and mouth; thoracic fraction refers to inhaled particles penetrating beyond the larynx; the respirable fraction is the inhaled particles penetrating to the unciliated airways (Brown et al. 2013). Mass median diameters (MMDs) for thoracic fraction and respirable fraction are reported to be 10 and 4 μm , respectively (Hieu and Lee 2010).

The US Environment Protection Agency (US EPA) suggested PM_{10} sampling instead of total suspended particulate (TSP) in 1987; later on, in 1997, $PM_{2.5}$ sampling was proposed to evaluate fine fraction (Brown et al. 2013). US EPA, in a report, suggested using PM_1 as an indicator of fine particles (Hieu and Lee 2010). As the advantage of PM_1 over $PM_{2.5}$ is not yet clear enough, it is known from toxicological studies that ultrafine particles show more toxic behavior than coarser particles (Lingard et al. 2005). When particle size decreases, surface area and toxicity per unit mass increase. Particles, having diameter less than 1–2 μm , tend to settle in the alveolar region, and trace element adsorption efficiency is reported to be 60–80% in this region (Infante and Acosta 1991). Additionally, in a roadside microenvironment, PM_1 is suggested to be a better indicator than $PM_{2.5}$ by Lundgren et al. (1996) due to minimized interference from natural sources.

PM is a complex matrix, including several solid and liquid particles of various sizes from distinct sources (Perrino et al. 2011). Within this complexity, modal parameters should be known in order to predict and model the evolution of PM precisely (Agus et al. 2007). Modern cascade impactors are capable of separating fine and ultrafine particles. Size range spectrum can broadly be evaluated by processing the sampling data with advanced mathematical algorithms (Kenneth Wolfenbarger and Seinfeld 1990; Talukdar and Swihart 2003). Charron and Harrison (2003) associated meteorological data to size-segregated particles. It was stated that wind speed did not alter nucleation mode particle concentration; however, obvious reductions were reported for the remaining

size fractions. Ambient particulate matter usually fit log-normal distribution (Whitby 1978). This distribution property helps to reveal modal diameters, which is given by MMD, and the width of the modal distribution, which is given by the geometric standard deviation (σ_g). In that way, particle formation mechanisms can be revealed, source identification can be conducted, and further atmospheric evolution can be explored (Agus et al. 2007).

Although there are some studies relating the meteorological conditions with PM concentrations, the relation between PM and temperature, relative humidity, wind, and atmospheric circulation is still not clear enough (Pateraki et al. 2012). Therefore, size-distributed particle concentrations were used in this study in order to make a further achievement. The main objectives of this study are to (i) reveal the modality of ambient particulate matter in an urban atmosphere, (ii) relate meteorological parameters with six different particle size stages, and (iii) extract thoracic fraction, respirable fraction, and PM_1 data from size-segregated particle data.

Materials and methods

Sampling and instruments

Ambient particle samples were collected within Yildiz Technical University Davutpasa Campus in Istanbul on the rooftop of Civil Engineering Faculty. The faculty is situated at 83 m above sea level. The coordinates of the sampling point are 41° 01' 26" N latitude and 28° 53' 16" E longitude. The total area of the campus is 100 ha. Small-scale industrial facilities are present to the east and west of the campus area, which do not produce too much particle emissions. The remaining directions are covered with residential areas. Heating supplied natural gas combustion in the houses. A major highway is located to 2 km south of the sampling location. The load of the highway is 172,000 vehicles per day. The ambient particles were collected by a five-stage (with an additional backup stage) Staplex™ Model235 high-volume cascade impactor (HVCI). It was operated at 1.1 $\text{m}^3 \text{min}^{-1}$ flow rate. When this flow rate is sustained, the aerodynamic cutoff diameters of the stages are as follows: 7.2, 3.0, 1.5, 0.95, and 0.49 μm . The remaining particles are collected through backup filter, placed downstream of the impaction stages. Particles were collected on glass fiber filters. The samples were collected between May 2012 and November 2014. Seventy-eight groups of air samples were collected during this period, making a total number of 468 samples. The average operation time of the sampler was 30 h when averaged for each set over the entire sampling. The average airflow was approximately 2000 m^3 .

An AND GR-202 microbalance was used for gravimetric measurements. The filters were conditioned before and after

the sampling at $50 \pm 5\%$ relative humidity and 20 ± 2 °C temperature. The microbalance was calibrated with internal weights before each measurement.

During each sampling, a Davis Vantage Pro-2 weather station was being operated adjacent to the HVCI. The weather station continuously measured and recorded basic weather parameters. The measurements included wind speed, wind direction, temperature, humidity, pressure, precipitation amount, and solar radiation. Air Resources Laboratory (ARL) produces mixing height data for any location around the world. The mixing height data and the back trajectories obtained from the NCEP reanalysis and HYSPLIT model output, respectively.

Calculations

Multiple linear regression

The simplest way of relating the meteorological data to ambient pollution data is the use of multiple linear regression (MLR). MLR is a system executed to resolve relation between the relationship of a set of independent variables to a single dependent variable (Aiken et al. 2003). This system can also be used in relating the air pollutant data to meteorological conditions, and this regression has been applied in many studies so far (Akyüz and Çabuk 2009; Cetin et al. 2007; Kuzu et al. 2014; Tai et al. 2010).

$$\ln\text{PM} = m_1(u) + m_2\cos(\text{WD}) + m_3(T) + m_4(\text{MH}) + c \quad (1)$$

where PM is the concentration of particulate matter ($\mu\text{g m}^{-3}$), u is the average wind speed (m s^{-1}), WD is the prevailing wind direction (in degrees), T is the atmospheric temperature (°K), MH is the mixing height (m), c is a constant term, and m_1 , m_2 , m_3 , and m_4 are regression parameters. Positive regression parameters indicate that particle concentration increases with the increasing value of the meteorological events. If the parameters are negative, then it means that the concentration is inversely related to these variables. Microsoft Excel Data Analysis add-in was used to execute MLR calculations.

Back trajectories

Apart from MLR, back trajectories show the route of the transported air mass. Potential source contribution function (PSCF) is a useful methodology to incorporate pollutant data with the back trajectories. In this methodology, probability field is generated that match the excessive concentrations at the receptor point with their probable source locations. In its theory, it is assumed that the end-point of each trajectory in a grid cell carries the pollutant from that location. Based on a criterion, trajectories are separated as “polluted” and “unpolluted.” The quotient of the number of polluted

trajectories to the number of total trajectories from cell i,j yields the PSCF score of i,j th cell (Eq. 2).

$$\text{PSCF}_{i,j} = \frac{\sum M_{i,j}}{\sum N_{i,j}} \quad (2)$$

In Eq. (2), $N_{i,j}$ refers to the total number of trajectories emerging from the i,j th cell, whereas $M_{i,j}$ is the number of polluted trajectories from i,j th cell. Small $N_{i,j}$ values lead to elevated PSCF results with high uncertainties. This occasion is usually encountered at more distant sources from the sampling location, because scattered nature of the trajectories causes sparse distribution at more distant grids. PSCF results are multiplied by an arbitrary weighting function $W_{i,j}$ in order to reduce the uncertainties of small $N_{i,j}$ values (Polissar et al. 1999). The selected $W_{i,j}$ values of this study are given in Eq. (3).

$$W_{i,j} = \begin{cases} 1.0 & n_{i,j} > 120 \\ 0.9 & 120 > n_{i,j} \geq 80 \\ 0.7 & 80 > n_{i,j} \geq 40 \\ 0.4 & 40 > n_{i,j} \geq 20 \\ 0.2 & 20 > n_{i,j} \end{cases} \quad (3)$$

Two different “polluted trajectory” selection criteria were suggested (Pekney et al. 2006). They used 90th percentile for the pollutant having low concentrations in most of the data set with high peak concentrations considerably different from background concentrations, while they selected the 75th percentile for the pollutant having more days with peak concentrations that are not much different from background concentration. Concentrations, which fall beyond the upper 25th quartile, were considered as polluted in this study. The grid resolution was $1^\circ \times 1^\circ$. Run time of the back trajectories was 24 h. TrajStat v.1.2.2.6 was used for the calculation of PSCF scores at each grid (Wang et al. 2009). The plots were generated through Surfer software.

Results and discussion

Particle size distribution data

The mean normalized size distributions of PM for different seasons, gathered through the sampling, are given in Fig. 1.

Particles exhibited tri-modal distribution at each of the seasons. The peaks were observed at particle sizes of <0.49 , 0.95 – 1.5 , and 3 – 7.2 μm . The finest particles were the most dominant in terms of mass for all of the seasons. Coarse mode particles were the second dominant fraction except for the winter season. The particles between 0.95 and 1.5 μm dominated the coarser particles in winter. In a different study, the particles between 0.95 and 1.5 μm were also enriched in the winter season in Thessaloniki (Chrysikou and Samara 2009).

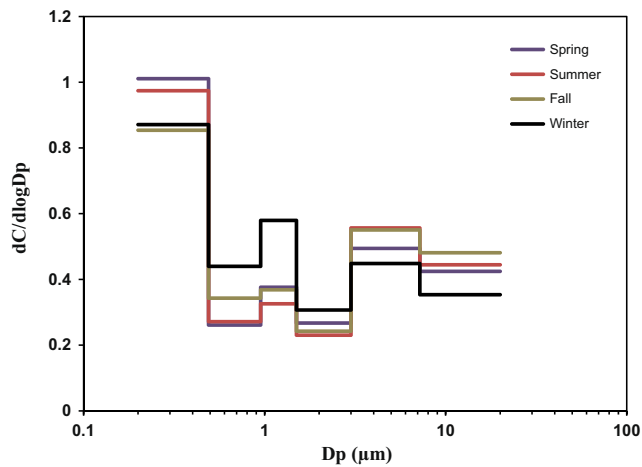


Fig. 1 Seasonality of particle size distribution

Increased anthropogenic activities could be the reason for this shift in the winter; air quality in Istanbul is reduced due to domestic heating and diesel vehicle emissions in the winter (Kuzu 2016b). The primary emitted pollutants accumulate to form larger particles. Increased relative humidity in the winter may be the driving factor for the particle growth in coarser mode. In several previous studies, mostly bi-modal particle size distribution was reported (Chrysikou and Samara 2009; Hien et al. 2007; Kuzu et al. 2013). Şahin et al. (2012) sampled ambient aerosols at five different locations in Istanbul by a low-volume cascade impactor. The particles exhibited bi-modal or tri-modal distributions. Although weather conditions do not alter too much within the city, the local sources may be effective in changing the modality of the particles.

Log-probability plots were generated for each season. These plots provide beneficial information about the particles. It is possible to extract PM concentration of different aerodynamic particle diameters. The plots are given in Fig. 2. MMDs were 1.1, 1.2, 1.4, and 1.1 μm for spring, summer, fall, and winter, respectively. In many of the previous studies, ambient particle MMDs were close to 1 μm (Bi et al. 2005; Kuzu et al. 2013; Lin et al. 2008; Şahin et al. 2012). The average MMD and PM concentrations of different sizes derived from log-probability plots are given in Table 1 along with their statistical analysis.

The average TSP concentration was $75 \mu\text{g m}^{-3}$, ranging between 38.8 and $182.2 \mu\text{g m}^{-3}$. TSP samples were taken at three different locations in Istanbul in a long-term study by high-volume sampler (Hanedar et al. 2011). The average concentrations observed in the urban sampling sites were 101 and $152 \mu\text{g m}^{-3}$, whereas the average concentration in a rural area was $50 \mu\text{g m}^{-3}$. Şahin et al. (2012) collected ambient particles at close places to the previous study by a low-volume cascade impactor. However, concentrations reported of that study was one third of Hanedar et al. (2011) at each of the sampling locations. Particle losses between the stages could have led this difference. TSP concentrations of this study are between

rural and urban concentrations reported by Hanedar et al. (2011). High-volume cascade impactors are more accurate than low-volume cascade impactors in TSP sampling (Şahin et al. 2012), because less loss occurs due to wider slot space of each sampling plate.

The average PM_{10} , PM_4 , $\text{PM}_{2.5}$, and PM_1 concentrations, derived from log-probability plots, were 62.5, 52.9, 46.9, and $34.2 \mu\text{g m}^{-3}$, respectively. EU limit for PM_{10} concentration is $50 \mu\text{g m}^{-3}$ (European 2008). Both thoracic and respirable fractions of the particles were above the limit value. PM_1 was sampled in Italy, and the average concentration was reported to be $8 \mu\text{g m}^{-3}$ (Trippetta et al. 2016). PM_1 measured in Istanbul was substantially higher. The ratios between PM_1 , $\text{PM}_{2.5}$, and PM_{10} give an idea of the formation sources of the particles. PM_1 is mostly from fresh combustion sources. $\text{PM}_{1-2.5}$ shows anthropogenic contribution, whereas $\text{PM}_{2.5-10}$ occurs from natural sources. The $\text{PM}_{2.5}/\text{PM}_{10}$ ratio usually ranges between 0.4 and 0.8 (Pateraki et al. 2012). In this study, $\text{PM}_{2.5}/\text{PM}_{10}$ ratio was 0.75, consistent with previous studies (Gonçalves et al. 2017). The average $\text{PM}_1/\text{PM}_{2.5}$ ratio was 0.73 in this study. Substantially lower $\text{PM}_1/\text{PM}_{2.5}$ ratios were previously reported in Istanbul at five different locations, which ranged between 0.45 and 0.57 (Şahin et al. 2012).

The relation between different PM sizes and meteorological conditions

The relationships between different particle sizes and meteorological conditions were investigated through Pearson's correlation analysis. Meteorological conditions during the sampling campaign are given in Table 2. The correlation matrix including correlation coefficients is given in Table 3. Positive numbers show that two variables are directly related, whereas negative numbers show an inversely correlation. Significance levels of the correlation coefficients were investigated, and they were grouped under three distinct classes. The classes show reasonably high ($p < 0.05$), high ($p < 0.01$), and very high ($p < 0.001$) significance levels.

Inter-relation of the particles having aerodynamic diameter larger than 1.5 μm was positive and significant. Especially, the value of r was close to 1 for particles having diameter larger than 3 μm . The particles larger than 2.5 μm are regarded as coarse mode particles. They are generated from mechanical processes. This shows that coarse particles in the study area are produced from the same source.

The most significant parameter affecting the particle concentrations was mixing height. Each particle size was correlated inversely and strongly with the mixing height. When the mixing height is deeper, the particle mass is diluted in a higher air volume. Thus, particle concentrations reduce. In the contrary case, the particles are concentrated in a lower air volume.

Particles were least associated with relative humidity. The particles $>3 \mu\text{m}$ had tendency to decrease with the increasing

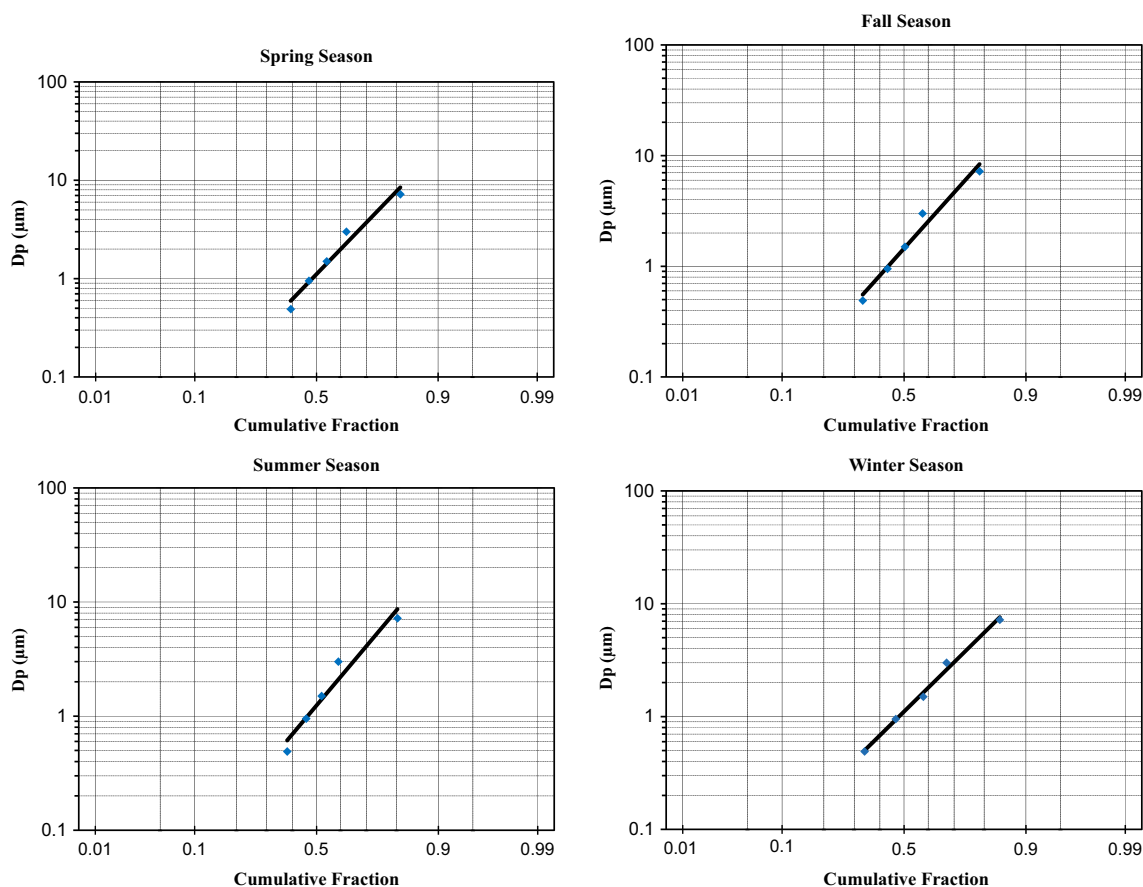


Fig. 2 Log-probability plots of each season

humidity. This is an expected case for coarse particles because they are strongly related with dust resuspension (Cheung et al. 2011). However, the statistical significance of this study was not enough to correlate humidity and the decrease in coarse PM concentrations. Additionally, during humid conditions, aerosols absorb water vapor. This situation leads a change in aerosol volume and consequently a shift of fine/coarse mode particle ratio (Vasilakos et al. 2007). The strongest correlation of humidity was with the particles between 0.95 and 1.5 µm.

Table 1 Statistics of particle size distribution and extracted data

	Average	Standard deviation	Min	25th quartile	75th quartile	Max
MMD ^a	1.3	0.6	0.4	1.0	1.5	3.5
TSP concentration ^b	75.0	25.0	38.8	55.1	91.6	182.2
PM10 ^b	62.5	21.2	30.8	47.1	75.1	152.3
PM4 ^b	52.9	17.7	26.0	40.3	63.4	126.6
PM2.5 ^b	46.9	15.5	23.2	35.8	56.2	110.5
PM1 ^b	34.2	11.3	15.7	25.9	40.3	76.8

^a Unit is µm

^b Unit is µg m⁻³

Particles increased with the increasing humidity. The shifted dominance of this particle fraction in winter can be attributed to the relative humidity in this case.

Particles larger than 3 µm had positive relation with the ambient temperature. However, the relation was not significant. There was a negative relation with smaller particles. The significance was not clear only for particles smaller than 0.49 µm. Similarly, Akyüz and Çabuk (2009) reported an inverse relation between temperature and PM_{2.5} in Zonguldak, Turkey. However, the Pearson’s correlation coefficient was positive for PM_{2.5–10} in their study. Contrary to this, Vasilakos et al. (2007) stated that fine particles were influenced by photochemical processes during hotter temperatures. Temperature increase favors the formation of secondary organic aerosols (Pateraki et al. 2012). Sulfate and nitrate are principal ions in the formation of secondary aerosols. Higher nitrate concentrations were observed during night sampling in Istanbul (Kuzu et al. 2013). Seasonal differences in primary emissions and their transformation in the atmosphere mask the effect of temperature on aerosols in Istanbul atmosphere.

Aerosol concentrations decreased during higher wind speeds at each particle size. Dilution effect was more obvious for smaller particles because the correlation was significant for

Table 2 Meteorological conditions during the sampling campaign

Season	Wind speed (m/s)	Temperature (°C)	Mixing height (m)	Relative humidity (%)
Spring	2.9 ± 0.7	14.9 ± 4.7	506 ± 250	73.9 ± 9.2
Summer	3.1 ± 1.1	25.5 ± 3.4	639 ± 273	68.9 ± 8.0
Fall	2.7 ± 1.2	20.6 ± 4.6	646 ± 230	71.6 ± 7.5
Winter	3.0 ± 2.3	9.2 ± 3.4	427 ± 220	81.0 ± 22.4

PM_{1.5}. The less significance level for coarser particles may be due to transportation of coarse particles to the sampling location at higher wind speeds. Primary sea salt aerosols and long-range transported dusts can be the main components of the coarse particles. Kuzu et al. (2013) declared that mechanical disruption of the sea surface was dominant in producing sea salt aerosols in Istanbul. The disruption occurs through the wind. Kuzu et al. (2013) also stated that chlorine and fluoride ions had higher concentrations at coarse particle modes in Istanbul. This proves the contribution of marine aerosols at coarse sizes. In another study, it was concluded that marine aerosol contribution to the ambient aerosol had the highest share in Istanbul (Uygur et al. 2014). Istanbul has shorelines at both south and north of the city. Therefore, regardless of the dominant wind directions (NE and SW), the city is exposed to marine aerosols. Long-range transportation was studied during a dust transportation event from Saharan Desert in Istanbul (Kuzu 2016a). Highest enrichment was observed between 3 and 7.2-µm particle sizes.

As a general evaluation, it was observed that correlation between >PM₃ and meteorological conditions was not significant. The active sources previously stated as marine aerosol contribution and long-range transported dusts as well as resuspension of dust from urban activities are thought to reduce the correlation significance between meteorological conditions and coarse particle concentrations.

The multiple regression between different PM sizes and meteorological conditions

MLR was applied to the dataset to determine the combined effect of meteorological conditions on particle concentrations. At first, individual regression was applied to each particle size and each meteorological parameter. The explanatory variables, used in multiple regression analysis, were selected based on their significance. Humidity had the weakest relation, and therefore, it was not included to MLR. Two thirds of the data was used in order to train the model. The remaining data were used for evaluating the model results. The data used in the assessment of the model covered a whole year. These dates were between December 2013 and November 2014. The resulting equations are given below for each particle fraction.

$$\ln PM = -0.038(u) - 0.095 \cos(WD) + 0.017(T) + 0.001(MH) + 2.70 \quad (\text{for } > 7.2 \mu\text{m})$$

$$\ln PM = 0.009(u) - 0.050 \cos(WD) + 0.021(T) - 0.001(MH) + 2.69 \quad (\text{for } 7.2\text{--}3 \mu\text{m})$$

$$\ln PM = -0.006(u) - 0.101 \cos(WD) - 0.004(T) - 0.001(MH) + 2.19 \quad (\text{for } 3\text{--}1.5 \mu\text{m})$$

Table 3 Correlation matrix of particle sizes and meteorological factors

	>7.2	7.2–3	3–1.5	1.5–0.95	0.95–0.49	<0.49
>7.2	1					
7.2–3	0.807***	1				
3–1.5	0.481***	0.448***	1			
1.5–0.95	0.330**	0.282*	0.531***	1		
0.95–0.49	0.200	0.145	0.377***	0.620**	1	
<0.49	0.358**	0.266*	0.486**	0.498**	0.195	1
<i>u</i> (m/s)	–0.187	–0.043	–0.229	–0.335**	–0.343**	–0.256*
<i>T</i>	0.190	0.189	–0.283*	–0.438**	–0.282*	–0.194
<i>H</i> (m)	–0.347**	–0.371**	–0.482***	–0.537**	–0.334**	–0.422***
Hum	–0.111	–0.138	0.059	0.284*	0.246*	–0.280*

*Correlation is significant at 0.05 level

**Correlation is significant at 0.01 level

***Correlation is significant at 0.001 level

Table 4 Statistical indicators of each simulated/observed particle fraction

Descriptive statistics	>7.2 μm	7.2–3 μm	3–1.5 μm	1.5–0.95 μm	0.95–0.49 μm	<0.49 μm
R^2	0.14	0.15	0.49	0.54	0.57	0.23
IA	0.56	0.59	0.77	0.78	0.75	0.63
MBE	-1.58	-1.46	-0.44	-0.02	0.43	-0.27
RMSE	3.81	3.98	1.35	2.84	3.18	7.65
MAE	2.91	3.07	1.11	2.04	2.32	5.67

$$\begin{aligned} \ln\text{PM} &= -0.049(u) \\ &+ 0.056 \cos(\text{WD}) - 0.023(T) - 0.001(\text{MH}) \\ &+ 2.89 \quad (\text{for } 1.5\text{--}0.95 \mu\text{m}) \end{aligned}$$

$$\begin{aligned} \ln\text{PM} &= -0.071(u) \\ &+ 0.134 \cos(\text{WD}) - 0.013(T) - 0.001(\text{MH}) \\ &+ 2.80 \quad (\text{for } 0.95\text{--}0.49 \mu\text{m}) \end{aligned}$$

$$\begin{aligned} \ln\text{PM} &= -0.041(u) - 0.158 \cos(\text{WD}) - 0.004(T) - 0.001(\text{MH}) \\ &+ 3.65 \quad (\text{for } < 0.49 \mu\text{m}) \end{aligned}$$

The above equations formulate the meteorological conditions to give particle concentrations. The strength of the equation is tested with some statistical indicators. Determination coefficient (R^2), index of agreement (IA), mean bias error (MBE), root mean square error (RMSE), and mean absolute error (MAE) are used to determine the goodness of relation. The calculated indicators for each particle size are given in Table 4.

The models for the particle size fractions between 3 and 1.5, 1.5–0.95, and 0.95–0.49 μm produced substantially better estimates relative to remaining particle size fractions. Determination coefficients suggest that these three models approximately predicted half of the data accurately. The fractions of 1.5–0.95 and 0.95–0.49 μm had the best estimates among the three models. Simulated and observed data were plotted for these two particle size fractions and shown in Fig. 3.

Apart from statistical indicators, a good visual fit between the observed and predicted values is present in Fig. 3. Severe deviations were observed in 2 of the 26 samplings. If there were no underestimations, the indicators would have been much better. The meteorological conditions checked for these 2 days. It was observed that there was no single dominant wind direction. Wind blew from different directions at equal shares in those days. This could lead to transportation of particles from different sources. Therefore, the observed concentrations deviated from the usual trend. Additional outcome of the model results are that the relation of the particles >3 and $<0.49 \mu\text{m}$ with the combined effect of the meteorological parameters is weak.

Trajectory analyses

In order to facilitate a relation between transported air masses to the sampling point and observed excessive concentrations, trajectories were examined through PSCF. Previously, long-range PM_{10} sources was investigated through PSCF in Istanbul (Karaca et al. 2009). Southern Mediterranean region had high PSCF scores at long range. Also, close areas at the south of Istanbul had high PSCF scores. In this study, we applied PSCF at shorter run time in order to evaluate regional transportation. This methodology was applied for each particle sizes. The results are shown in Fig. 4.

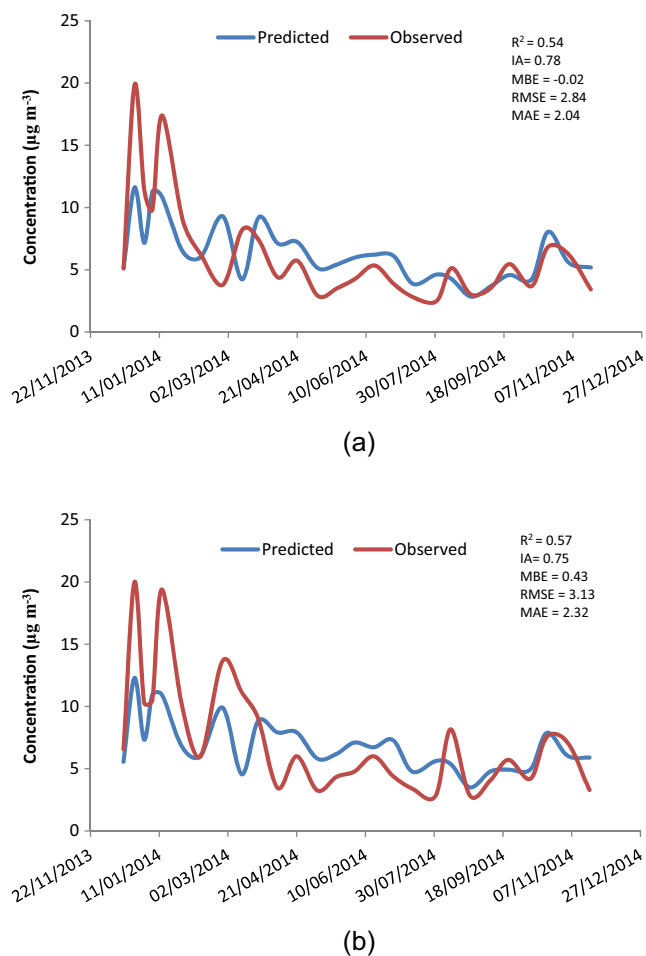


Fig. 3 Observed/predicted plot of **a** 1.5–0.95 and **b** 0.95–0.49 μm particle fraction

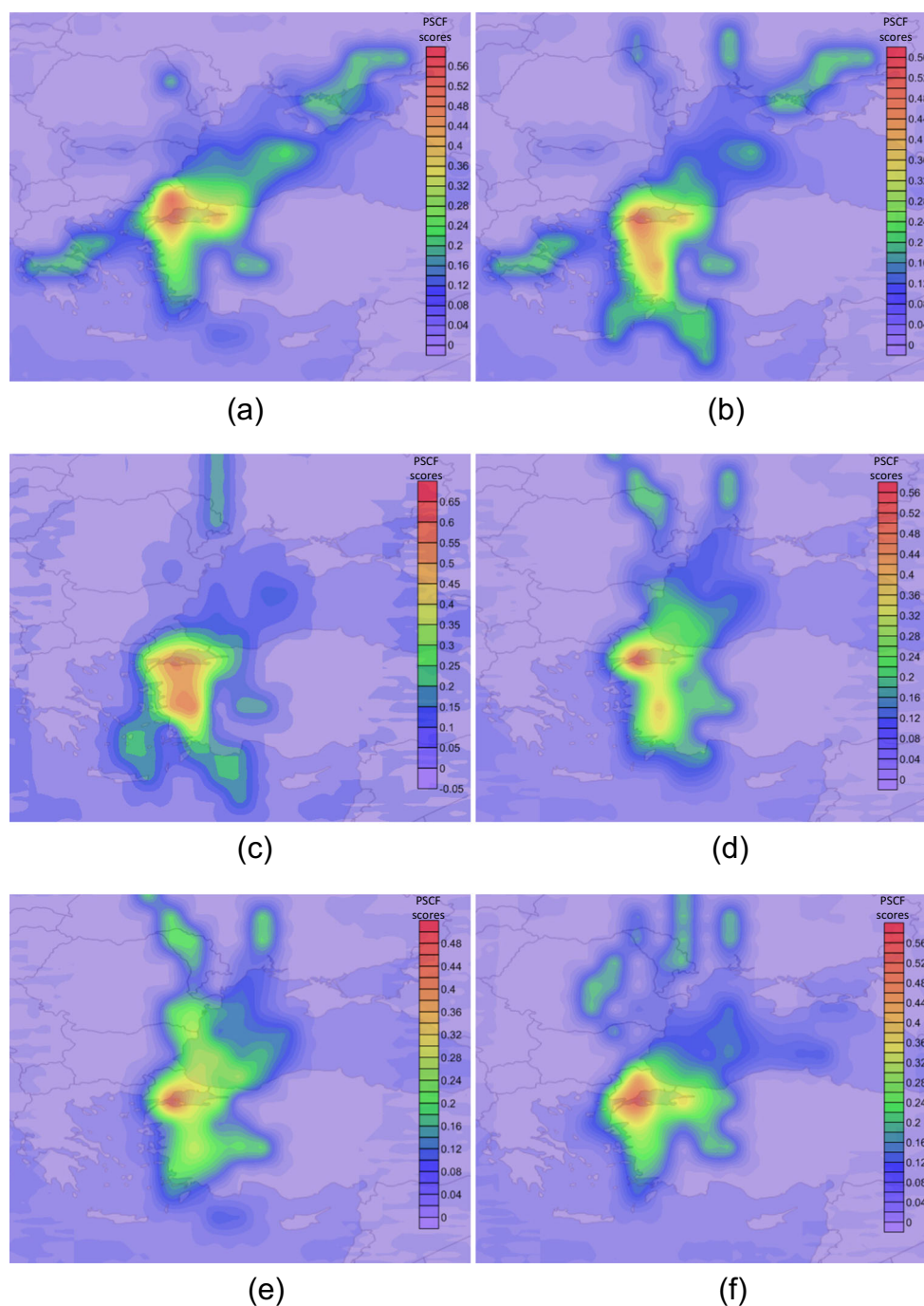


Fig. 4 PSCF scores for **a** >7.2 μm , **b** $7.2\text{--}3$ μm , **c** $3\text{--}1.5$ μm , **d** $1.5\text{--}0.95$ μm , **e** $0.95\text{--}0.49$ μm , and **f** <0.49 μm

For each of the particle sizes, trajectories over Marmara Sea had the highest PSCF scores. This also confirms the previously stated marine aerosol contribution. Apart from marine aerosol contribution, the particle sizes between $7.2\text{--}3$ and $3\text{--}1.5$ μm seemed to have crustal origin from the south of Istanbul. The particles >7.2 μm also had transportation over crustal area. But this fraction a bit differed from the sizes between 7.2 and 1.5 μm . Due to their larger size, particles of >7.2 μm are settled during

transportation. This fraction was transported from close areas over Trachea region. For finer particle sizes, the highest PSCF scores were distributed in a narrower area. Fine particles in urban area are formed usually through combustion sources and gas to particle formation mechanism. For that reason, sources of fine particles had to be close to sampling area. According to results in Fig. 3, PSCF did not performed reasonable results for particles <1.5 μm .

Conclusions

Particle size distribution is crucial in determining the sources and fate of the particles. Association between particles and meteorological parameters has been reported in several studies so far, but the relation is still not clear enough. Especially, fewer studies are present for size-segregated particles. Additionally, there is no universal consensus for this topic, because each study is unique due to its own source and meteorological parameter characteristics. In this scope, samples were collected during a long sampling period in Istanbul which is a complex urban area. The city is being affected by anthropogenic sources and natural sources. Anthropogenic sources show variations between heating season and non-heating season. Therefore, dominance of the particle fraction between 0.95 and 1.5 μm was enriched in winter with the help of higher relative humidity level. Coarse particles were not correlated with the meteorological parameters significantly, because crustal dust and marine aerosol contribution was more effective than meteorological conditions on coarse particle fraction. NE and SW are the prevailing wind directions in Istanbul. Both directions blow over the sea surface, bringing marine aerosols to the sampling point. Particles between 0.49 and 3 μm were inversely related to ambient temperature, showing that they were increased during colder periods. Dilution effect of the wind was significant for $\text{PM}_{1.5}$, which is generated from fresh sources. Multiple regression was applied in order to model each fraction with meteorological parameters. The best performance was achieved for 1.5–0.95 and 0.95–0.49- μm particle fractions. These fractions had appreciable relation with meteorological parameters. PSCF generated reasonable results for the particles $>1.5 \mu\text{m}$; however, this methodology was poor in estimating the source regions of $\text{PM}_{1.5}$.

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