Environ Sci Pollut Res (2016) 23:11085–11099
DOI 10.1007/s11356-016-6323-z

RESEARCH ARTICLE

Mass size distribution and source identification of particulate matter metal components at four urban sites and a background site of Istanbul

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Received: 17 November 2015 / Accepted: 16 February 2016 / Published online: 24 February 2016

Abstract In this study, the size distribution characteristics and metal contents of particulate matter (PM) have been determined. In this scope, PM sampling has been done at five stations in Istanbul. PM filter samples were collected for eight different sizes using the Anderson cascade impactor. PM filters were decomposed and analyzed for 20 metals. The highest median concentration for Fe, Ca, K, and Mg, known as soil metals, were observed as follows: Fe and Ca were observed at Goztepe station (1.20 and 8.28 μg/m^3), K was observed at Kilyos station (0.33 μg/m^3), and Mg was observed at Avcilar station (0.37 μg/m^3). The highest median concentrations for Zn, Cu, Pb, Ni, Cr, V, As, Se, Co, and Cd, known as anthropogenic metals, were observed at Avcilar, Goztepe, and Besiktas stations. Although the lowest metal concentrations was determined at Kilyos stations that was selected as the urban background. The enrichment factors (EFs) of most metals in the fine PM is higher than those in the coarse mode. According to the factor analyses, the most important emission source was observed to be industrial facilities at Avcilar; traffic at Besiktas; traffic and domestic heating at Goztepe; and domestic heating, sea salt aerosols, and ship traffic (in the Bosphorus Channel of Istanbul) at Rasathane.

Keywords Particulate matter · Size distribution · Metals · Enrichment factor · Sources · Istanbul

Responsible editor: Constantini Samara

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Introduction

Istanbul is an important city in Turkey owing to the extent of industrialization, steady rise in population, and urbanization. It also plays a role as a center of economic activity as a result of its geographical location between Asia and Europe. However, these factors contribute to worsening air quality. The characterization of the air pollution brought about by this regional development and the determination of the precautionary measures constitute an important environmental issue. Many cities in Turkey move toward the actualization of the clean urban air action plan. A considerable reduction in sulfur dioxide (SO_2) contamination was achieved in the Marmara region in Turkey, where Istanbul is located, during the 20-year-long transition process of heating systems that employ natural gas, replacing coal as a source of energy. In contrast, the concentration of particulate matter (PM) in the atmosphere cannot be reduced. It is difficult to determine the quality of air in a metropolitan city such as Istanbul, populated by 13 million people residing in an area of 5712 km^2. Regional atmospheric conditions and the geographical landscape are two of the most important parameters affecting air quality of a city. Istanbul resides on seven strategic hills; a different meteorological weather event can be observed in different districts within the city, making that determination even more difficult.

The number of publications on the particle size distribution and metallic content of PM has been increasing in the last 5 years. A number of studies were conducted on PM_{10} (particulate matter less than 10 μm in diameter) and $PM_{2.5}$ (particulate matter less than 2.5 μm in diameter). The physical and chemical characteristics of PM measured in 31 different locations in Europe were investigated in studies by Dingenen et al. (2004) and Putaud et al. (2004). These authors have reported that metal salts constituted 13–22 % of PM_{10}, an undetermined fraction constituted 15–28 %, whereas the

remainder was composed of ionic materials. A similar study was conducted in 19 different cities in Europe by Künzli et al. (2006). The Pb and As values reported in that study were as follows: Pb, 13.8 ng/m^3 and As, 4.1 ng/m^3 in Erfurt, Germany; Pb, 15.4 ng/m^3 and As, 3.7 ng/m^3 in Paris, France; Pb, 59 ng/m^3 and As, 14 ng/m^3 in Barcelona, Spain; and Pb, 62.2 ng/m^3 and As, 14.7 ng/m^3 in Turin, Italy.

Scientists have initiated studies identifying the concentrations of different toxic elements in different fractions of the breathable particulate dust. The particle size distribution of the PM collected in Germany ranged from 0.05 to 10 μm; the PM having a size below 1.2 μm was determined to be 2- to 7-fold over-represented for Cr, Ni, Pb, Cu, Zn, and Mn (Brüggemann et al. 2009). Similar observations were reported in studies conducted in Greece and in the United Kingdom (Karanasiou et al. 2007; Taiwo et al. 2014; Voutsa et al. 2015). PM was sampled in six different particle sizes in a study conducted in Spain: 74.8 % of As, 69 % of Pb, 61.3 % of Cd, 57.5 % of Ni, and 40.4 % of Hg were measured in the fraction of fine PM (Alvarez et al. 2004).

A number of studies have been conducted on the identification of metal concentrations in PM_{10} and $PM_{2.5}$ in Turkey (Dündar and Deryaoğlu 2005; Yatkın and Bayram 2007; Gaga and Arı 2011; Onat and Şahin 2012; Onat et al. 2013). However, only a limited number of those studies were conducted in and around Istanbul, which is the most highly populated city in Turkey and also an important region of industrial activity. Karaca (2008) investigated the concentration of metals in atmospheric $PM_{2.5}$ and $PM_{2.5–10}$ aerosols in the Buyukcekmece region in Istanbul. Onat and Sahin (2012) determined the particle size distribution of PM and its metallic content at single locations on the European (in Yenibosna) and on the Asian (in Goztepe) sides of Istanbul. Approximately 50–90 % of all metals were determined to be within the fraction having <3.3 μm PM size. Another study conducted in Istanbul reported the content of PM_{10} samples at a single location and identified the source distribution (Koçak et al. 2011). That study reported approximately 80 % of PM_{10} in the Istanbul atmosphere to be of anthropogenic sources.

In the present study, five different locations were selected in Istanbul to represent the overall air quality, taking the source characteristics and geographical locations into consideration. The seasonal variation in the concentration of 20 different elements in 8 different PM size fractions was investigated for 2 years.

Material and method

Area of study and sample collection

Istanbul, located on the extremities of Asia and Europe, is the most populous city in Turkey, with a population of approximately 13 million. The city has 11,143 industrial establishments and 2,500,000 registered motor vehicles. The sampling locations and size distribution analyses of total suspended particulate matter (TSP) have been described in detail in our previous studies (Şahin et al. 2012a; Onat and Şahin 2014).

TSP was sampled at five stations (Fig. 1). The Avcilar urban station (40° 59′ 21.45″ N, 28° 43′ 19.64″ E) has high levels of vehicle, industrial, and residential emissions. Residential emissions are caused by burning household coal, natural gas, and fuel oil. The Besiktas urban sampling station (41° 3′ 6.59″ N, 29° 0′ 35.82″ E) is located in a residential and commercial area, where the main pollution sources are residents' activities and vehicular emissions. The Rasathane urban sampling station (41° 3′ 48.54″ N, 29° 3′ 52.26″ E) is located in a residential area. The Goztepe urban sampling station (40° 59′ 47.36″ N, 29° 3′ 58.58″ E), which is near the D100 highway, has vehicle and residential emissions. The fifth sampling station is the Kilyos rural sampling station (41° 14′ 36.60″ N, 29° 0′ 48.60″ E), occupied by very few residents.

In this study, metals were collected using Andersen cascade impactor with an airflow rate of 90 lpm. Quartz fiber filters (QF) with 81-mm diameter were used for the gravimetric and metal analysis. The manufacturer for the quartz filter was Whatman and the supplier company is Thermo Scientific. All laboratory ware was thoroughly cleaned. Both the field blank filter and the laboratory blank filter were used for each campaign-sampling period. Corrections were made according to the values of the field and lab blank filters during analysis of each campaign filter. The Anderson consists of eight stages (labeled 0 to 7) and an additional stage F (backup filter), with size selection parameter divisions in terms of the particle aerodynamic diameter (dp) as follows: >8, 8–6.5, 6.5–5.2, 5.2–3.5, 3.5–2.6, 2.6–1.7, 1.7–1, 1–0.43, and <0.43 μm. The TSP concentrations of the nine PM fractions were determined by gravimetric analysis with an electronic balance that could detect levels as low as 0.01 mg. All blanks and the sample filters were kept in a conditioned special room for 48 h at a relative humidity of 50 ± 5 % and a room temperature of 20 ± 1 °C prior to their gravimetric measurements, both before and after sampling. After weighing, the samples were stored in the refrigerator at approximately 4 °C prior to extraction to prevent the loss of the volatile metals. The sampling procedure and gravimetric analysis of the TSP have been described in detail in our previous studies (Şahin et al. 2012a, b; Onat et al. 2012; Onat and Şahin 2014).

Sampling was carried out from July 2008 to August 2010. A total of 16 TSP samples were collected at each station. The TSP sampling was performed a total of 80 times. The duration of sampling in a single station required approximately 7 ± 2 days. The filters within the impactor were transferred to clean Petri dishes with Teflon-coated tweezers in a closed

Fig. 1 Study area and sampling stations

environment and were then brought into the laboratory. Following removal of the filters, the impactor was cleaned. Clean filters were then installed before the system was taken to the next station.

Extraction and analysis of metals

The dust filters were decomposed in three different steps using microwave equipment (CEM MARSXpress microwave digestion system, USA). In the first step, 6 ml of HNO_3 and 2 ml of H_2O_2 were added to the dust filters and were allowed to decompose for 40 min at 180 °C. In the second step, 2 ml HCl was added to the extract; the decomposition time was 35 min at 175 °C. In the final step, 2 ml HF was added to the extract; the decomposition time was 30 min at 160 °C. The most effective results were observed with this three-step decomposition process. At the end of the decomposition, the filters were completely dissolved. A rotating turntable (3–6 rpm) capable of holding ten closed digestion vessels (PTFE) ensured even sample heating. The samples were digested and cooled as per the program setting, after which the sample solution was directly diluted to 30 ml at 25 °C with deionized distilled water and stored in the refrigerator at approximately −4 °C prior to analysis.

To measure the metal concentrations in the solution, graphite atomic absorption spectrophotometry (PerkinElmer GF-AAS 600) and flame atomic absorption spectrophotometry (PerkinElmer F-AAS 400) were employed. The GF-AAS unit was operated with the Zeeman background correction on, and the peak area was integrated for all signals. To evaluate the accuracy of the analytical methods and to establish the optimal digestion conditions, the NIST Urban Particulate Matter (SRM 1648) and standard solution (Merck) were used as references. One milligram of SRM 1648 was added to a blank filter and digested using the method described previously to examine the capabilities of the analytical methods and to perform the recovery procedure. We determined between 85 and 105 % recovery of all metals was achieved using SRM 1648.

The accuracy of the measurement was accepted when the repeatability of the measurement for each sample was less than 2 %. Five standard solutions were prepared for the calibration curve and the $R^2 > 0.99$ condition was provided. All readings were repeated three times. We collected one filter from each measurement campaign to determine the background of pollution. For every digestion, these filters have been used as a blank and the metal contents analyzed.

Enrichment factor analysis

The enrichment factors (EFs) of the metallic elements were utilized to identify the origin of the elements in the atmosphere. Many studies have reported that an EF greater than 10 would be an indicator of anthropogenic sources for the existence of these elements, whereas an EF less than 10 would be an indicator of the dominance of natural sources (Cyrys et al. 2003; Slazekova et al. 2007; Ny and Lee 2010; Wang et al. 2006). Fe was selected as the reference element for the crust, as indicated in previous studies (Ny and Lee 2011; Wang et al.

2006; Samara and Voutsa 2005; Manoli et al. 2002). There is no widely accepted rule for the choice of the reference element; however, Fe, Si, and Al are usually used for this purpose (Wang et al. 2016; Duan et al. 2012). Some studies reported the abundance of Fe element in the upper continental crust (Taylor and McLennan 1985; Duan et al. 2012). Additionally, we could not measure Al and Si in this study.

The enrichment factors were determined with respect to a reference sample in previous studies. However, similar to the variations in the concentration of particulate matter in the atmosphere, regional differences also exist in soil samples. Coşkun et al. (2006) collected soil samples from 73 locations in the European portion of Turkey and identified 35 elements. The results obtained indicated that the soil elements in Turkey differed from the world average. All data available in the literature on the elemental composition of soil and road dust in Istanbul were evaluated, along with the results of the present study; the results were in accordance with those of previously conducted studies. The metal concentrations determined in the present study are provided in Table 1. The EF value, also provided in Table 1, was calculated to account for the metals in the soil composition of Istanbul, using Fe as the reference element.

Results and discussion

Element concentrations

The size distribution of TSP at the five sites has been reported in our previous study (Şahin et al. 2012a). The elemental concentrations found in the eight fractions at the five sites, along with statistical analyses, are shown in Table 2. The highest median values for the concentration of metals of soil origin—Fe and Ca (measured as 1.20 and 8.28 μg/m^3, respectively)—were determined in the Goztepe, of the K (measured as 0.33 μg/m^3) in the Kilyos, and of Mg (measured as 0.37 μg/m^3) in the Avcilar sampling locations where PM was collected. The concentration of these metals of soil origin were observed to be 1- to 2.6-fold higher in Besiktas, Avcilar, and Goztepe than in Kilyos, which was selected as the background location in our study; however, a considerable difference could not be observed for the samples collected from Rasathane.

Similar studies conducted in other cities in Turkey reported the concentration of these metals to be higher in the cities under investigation than in other European cities, especially in coarse dust (Yatin et al. 2000; Cyrys et al. 2003; Samara et al. 2003; Yatkın and Bayram 2007; Karaca 2008). Several studies attributed this observation to differences in the

Table 1 The elements contents of Istanbul soil and road dust reported in the literature

Metals (mg/kg)	Belivermiş et al. 2008	Guney et al. 2010		Coşkun Et al. 2006	Sezgin et al. 2003	Yetimoğlu et al. 2007	Mean	
	Soil	Soil	Road dust	Soil	Road dust	Road dust	Soil	Road dust
Cu	28.7	23.4	111	20	111.3	175.0	24.0	132
Ba				550			550	
Cd	0.14			0.2	1.91	0.3	0.17	1.1
Ca				30,700			30,700	
Cr	35.6			173			104.3	
Co	8.2			11			9.6	
Fe	9600			26,900			18,250	
Pb		29.7	177	33	211.9	198.3	31.3	195.7
Mg[a]				–				
Mn				600	397.9	6573	600	417.7
Ni	23.9			50	30.0	26.2	36.9	28.1
K				20,100			20,100	
Se[a]				–				
Na				7800			7800	
V				78			78	
Zn	82.0	96.6	245	45	447.7	414.7	74.5	369.1
Mo				0.6			0.6	

The EF of the elements was obtained using the following formula: $EF_i = [C_x / C_{Fe}]_{air} / [C_x / C_{Fe}]_{soil}$

where $[C_x / C_{Fe}]_{air}$ is the concentration ratio of elements depending on the size of each PM sample and $[C_x / C_{Fe}]_{soil}$ is the concentration ratio of elements in the soil

[a] Reference soil values, Mg: 529 mg/kg, Se 1.5 mg/kg

structure of the soil in Anatolia and to the scantiness of vegetation (Yatin et al. 2000; Bayraktar et al. 2010). Istanbul is a city surrounded by seawater, and sea aerosols blending into the air were also thought to be a factor in the finding of such high concentrations of these metals. As a matter of fact, studies conducted in other cities situated by the sea, such as Antalya and Izmir, also reported high concentrations of metal ions, specifically those of Na and Mg, as was the case in the present study (Na, 0.52–1.90 μg/m^3 and Mg, 0.09–0.36 μg/m^3) (Güllü et al. 2000; Yatkın and Bayram 2007). Sea salt is composed of 30 % Na and 1.5 % Mg. A high Na/Mg ratio is an indicative parameter for the blending of sea salts into the PM in air. This ratio was determined as the highest (nearly 14) in Goztepe, followed by Kilyos (nearly 7.5). Although the Mg concentrations at these two locations were similar, the concentration of Na was almost twofold higher in Goztepe, resulting from the structural composition of the soil as well as from use of coal as a source of heating. Other studies conducted in the Marmara region also reported a substantial contribution of dust transported from Asia and of sea salt to the measurement of high concentrations of PM (Rodriguez et al. 2004; Karaca et al. 2009; Koçak et al. 2011; Theodosi et al. 2010).

The highest concentrations of the metals that are thought to be of anthropogenic origin—Zn, Cu, Pb, Ni, Cr, V, Se, Co, and Cd—were determined in Avcilar, Goztepe, and Besiktas (Table 2). The highest value of Pb and Cd was measured in Avcilar (3.5-fold and 2.1-fold higher, respectively, than the values measured in Kilyos). Pb and Cd were reported as

Table 2 Elemental concentration (mean and standard deviation, ng/m^3) in total particulate matter (TPM), fine (<2.6 μm) to coarse (>2.6 μm) ratios (F/C), MMADs, and differs from background station in all stations (Avcılar, Beşiktaş, Göztepe, Rasathane, Kilyos)

Stations	Metals																	
	Cr	Ni	Pb	Cu	Cd	Mn	V	Sn	Co	Mo	Se	Ba	Zn	Fe	Na	K	Mg	Ca
Avcılar																		
Mean	7.0	4.8	16.7	5.9	0.9	16.4	2.6	62.6	0.3	0.3	2.2	117.5	213.3	1100.6	2285	399	419	7727
SD	3.1	3.9	10.6	3.6	0.5	11.2	2.1	32.9	0.1	0.3	1.8	61.9	88.5	666.7	1041	122	263	4504
F/C	0.6	0.7	3.2	0.5	1.1	0.5	0.9	1.0	0.5	0.5	0.9	0.6	1.0	0.5	0.7	0.8	0.6	0.3
MMADs	3.3	2.9	1.3	4,1	2.3	3.9	2.2	2.3	4.2	3.9	2.4	3.2	2.6	4.2	3.0	2.9	3.6	4.6
A/Background	2.0	1.5	2.3	0.8	2.0	1.6	1.5	1.8	3.5	1.8	1.6	2.3	2.2	3.4	1.6	0.9	2.1	2.0
Beşiktaş																		
Mean	7.9	4.3	7.3	15.0	0.7	13.6	3.1	52.4	0.4	0.9	2.4	109.9	216.0	1034.9	1862	494	272	5454
SD	3.7	1.8	4.8	7.2	0.3	6.7	1.9	20.9	0.2	0.3	1.2	36.1	181.9	364.9	1395	203	131	2276
F/C	0.7	0.6	1.9	0.4	1.1	0.4	1.2	0.8	0.4	0.5	0.8	0.8	0.9	0.3	0.6	0.6	0.9	0.5
MMADs	3.1	3.4	1.4	4.0	2.2	4.4	1.7	3.0	4.0	3.6	2.9	2.7	2.7	4.4	3.4	3.5	3.0	3.6
B/Background	1.4	1.2	2.1	1.0	1.3	1.5	1.3	1.5	1.5	2.1	1.5	1.2	2.7	3.6	1.3	1.1	1.4	1.4
Rasathane																		
Mean	4.4	3.1	5.5	5.4	0.5	7.6	1.6	49.6	0.2	0.2	0.8	97.2	188.4	564.4	1480	453	310	3194
SD	2.2	1.8	3.7	4.3	0.3	4.2	1.1	30.5	0.1	0.1	0.6	54.6	143.6	261.9	727	217	212	1235
F/C	0.7	0.7	3.0	0.6	1.6	0.7	1.6	0.6	0.5	0.8	0.9	0.5	0.6	0.5	0.5	0.5	0.7	0.3
MMADs	2.9	3.1	0.8	3.8	1.8	3.0	1.2	3.3	3.7	2.9	2.8	4.0	3.3	3.7	3.5	3.4	3.1	4.4
R/Background	0.8	1.0	1.2	0.9	1.5	1.4	1.3	0.9	1.2	1.2	1.1	1.1	1.4	1.3	1.0	1.0	1.6	0.8
Göztepe																		
Mean	7.5	3.8	10.8	19.1	0.6	16.7	2.7	62.8	0.6	0.5	2.9	114.8	152.4	1303.4	2823	474	191	9908
SD	3.2	2.5	14.8	10.6	0.6	5.5	2.0	23.7	0.6	0.5	2.5	55.9	83.8	595.2	1679	246	93	5673
F/C	0.6	0.3	2.1	0.5	2.0	0.5	0.9	0.6	0.3	0.7	0.6	0.4	0.7	0.5	0.6	0.8	0.5	0.4
MMADs	3.3	4.2	1.7	3.6	1.0	3.7	2.5	3.4	4.4	3.2	3.5	4.3	2.8	4.0	2.5	2.7	4.0	4.2
G/Background	2.6	2.0	2.7	1.0	1.0	1.5	1.5	1.9	2.2	2.0	1.3	2.6	2.3	4.5	2.0	1.0	1.0	2.6
Kilyos																		
Mean	3.8	2.9	4.8	2.2	0.4	8.9	1.2	41.0	0.2	0.1	0.7	65.0	99.3	486.2	1443	460	197	3804
SD	2.1	1.6	3.6	0.9	0.3	4.1	0.4	26.9	0.1	0.1	0.9	46.3	52.5	263.5	694	376	99	2017
F/C	0.5	0.5	1.8	0.5	1.1	0.5	1.3	0.6	0.4	0.5	0.5	0.5	0.6	0.5	0.5	0.5	0.5	0.5
MMADs	3.7	3.9	2.0	4.2	2.1	4.1	1.8	3.3	4.1	3.8	3.5	3.5	3.1	4.1	3.4	3.9	3.8	3.7

SD standard deviation, *F/C* fine to coarse ratio, A/Background: Avcılar/Kilyos, B/Background: Beşiktaş/Kilyos, R/Background: Rasathane/Kilyos, G/Background: Göztepe/Kilyos

important emissions from the cement industry (Yatkın and Bayram 2007). Because a cement plant is situated to the north of the Avcilar sampling station, the high concentrations of Pb and Cd were assumed to be associated with this source of emissions. Previous studies have also reported Pb and Cu as the indicator elements of traffic-sourced emissions (Querol et al. 2001; Manoli et al. 2002; Fang et al. 2003). Indeed, traffic-sourced PM formation was identified to be important in Besiktas, Goztepe, and Avcilar, where the highest concentrations for those two elements were measured. Specifically, Cu concentration was 2.6- to 8.5-fold higher in those stations than the concentration measured in Kilyos. Similarly, V and Fe, possible constituents of road dust, were measured at high concentrations in these stations. A comparison of the fine and coarse dust indicated that Pb, Cd, and V were over-represented in fine PM throughout the measurements made at all stations in Istanbul (F/C > 1). MMAD values and F/C ratios were observed to be strongly negatively correlated and having an average coefficient of 0.87 ranging between 0.55 and 0.99. This situation indicated that a lower mean diameter in the distribution of PM throughout the atmosphere in Istanbul is associated with a growing presence of anthropogenic elements in fine dust. The inhalation of toxic elements such as Pb and Cd in the fine dust imposes a potential public health risk.

The concentrations measured in the present study all fall below the limits dictated by national and international institutions (99/30/EC EU-Directive, US-EPA, WHO). However, WHO and EPA classify Cr and Ni as carcinogenic, and V, Mn, and Cd as non-carcinogenic, but as toxic elements imposing health risks. Theodosi et al. (2010) reported Ni and Cd concentrations in Istanbul as 7 and 1 ng/m^3, respectively, both below the limits as reported in the present study.

Size distribution of elements

The size distribution of elements expressed as dC/dlogDp versus Dp (PM size) is presented in Fig. 2. The size distribution and As and Hg concentrations in PM was reported in our previous studies (Şahin et al. 2012a; Şahin et al. 2013; Onat and Şahin 2014). We observed the tri-modal distribution with two peaks in the fine and coarse mode of PM; however, Fig. 2 shows that in contrast to PM, most elements showed a unimodal distribution, with one peak either in fine or coarse mode. Generally, elements accumulated in the fine mode (<1 μm) were Pb, Cd, Ni, and V, whereas Ca, K, Mg, Fe, Co, and Mn elements accumulated in the coarse mode (>3 μm). The results for As and Hg were reported in our previous study, and As occurred predominately in the fine size (Onat and Şahin 2014). Similar results for the size distribution of elements were observed in Thessaloniki, Greece (Voutsa et al. 2015; Taiwo et al. 2014). However, Lü et al. (2012) reported in their study in Shanghai that V and Ni had a wide range of distribution. Therefore, it appears that the size distribution of elements depends on the location and sources.

The maximum peaks for Se, Sn, and Zn were observed in PM with size less than 1 μm in the samples collected from the Avcilar station (Fig. 2). The high concentration of these elements in fine PM in the samples collected during the summer was thought to be a consequence of the industrial activity situated around the Avcilar sampling station. A major fraction of Pb and V were measured in PM having size <2.6 μm. These elements are pollutants of anthropogenic sources, caused by the employment of fuel oil for heating around the area. High Pb and V concentrations were thought to be caused by the thermal power plant situated in the western part of the area, which uses fuel oil and natural gas. Pb, Cd, and Zn showed distributions similar to a typical industrial city, Beijing in China and Ulsan in Korea (Wang et al. 2016; Duan et al. 2012; Nyand Lee 201). The PM < 2.6/PM > 2.6 ratio for Na and K was close to 1, whereas it was nearly 0.3 for Ca and Mg. Natural elements and sea aerosols were measured at the sampling station. Street dust was observed to be a substantial contributor to the PM composition at the Besiktas sampling station, with Mn, Cu, and Fe elements having slightly higher concentrations during seasonal transitions, specifically in the coarse PM.

The high concentration of anthropogenic pollutants (Ni, Pb, Cd, and V) in fine dust in the Rasathane area, where traffic is not an important contributing factor to pollution, indicated that the pollution was mostly due to domestic heating. Furthermore, Rasathane is situated by the Bosphorus and thus easily affected by the metallic pollution resulting from the vessel traffic. The concentration of all metals in the PM was observed to be periodically higher during fall. The Goztepe sampling station is a highly residential area with traffic congestion and is geographically situated at a low altitude. For that reason, the pollutants cannot always be dispersed as the atmospheric conditions change and reach considerable levels. The PM < 2.6/PM > 2.6 ratio in Kilyos was determined to be in the range of 0.4 to 0.7 for Na, Mg, and Ca. Sea aerosols were thought to be an important factor contributing to this outcome.

Seasonal variation of elements

The sampling periods in this study were arranged to include seasonal transition periods representing all four seasons. The effect of four seasons on the particulate matter concentration was determined in our previous study (Şahin et al. 2012a). However, the limited number of samples which is representative of each season does not allow the deduction of definitive conclusions from the values obtained for the concentrations of the metals under investigation. In many studies on air pollution, the heating and the non-heating seasons are different (Yatin et al. 2000; Voutsa and Samara 2002; Bayraktar et al. 2010). Domestic heating in densely populated residential

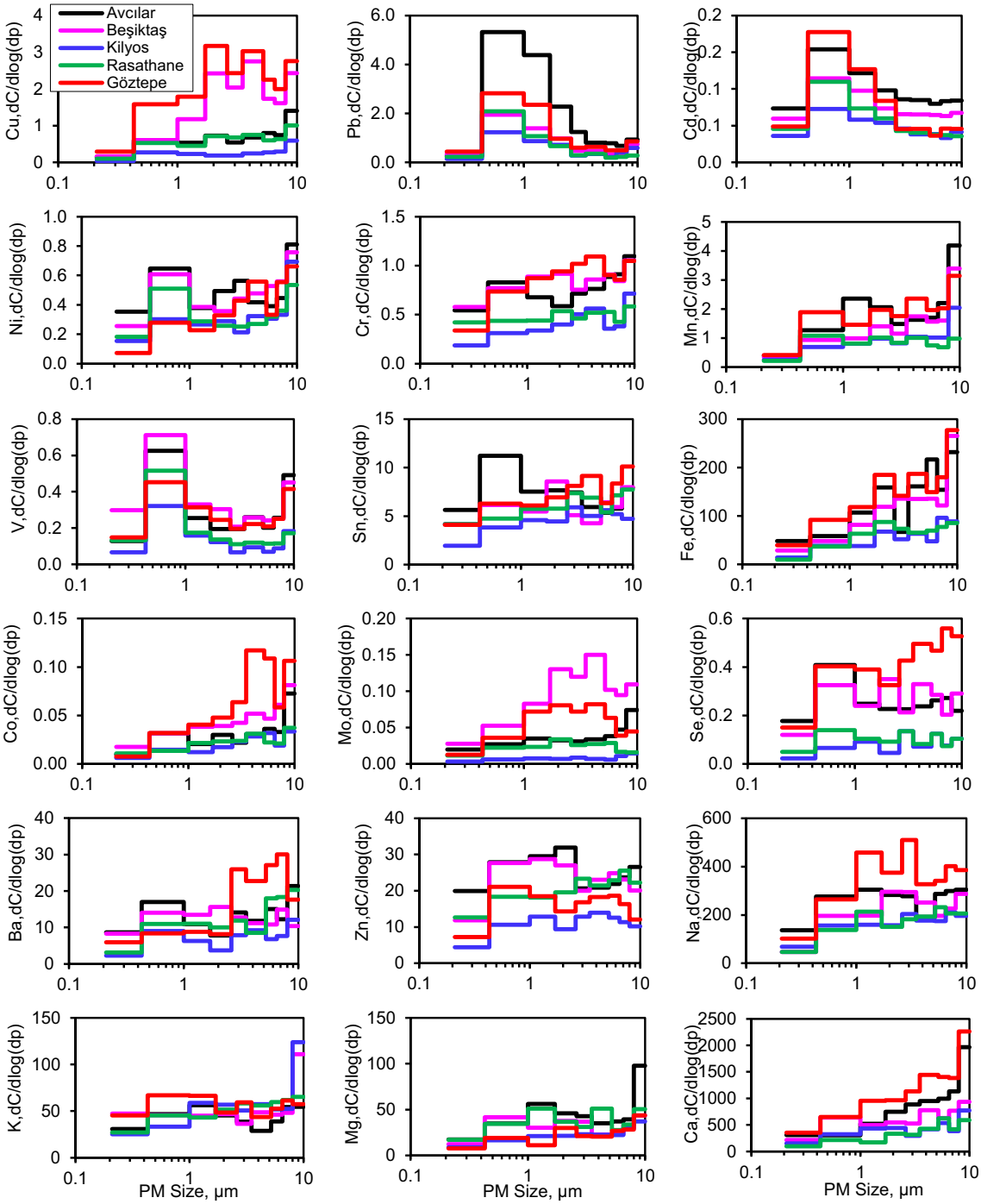

Fig. 2 Size distribution of elemental concentrations at five measurement stations

areas is a significant source of air pollution in Istanbul. For this reason, the data obtained in this study were considered in terms of heating and non-heating periods. In determining these periods, the local air temperatures during the sampling periods were considered. Periods when the air temperature was above 18 °C were considered non-heating periods, and the rest were considered heating periods. The mass percentage of the elemental size fraction of particulate matter for two seasons in the five areas of Istanbul atmosphere is given in Fig. 3.

As seen in Fig. 3, 65–80 % of all soil-based metals (Ca, Mg, Fe, and Mn) were measured in the coarse PM fraction throughout the sampling periods. On the other hand, 82 % of Pb, 53 % of Cd, as well as 48 % of V, Sn, and Se were measured in PM of size <2.6 µm in Avcilar. Slight increases were observed in the concentration of these elements during

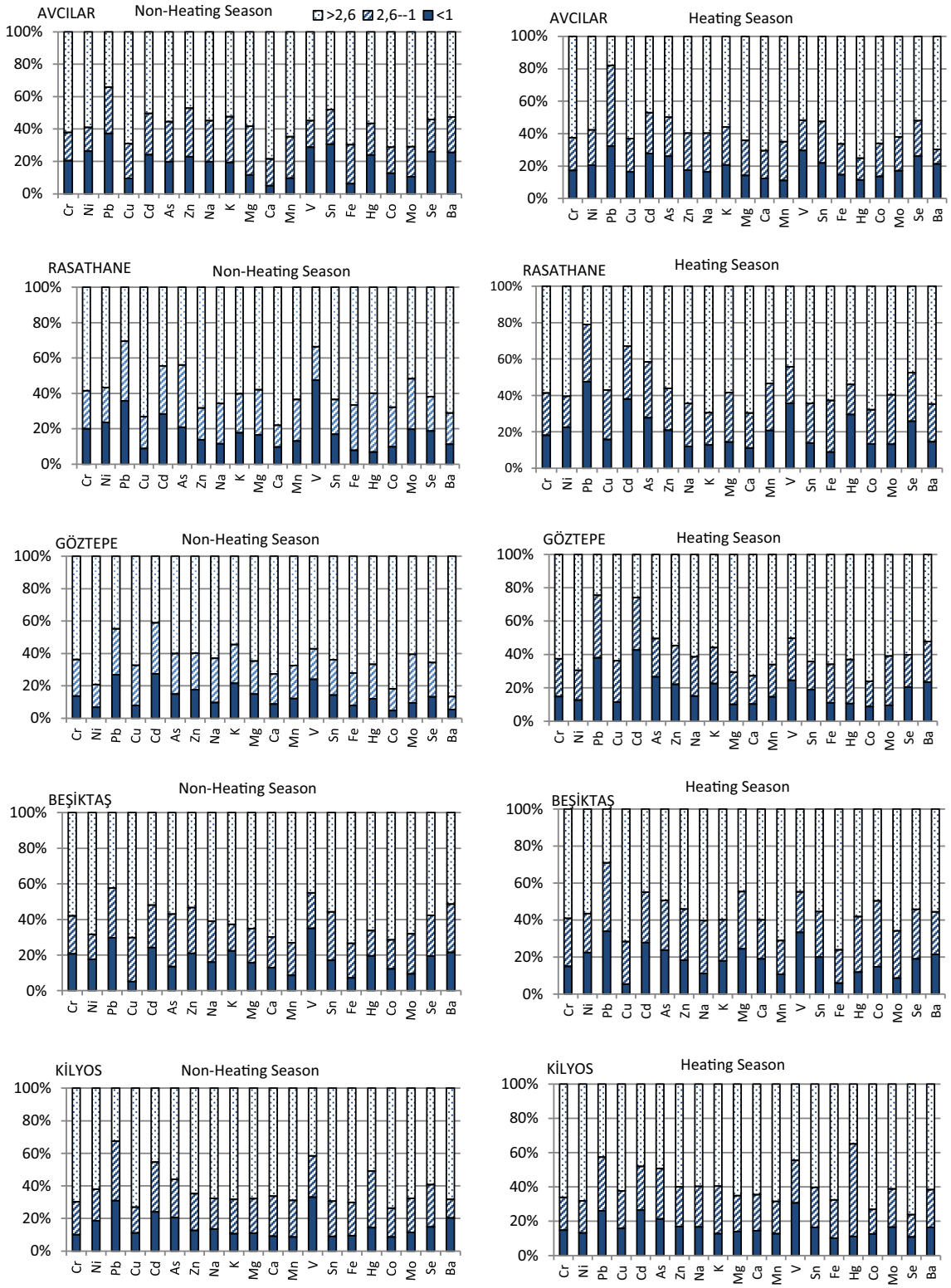

Fig. 3 Mass percentage of the elements as size fraction of particulate matter for two seasons in the five areas of Istanbul atmosphere

periods of heating, with this increase for Pb in the fine PM fraction being as high as 25 %. A similar situation was observed for the samples collected in Besiktas, with 71 % of the total Pb to be measured in the fine PM fraction during periods of heating, whereas this fraction was 58 % during the remaining months of the year. Furthermore, 50–55 % of Cd, V, and Co were also deposited in the fine dust during periods when heating systems were operating. In addition, a substantial

fraction of total Mg content (55 %) was also determined among the fine dust particles. The 50–75 % of the fine dust fraction samples in Goztepe was identified to be composed of Pb, Cd, and V, and the concentration of these elements increased by 20–40 % during the period when heating was needed. Additionally, 48 and 31 % of Ba and Ni, respectively, were identified in the fine fraction during the heating period, whereas these numbers were reduced to 13 and 21 % at other times. Domestic heating and the use of fossil fuels in transportation were observed to increase the relative composition of these pollutants in the fine fraction during periods when heating systems were in operation.

The relative ratio of Pb, Cd, and Se in the fine PM fraction was determined as 79, 67, and 53 %, respectively, in the samples collected in Rasathane during the period when heating systems were on, whereas these ratios were determined as 14, 20, and 38 % when heating systems were not operating. In addition, the ratio of Cu in the coarse PM fraction was determined as 57 % during the period when heating was on, whereas it was determined to be 73 % at other times. A similar increase in the ratio of Zn, Ca, Ba, and Mn was observed in the coarse fraction when heating was not required. The highest ratios of Pb and V in the fine PM fraction were determined as 65, 57, and 55 % in Kilyos during the period of heating. Approximately 60–65 % of the elements originating from natural resources, including Ca, Mg, Na, and Fe, were identified in the coarse PM, and a further increase was observed during summer months.

Enrichment factor analyses

Figure 4 shows the EFs of elements in coarse and fine particles at the five stations. Many elements were observed to assume higher EF values in fine particles than in coarse particles in samples collected from all five stations, and therefore, were more enriched. Specifically in Besiktas, all elements except for Hg, Na, and Cu were enriched in fine dust particles 4.4-(Mo) to 17.1-fold (Pb), indicating a greater contribution of non-crustal sources associated with fine particles (Wang et al. 2016). The EF of Cd, Zn, As, Se, and Pb in fine dust was determined to be above 100 in Avcilar and above 50 in Goztepe and Besiktas. The fine dust samples collected at these stations were most likely of combustion origin. In addition, the effect of industrial plants was also notable in Avcilar. Se and As are markers for coal combustion (Duan et al. 2012). There is a thermic power plant and cement industry in Avcilar, and these areas have the greatest population. The enrichment factor of Mn, K, and V in coarse dust samples was determined as below 1, identifying these as the least enriched elements. The enrichment factors of Pb, Cu, Ni, Cr, and Co were determined to vary in the range from 1 to 10. This result show that these elements should mainly come from the re-suspended soil containing previously deposited. The enrichment of Mg, an important indicator of sea aerosols, in fine dust was approximately 7.2-fold higher than in coarse dust, with EF of nearly 10 in coarse dust and nearly 100 in fine dust. This observation was an indicator of the possibility of having sea salts in PM, as was previously reported in the literature (Wang et al. 2006; Ny and Lee 2010). The EFs of Ca in coarse mode (7.7–13.6) were lower than those in fine mode (14.5–26.5). The abundant Ca particles originated from local construction activities (Duan et al. 2012). Istanbul is a city that routinely allows immigration, and consequently there is incessant construction activity. The enrichment of Ca in fine dust particles was therefore interpreted as an indicator of the construction activity in the area.

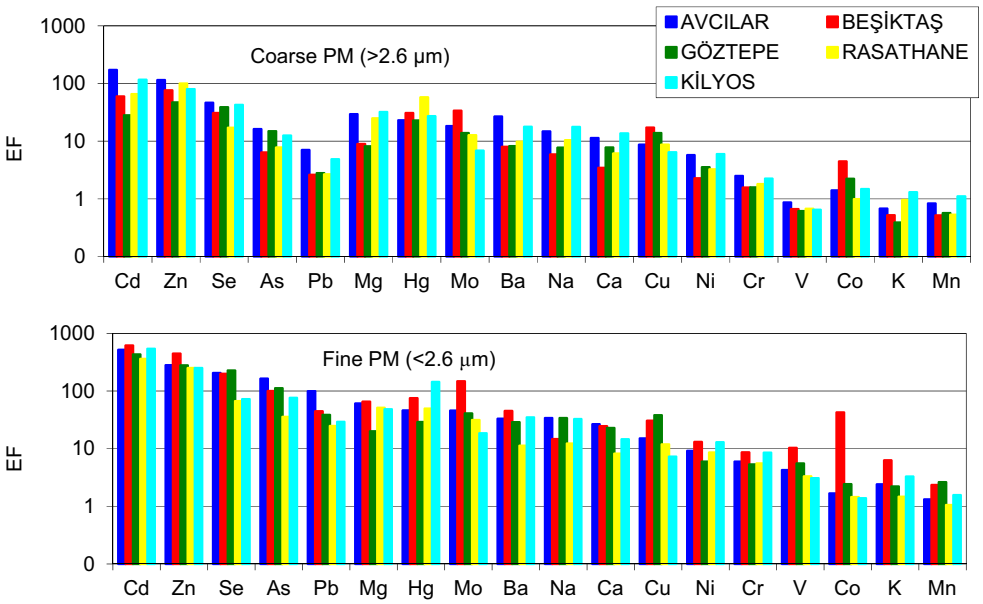

Fig. 4 The enrichment factors of elements in coarse (>2.6 μm) and fine (<2.6 μm) particulate matter

Principle component analyses

In this study, we used principal component analysis (PCA) using varimax rotation and retention of the principal component with eigenvalues below 1 to determine and estimate possible sources of fine and coarse size fractions of PM (SPSS 17). The impactor size ranges were divided into two categories: fraction 1 (<2.6 μm: fine particulate matter) and fraction 2 (>2.6 μm: coarse particulate matter). We determined five possible basic sources: soil, road dust, sea aerosol, industry, residential, and traffic; identified as factors, they explained a total variance of almost 85 %. Table 3 shows the varimax rotated principal component loading for the fine and coarse fraction of particulate matter collected at the five stations in Istanbul.

The atmospheric particulate matter in Istanbul was shown to originate from a variety of sources. A similar observation was made in other studies available in the literature on Istanbul; the effect of a number of sources on the mass concentration of PM was discussed using a source analysis (Hanedar et al. 2011; Koçak et al. 2011; Karaca et al. 2009; Theodosi et al. 2010). More specifically, Koçak et al. 2011 identified seven sources contributing to the PM_{10} concentration in Istanbul—secondary, combustion, traffic, fuel oil, soil, crystal, and sea aerosols—in the source analysis they carried out. The pollutants from these sources were determined to constitute 98 % of the total mass. However, this study was conducted in a non-industrialized area, and therefore the contribution of industrial activity to the pollution was not evaluated.

In the present study, the factor analysis carried out on samples collected from Avcilar revealed fuel oil and coal combustion as those affecting the concentration of Cr, Ni, Cd, and Mn elements. Industrial processes (specifically those of metal casting and oil combustion processes) were the factors affecting the concentration of Zn and Mn elements in both fine and coarse fractions as the first and the second most effective factors, respectively. Additionally, the measurements regarding Pb, Cu, Mn, V, Fe, and Hg made in Avcilar identified road dust as an important secondary source in coarse PM. Furthermore, the sea salt elements Na and Mg highlighted as the third factor indicated the importance of sea aerosols. The overrepresentation of Sn, Mo, and K elements in the third factor of the fine dust was thought to result from industrial activity (specifically, the metal industry) in the area. The fourth factor elements in fine dust were identified as Cu, V, and Ba. The identification of these elements indicated the sampling location to be an important spot for the traffic activity because of the E-5 motorway.

The first factor effective in the composition of fine dust samples in Besiktas was determined as pollution resulting from intense traffic activity, whereas the next factors (second, third, fourth, and fifth) were associated with domestic heating. In addition, the second and sixth factors affecting the composition of the fine dust identified sea aerosols as an important contributor. Coarse PM was mainly due to road dust and traffic activity.

Nearly 60 % of the variation in the samples collected from Goztepe sampling station were observed to be explained by only the first two factors; the samples comprising anthropogenic elements such as Cr, Ni, Pb, Cd, Cu, Mn, V, Hg, Mo, Se, and K were detected due to the presence of some combustion event. The presence of As, Mn, Sn, and Ca in the third factor was an indicator of the presence of road dust as well as the intensive residential settlements and associated emissions caused by the construction activity in the area. A similar situation was observed in the second factor of the coarse PM analysis through the detection of Sn and Fe metals. The first factor of the coarse PM explained 30.4 % of the variance in the data by representing the subset of elements associated with the road dust (Pb, Cu, Mn, V, Fe, and Mo).

The first and the second factors explain 59.2 % of the variance in the fine dust in Rasathane, and they represent the elemental groups of heating (Cr, Cu, Cd, As, Zn, V, Ba, K, Fe, Hg, and Se) and sea aerosol (Na and Mg) origin. The third factor explaining the variance in the fine dust included Pb, Cu, Mn, and Co, indicating that pollution was caused by the traffic as carried through the fine dust in the area. The first factor in the coarse PM fraction was representative of the soil and road dust elements, whereas the second and the third factors represented heating-associated elements. Elements of soil origin were determined to be effective in the accumulation of PM as explained by the fourth and the fifth factors.

The first two factors were observed to explain 50 % of the variation in the coarse PM in Kilyos, and these two factors were populated with elements of soil origin. The group of elements detected as a result of the combustion of petroleum derivatives—Ni, Pb, Cd, Zn, Mn, and Co—were identified as the third and the fourth factors. The anthropogenic elements Pb, Cd, and Co comprised the first factor group in fine PM, explaining 22.2 % of the total variance. Sn, Hg, K, and Mg populated the second factor, representing the substantial contribution of sea salt to fine dust pollution. Other factor groups in fine dust indicated a combustion-based pollution. The transport model studies conducted in Istanbul reported a major contribution of long-distance transport to the PM concentration in the atmosphere in Istanbul (Koçak et al. 2011). Fine dust from Europe and Russia could possibly be carried toward Istanbul. Additionally, Kilyos is situated where sea vessels enter the Bosphorus from the Black Sea.

Conclusion

A study on size distributions of elements was carried out at urban and rural sites in Istanbul, Turkey. For this proposes,

Table 3 Varimax rotated principal component loading for the fine and coarse fractions of particulate matter collected at the five stations in Istanbul

Avcilar															
Coarse PM (>2.6 μm)						Fine PM (<2.6 μm)									
Metals	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5	F6	F7			
Cr	0.86					0.90									
Ni	0.70			0.51		0.53									
Pb		0.69		0.67							0.65				
Cu		0.89							0.73						
Cd	0.85					0.82									
As			0.72							0.87					
Zn	0.77						0.78								
Mn	0.61	0.64					0.49					0.50			
V		0.88							0.82						
Sn			0.84					0.82							
Fe		0.84					0.53				0.59				
Hg		0.65					0.96								
Co					0.84						0.84				
Mo				0.65	0.60		0.63	0.68							
Se			0.57									0.95			
Ba					0.76	0.45			0.65						
Na	0.69					0.88									
K				0.80				0.86							
Mg	0.57		0.67							0.85					
Ca			0.81							0.60					
Var. %	30.56	17.36	14.29	10.29	7.96	26.09	19.1	11.42	9.135	8.027	6.384	5.389			
Cum %	30.56	47.91	62.21	72.49	80.45	26.09	45.2	56.66	65.79	73.82	80.2	85.59			
Sources	IN, SA	TR, RD	SA	RE	RE	RE, IN	IN	IN	TR	SO	RD	RE, IN			
Beşiktaş															
Coarse PM (>2.6 μm)									Fine PM (<2.6 μm)						
Metals	F1	F2	F3	F4	F5	F6	F7	F8	F1	F2	F3	F4	F5	F6	F7
Cr			0.94										0.78		
Ni					0.90						0.70				
Pb		0.78							0.90						
Cu	0.60									0.62			0.52		
Cd	0.91												0.69		
As		0.86							0.60						
Zn				0.62								0.91			
Mn		0.86							0.87						
V				0.59								0.7			
Sn						0.77									0.84
Fe	0.75										0.77				
Hg							0.60						0.73		
Co						0.88									0.84
Mo								0.88	0.60	0.5					
Se	0.85									0.74					
Ba				0.86								0.79			
Na							0.84		0.53					0.71	
K			0.90								0.76				
Mg							0.66			0.81					
Ca					0.66									0.90	
Var. %	21.4	14.9	13.7	10.3	7.6	6.9	6.3	5.8	27.5	14.2	12.0	10.3	10.0	7.3	5.2

Table 3 (continued)

Cum %	21.4	36.3	50.0	60.2	67.8	74.7	81.0	86.8	27.5	41.7	53.7	64.0	73.9	81.2	86.4
Sources	RD	TR	RE	RD	SO	RD	SA	IN	TR	RE, SA	RE	RE	RE	SA	RD
Göztepe															
Metals	Coarse PM (>2.6μm)						Fine PM (<2.6μm)								
	F1	F2	F3	F4	F5	F6	F1	F2	F3	F4	F5				
Cr	0.59						0.80								
Ni					0.81		0.76								
Pb	0.91						0.97								
Cu	0.88						0.94								
Cd		0.79					0.96								
As			0.94						0.84						
Zn					0.57						0.85				
Mn	0.86						0.79		0.50						
V	0.62						0.94								
Sn		0.53							0.55						
Fe	0.73	0.49								0.71					
Hg			0.83				0.98								
Co					0.92					0.89					
Mo	0.91						0.97								
Se				0.90				0.83							
Ba				0.93							0.77				
Na						0.87	0.84								
K		0.94						0.77							
Mg		0.78						0.92							
Ca			0.88						0.82						
Var. %	30.36	18.77	16.09	11.51	7.55	5.54	44.29	15.67	10.74	8.87	6.93				
Cum %	30.36	49.13	65.23	76.74	84.29	89.83	44.29	59.95	70.69	79.56	86.49				
Sources	RD, RE	RD	SO	TR	RE	SO	RE, TR	RE	IN	IN	IN				
Rasathane															
Coarse PM (>2.6μm)							Fine PM (<2.6μm)								
Metals	F1	F2	F3	F4	F5	F6	F1	F2	F3	F4	F5				
Cr		0.54				0.56		0.69							
Ni			0.80							0.55					
Pb	0.87								0.81						
Cu	0.91						0.68		0.52						
Cd		0.83						0.81							
As	0.77						0.61								
Zn		0.83					0.72								
Mn	0.54			0.46					0.95						
V						0.95	0.63								
Sn	0.68							0.85							
Fe	0.74							0.70							
Hg		0.89						0.91							
Co			0.60	0.55					0.54	0.68					
Mo					0.88					0.82					
Se	0.80							0.69							
Ba	0.73						0.91								
Na				0.87			0.85								
K			0.78				0.78								
Mg	0.62			0.60			0.82								
Ca				0.59	0.59						0.87				
Var. %	41.70	15.01	9.24	8.93	7.42	5.96	45.38	13.80	12.14	8.21	5.99				
Cum %	41.70	56.71	65.95	74.89	82.31	88.27	45.38	59.19	71.33	79.54	85.53				
Sources	SO, RD	RE	RE	SO	SO	RD	RE, SA	RE	TR	RD	SO				
Kilyos															
Coarse PM (>2.6μm)							Fine PM (<2.6μm)								
Metals	F1	F2	F3	F4	F5	F6	F1	F2	F3	F4	F5	F6	F7		
Cr	0.63									0.74					
Ni				0.73									0.88		
Pb			0.81				0.88								
Cu		0.70							0.81						
Cd			0.63				0.80								
As		0.88									0.93				
Zn	0.65			0.50						0.69					

Table 3 (continued)

Mn	0.70			0.50								0.81	
V			0.81							0.73			
Sn	0.89							0.83					
Fe	0.86								0.60				0.57
Hg	0.51				0.80			0.78					
Co				0.64			0.83						
Mo		0.93									0.83		
Se					0.95							0.74	
Ba						0.87			0.89				
Na		0.73							0.64				
K	0.93							0.89					
Mg		0.62						0.51				0.63	
Ca	0.58					0.69				0.86			
Var. %	34.00	16.14	13.31	8.90	7.61	5.34	22.17	18.18	12.96	12.12	11.06	6.41	6.15
Cum %	34.00	50.15	63.45	72.35	79.96	85.30	22.17	40.34	53.30	65.42	76.48	82.89	89.04
Sources	SO	SO, SA	RE	RE	SO	SO	RE	SO, SA	SO	RE	RE	RE, SA	RE

PM samples were collected from five different sampling stations (Avcilar, Besiktas, Kilyos, Rasathane, and Goztepe) throughout the city; these were thought to represent the source variability, although none of them could specifically represent a single source.

Among the measurements made on metals with soil origin, the highest values of Fe and Ca were measured in Goztepe, and the highest values of K and Mg were measured in Kilyos and Avcilar, respectively. The concentration of these metals was 1- to 2.6-fold higher in Besiktas, Avcilar, and Goztepe in comparison with their levels determined in the samples collected from the Kilyos station, situated in a rural area of the city. However, an important difference could not be observed in the measurements made on the samples collected in Rasathane. Similar studies conducted in other cities in Turkey reported high concentrations for these metals, specifically in coarse dust, in comparison with measurements made in other cities in Europe; this was attributed to the particular compositional structure of the soil in Asia Minor and the scarcity of vegetation.

The high number of factors identified to affect the composition of the atmospheric dust in Istanbul indicated a diversity of sources responsible for this pollution. A similar situation has been reported repeatedly in earlier publications on Istanbul, and the source analysis revealed a variety of sources to be effective on the mass concentration of the PM. Specifically, the substantial presence of anthropogenic metals in fine particulate matter was attributed to industrial activity in Avcilar, traffic congestion in Besiktas, and traffic congestion together with domestic heating in Goztepe. The three most effective factors in Rasathane were determined as domestic heating, sea aerosols, and traffic congestion.

It is rather difficult to determine the overall air quality in Istanbul. It is a large metropolitan city with a population of 13 million residing in an area of 5712 km^2, and it is also an industrial hub in Turkey. Additionally, within Istanbul itself, there are considerable regional differences in geographical location and meteorological conditions. Furthermore, the transport of pollutants from both Asia and Europe into the city atmosphere contributes substantially to the local pollution caused by the PM. Five different sampling locations representing source variability, but avoiding any bias toward any one of those particular sources, were employed in the study. The physical characteristic and the elemental composition of the PM characterized in this study can be thought to represent, at least partially, the city atmosphere.

Acknowledgments We gratefully acknowledge the Scientific and Technical Research Council of Turkey (TUBITAK) with project no 107Y161 and the Research Fund of the Istanbul University with project no T-3783 and UDP-29334 for financially supporting our study.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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