Trace element concentrations of size-fractionated particulate matter in the atmosphere of Istanbul, Turkey

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Abstract

In this study, the physical characteristics and metal contents of particulate matter (PM) have been determined. In this scope, PM sampling has been done at 5 stations (Avcılar, Beşiktaş, Kilyos, Rasathane, Göztepe) for 2 years. PM filter samples were collected for 8 different size($<0.43 \mu m$, 1-0.43 μm , 1-1.7 μm , 1.7-2.6 μ m, 2.6-3.5 μ m, 3.5-5 μ m, 5.2-6.5 μ m, 6.5-8 μ m and >8 μ m) using the cascade impactor. PM filters were decomposed and analyzed for 8 metals (Cr. Mn, Fe, Cu, Pb, Ni, Cd and V). According to the results, trace element concentrations at all the stations were at least 2 times higher than at the Kilyos station which was selected as the urban background station. Pb, Cd and V elements are mainly present in fine fraction of PM, while Cu, Mn and Fe mass is found in coarse fraction of PM. Mass Median Aerodynamic Diameter (MMAD) values for crustal element (Fe, Cr, Cu and Mn) were found to be in the coarse size fraction and anthropogenic element (Pb, Cd and V) were found to be in the fine particles. The major PM source was observed industrial facilities at the Avcılar; traffic and domestic heating at the Beşiktaş, Göztepe and Rasathane. Keywords: air pollution, particulate matter, trace metals, size distribution, Istanbul.

1 Introduction

Atmospheric particles have a direct impact on health through respiration, and this effect is related to the physical and chemical properties of the particles. The efficiency of inhalation and respiratory deposition of particulate matter (PM) is dependent on the size of the particles [1]. Therefore, knowledge of particle size



is vital in understanding the effects of PM on human health. Particles that are less than 10 μ m in diameter have the potential to reach the lower respiratory tract from the nose down to the alveoli. Particles larger than 5 μ m in diameter are usually filtered out by the nose. Fine particles with diameters of less than 2 μ m reach deep into the alveolar regions of the human lung, where the absorption efficiency for trace elements is in the range of 60–80%. The chemical components in anthropogenic PM range from 0.001 to 2 μ m in diameter [1, 2].

Many researchers in Europe and Asia have conducted experiments to study the size distribution of PM in the last decade [3–15]. However, there have only been a few studies conducted in Turkey investigating PM_{10} and even fewer studies investigating $PM_{2.5}$ [16–19]. Furthermore, the size distribution of PM has been studied in Turkey limited number in the last year [20, 21]. In this study, the size distributions of particles at different locations in Istanbul across four seasons from 2008 to 2010 were investigated.

2 Materials and methods

Istanbul, located on the extremities of Asia and Europe, is the most populated city in Turkey, with a population of approximately 13 million. The city has 11,143 industrial establishments and 2,500,000 registered motor vehicles. In this study, inhalable particulate matter (PM) was sampled at five stations (Figure 1). The Avcılar urban station (40°59'21.45"N, 28°43'19.64"E) has high levels of vehicle, industrial and residential emissions. The Beşiktaş urban sampling station (41°3'6.59"N, 29°0'35.82"E) is located in a residential and commercial area, where the 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 Göztepe urban sampling station (40°59'47.36"N,



Figure 1: The particulate matter sampling locations of the study area in Istanbul, Turkey (<u>www.maps.google.com</u>).

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), which is occupied by very few residents. The sampling locations and size distribution analyses of PM were described in detail in our previous study [21].

PM were collected using a non-viable Andersen impactor with an airflow rate of 90 lpm. Quartz fiber filters (QF) with 81-mm diameters were used for the gravimetric and metal analysis. The Anderson impactor consists of eight stages (labeled 0-7) 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. Sampling was carried out from July 2008 to August 2010. A total of 16 PM samples were collected at each station. The duration of sampling in a single station was approximately 7±2 days. The filters within the impactor were transferred to clean Petri dishes with tweezers in a closed environment and were then brought into the laboratory. Following the removal of the filters, the impactor was cleaned. Clean filters were then installed before the system was taken to the next station. The sampling procedure and gravimetric analysis of the PM were described in detail in our previous studies [20, 21]. After tha gravimetric analysis, the aerosol samples were stored in the refrigerator at approximately 4°C prior to extraction.

The dust filters were decomposed in three different steps using microwave equipment (Cem Mars Xpress, microwave digestion system, USA). At the first step, 6 ml of HNO₃ and 2 ml of H₂O₂ were added to the dust filters and were allowed to decompose for 40 minutes at 180°C. At the second step, 2 ml HCl was added to the extract and the decomposition time was 35 minutes at 175°C. In the final step, 2 ml HF was added to the extract and the decomposition time was 30 minutes 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 10 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 concentration in the solution, graphite atomic absorption spectrophotometry (Perkin Elmer GF-AAS 600) was employed. This 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) was used as references. One mg of SRM 1648 was digested using the method described earlier to examine the capabilities of the analytical methods and to perform the recovery procedure. All elements had recoveries between %95 and %103. We selected one quartz filter as blank every PM sampling. Limits of detection (LODs), calculated as three times the standard deviation of blank samples, and were satisfactory for environmental monitoring. The mass concentrations of metals are well above these blank values.



3 Results and discussion

The concentrations of inhalable particulate matter and metal contents of eight size ranges at different locations in Istanbul were determined. The total metal concentration in total particulate matter (TPM), fine to coarse ratio (<2.6 μ m />2.6 μ m, F/C), mass median aerodynamic diameters (MMADs) of metals and differs from background station (Kilyos) in all stations (Avcılar, Beşiktaş, Rasathane, Göztepe) are displayed in Table 1.

The highest mean Pb, Ni, Cd, Cr, V, Mn, Cu, Fe concentrations in TPM was measured at the Avcılar, Beşiktaş and Göztepe stations (Table 1). The lowest mean metal values were measured at the Kilyos station. The total mean Pb, Cd and Ni concentrations at the Avcılar station were 3.5, 2.3 and 1.7 times higher, Cr and V at the Beşiktaş station were 2.1 and 2.6 times higher and Cu, Mn and Fe 8.7, 1.9 and 2.7 times higher than at the Kilyos station, which was selected as the city background. The Pb and Cd metals are major emissions of the cement factory [16]. The cement factory located in the north of the Avcılar sampling station can cause an increase of the Pb and Cd concentrations in Avcılar. Some studies showed that Pb and Cd elements were the indicator elements of the traffic emissions [22–24]. The highest concentrations were observed at Beşiktaş, Göztepe and Avcılar stations. The major PM source of these stations was traffic. Additionally, V and Fe, can be found in the composition of road dust, were also higher at the same stations.

The annual average limit values of Pb, Cd and Ni metal in PM_{10} are given as 500 ng/m³, 5 ng/m³ and 20 ng/m³ by 99/30/EC EU-Directive (99/30/EC EU-Directive), respectively. In this study, Cd, Pb and Ni concentrations were found below the limit values. In a recent study in Istanbul, Theodosi *et al.* [25] found that the Ni and Cd was 7 ng/m³ and 1 ng/m³, respectively, and their results were similar to this study.

The highest F/C ratios are observed for Pb at all stations, especially higher at the Avcılar and Rasathane stations. F/C ratios for Cd are changing between 1.1 and 2.0 and for V equal to 1. Pb, Cd and V elements are mainly present in fine fraction of PM, while Cu, Mn and Fe mass with F/C ratios approximately 0.5 is found in coarse fraction of PM. Similar results were reported by Karanasiou *et al.* [26].

Fig. 2 summarizes the size distributions of trace elements at all stations. The size distribution of Pb, Cd and V have their peaks mainly between 1 μ m and 0.43 μ m fraction at Avcılar, Göztepe and Beşiktaş stations, respectively. This fine particles accumulations showed to be anthropogenic sources at these areas. In general, the mass size distributions in crustal trace metals (Fe and Mn) and Cr showed coarse particle accumulation which was characterized by relatively higher peak at the size above 6.5 μ m. The other anthropogenic metal, Ni, showed bimodal patterns with primary peak at the size range of 0.43-1 μ m, and secondary peak at the size range of 2.6-5.2 μ m (Fig. 2). Cu is known that important finger print of brake wear source [27]. So the size distribution of Cu showed main peak at the size range of 1.7-3.5 μ m at the Göztepe and Beşiktaş stations which are near main traffic road (Fig. 1). The results showed that the



traffic is the first major source of PM and the second major source is domestic heating. In addition, road dust was an important component of atmospheric PM and Mn, Cu and Fe elements were found higher in coarse PM.

Table 1: MMAD values for metal concentration (mean and standard deviation, ng/m³) in total particulate matter (TPM), fine (<2.6 μ m) to coarse (>2.6 μ m) 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	Fe			
AVCILAR											
Mean	7.0	4.8	16.7	5.9	0.9	16.4	2.6	1100.6			
SD	3.1	3.9	10.6	3.6	0.5	11.2	2.1	666.7			
F/C	0.6	0.7	3.2	0.5	1.1	0.5	0.9	0.5			
MMADs	3.3	2.9	1.3	4,1	2.3	3.9	2.2	4.2			
A/Background	1.8	1.7	3.5	2.7	2.3	1.8	2.2	2.3			
BEŞİKTAŞ											
Mean	7.9	4.3	7.3	15.0	0.7	13.6	3.1	1034.9			
SD	3.7	1.8	4.8	7.2	0.3	6.7	1.9	364.9			
F/C	0.7	0.6	1.9	0.4	1.1	0.4	1.2	0.3			
MMADs	3.1	3.4	1.4	4.0	2.2	4.4	1.7	4.4			
B/Background	2.1	1.5	1.5	6.8	1.8	1.5	2.6	2.1			
RASATHANE											
Mean	4.4	3.1	5.5	5.4	0.5	7.6	1.6	564.4			
SD	2.2	1.8	3.7	4.3	0.3	4.2	1.1	261.9			
F/C	0.7	0.7	3.0	0.6	1.6	0.7	1.6	0.5			
MMADs	2.9	3.1	0.8	3.8	1.8	3.0	1.2	3.7			
R/Background	1.2	1.1	1.1	2.5	1.3	0.9	1.3	1.2			
GÖZTEPE											
Mean	7.5	3.8	10.8	19.1	0.6	16.7	2.7	1303.4			
SD	3.2	2.5	14.8	10.6	0.6	5.5	2.0	595.2			
F/C	0.6	0.3	2.1	0.5	2.0	0.5	0.9	0.5			
MMADs	3.3	4.2	1.7	3.6	1.0	3.7	2.5	4.0			
G/Background	2.0	1.3	2.3	8.7	1.5	1.9	2.3	2.7			
KİLYOS											
Mean	3.8	2.9	4.8	2.2	0.4	8.9	1.2	486.2			
SD	2.1	1.6	3.6	0.9	0.3	4.1	0.4	263.5			
F/C	0.5	0.5	1.8	0.5	1.1	0.5	1.3	0.5			
MMADs	3.7	3.9	2.0	4.2	2.1	4.1	1.8	4.1			

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.



Figure 2: The mass size distribution of the mean metals in the five stations.

As seen in Fig. 2 the mass size distribution of a particular compound-species is approximately lognormal and unimodal. So the mass median aerodynamic diameter (MMADs) and the geometric standard deviation (GSD) are able to define this type of distribution [1, 26, 28]. The cumulative size distribution was

constructed using the method which described detailed in our previous study [21]. Using the regression lines of the log-normal distribution, mean MMAD were calculated for every element. In the case of particle metal concentrations a least-square fit, R² values greater than 0.95 indicating that these metals have a unimodel distribution. MMADs results are given in Table 1. MMAD values for Fe, Cr, Cu and Mn were found to be in the coarse size fraction, indicating crustal contribution, Pb, Cd and V were found to be in the fine particles (<2.6 µm), indicating anthropogenic contribution. In addition to this, as can be seen in Table 1, there is a strong negative correlation between MMADs values and F/C values, change from 0.55 and 0.99. These results are similar to Karanasiou et al. [26] study results.

In many studies on air pollution, the heating and the non-heating seasons are differentiate [17, 21, 29, 30]. Domestic heating in densely populated residential areas is a significant source of air pollution in İstanbul. 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 metal concentration ratio of heating and non-heating season in three size fractions is shown Table 2.

Stations	PM Size (µm)	Cr	Ni	Pb	Cu	Cd	Mn	V	Fe
	<1	1.0	1.4	1.0	0.9	1.8	1.1	0.6	1.7
AVCILAR	1-2.6	1.3	2.6	1.9	0.5	1.5	0.9	0.7	0.6
	>2.6	1.1	1.7	0.6	0.5	1.5	1.0	0.5	0.7
BEŞİKTAŞ	<1	0.9	0.8	2.0	1.1	1.1	1.8	1.1	0.6
	1-2.6	1.5	0.9	2.3	1.0	1.1	1.4	1.2	0.7
	>2.6	1.3	0.5	1.2	1.1	0.8	1.4	1.1	0.8
GÖZTEPE	<1	1.2	2.2	3.3	1.9	2.0	1.4	1.8	1.5
	1-2.6	1.1	1.5	3.1	1.3	1.3	1.1	2.4	1.2
	>2.6	1.1	1.0	1.3	1.2	0.8	1.1	1.6	1.0
RASATHANE	<1	0.8	0.8	1.7	1.0	1.0	1.8	0.8	0.9
	1-2.6	1.0	0.7	1.2	0.8	0.8	1.3	1.1	0.9
	>2.6	0.9	0.9	0.9	0.4	0.6	1.0	1.4	0.7
KİLYOS	<1	2.4	0.8	1.8	1.1	2.7	1.3	1.0	1.0
	1-2.6	1.5	1.1	1.8	1.0	2.1	0.7	1.1	1.0
	>2.6	1.5	1.3	2.8	0.6	2.6	0.8	1.2	0.9

Table 2. The metal concentration ratio of heating and non-heating season in three size fractions ($<1\mu$ m, 1-2.6 μ m, $>2.6 \mu$ m).



We observed that Pb, Ni, Cd and V elements which are sourced from combustion, were higher in fine dust in heating season at Göztepe and Beşiktaş. Whereas, there was an increase of the Ni, Pb and Cd concentrations (between 1.4 and 2.6) in fine dust at Avcılar. The metal concentrations at Avcılar were higher than the other stations. We observed that there is no significant difference between heating and non-heating periods at Avcılar. It should be possible that the industrial emissions are the dominant source at Avcılar. There is no significant difference between heating and non-heating and non-heating periods at Rasathane, but at this station, Mn and Pb in fine dust were found 1.8 times and 1.7 times more, respectively (Table 2). The lowest metal concentrations were found at Kilyos background station. In the heating period, Cr, Cd and Pb concentrations in fine dust was found 1.3–2.7 times higher than the concentrations in the non-heating periods at Kilyos. It should be possible that there is an air mass transport from the Blacksea. Koçak *et al.* [19] found that the air transport affects the element concentration of particle in Istanbul.

4 Conclusions

This study was conducted over two years at five different sampling locations that characterized the variations metals in eight particle size fractions in Istanbul. Generally, the highest anthropogenic metals (Pb, Ni, Cd and V) concentrations were measured in the fine fraction of particles at all stations in the heating periods. The lowest metals concentrations were measured at the Kilvos, which is in a rural area in Istanbul. Determining the overall air quality in İstanbul is quite challenging. It is a large metropolitan city and it is the center of Turkish industry, with a population of nearly 13 million. In addition, it is a city with regional differences in terms of geographical and meteorological conditions. Furthermore, the transport of pollutants into the city atmosphere from Europe and Asia accounts for a significant portion of the regional PM pollution. The physical and chemical properties of the PM samples represented the diversity of pollution sources in the city atmosphere but did not represent any specific sources. More comprehensive studies are required with more regional attributions and continuous measurements to fully evaluate the air quality of the city. In addition, it is necessary to take other constituents of particles, especially in the fine particles, into consideration to determine the overall air quality and its sources in cities.

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