

Mass Concentrations and Elemental Compositions of PM₁₀ in Duzce, Turkey

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In this study, mass concentrations and elemental compositions of PM₁₀ were determined in the Duzce atmosphere. The Duzce province is located in north-western of Turkey. Two sampling campaigns were performed between 28 January and 25 March 2015 and between 22 June 2015 to 16 August 2015 to evaluate differences between summer and winter concentrations of pollutants. Concurrent measurements were carried out in the city center and the University Campus that has semi-urban property. PM₁₀ sampling was performed by high-volume air samplers. Average PM₁₀ mass concentration measured at urban station was 85.67 µg/m³ and 53.16 µg/m³ respectively winter and summer, while it was measured as 27.13 µg/m³ and 34.70 µg/m³ at semi-urban station. Seasonal variations of PM₁₀ concentration in the urban station showed significant differences in winter than in summer. Higher PM₁₀ concentrations were measured at an urban location often exceeding the limit values of 50 µg/m³ in winter. In this study the relationship between particulate matter concentration and meteorological variables such as wind direction, wind speed and rainfall was investigated and it was found that meteorological factors have significant effects on the measured particulate matter (PM) mass concentrations. PM mass collected on quartz fiber filters were digested in acid mixture and analyzed with ICP-MS. Concentrations of elements, Al, Fe, Sc, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Mo, Ag, Cd, Sn, Sb, Ba, Pb and Bi were determined in daily collected filters. Trace element concentrations measured at an urban station which is close to major sources such as traffic and domestic heating were found higher than semi-urban station.

Keywords: Air pollution, High volume air sampling, PM₁₀, Trace element, Turkey.

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Introduction

Particulate matters which have adverse effects on climate, human and environmental health are partly formed by the emission of anthropogenic or natural sources to the air and partly by condensation or nucleation of primer gases (Dongarra et al., 2010; Cheng et al., 2015). They are a complex mixture composed of liquid or/and solid, organic and inorganic materials (Aldabe et al., 2011). Suspended particles are classified as PM₁₀, PM_{2.5} or ultrafine particles depending on their aerodynamic diameters. Fine particles are generally released to the atmosphere from anthropogenic sources, whereas the coarse particles are released from natural sources (Hueglin et al., 2005; Romanazzi et al., 2014)

The determination of particulate matter size distribution and chemical content is important in terms of understanding their source, formation, transportation, and toxicity (Tian et al., 2015). The effects of particles on health differ depending on their size and chemical contents. Particles with a diameter less than 10 µm (PM₁₀) or 2.5 µm (PM_{2.5}) are considered to be especially dangerous as they may cause respiratory system disorders and increase the rates of death (Monn et al., 1997; Querol et al., 2001; Brunekreef and Holgate, 2002; Pekey et al., 2010; Wiseman et al., 2014). Particles comprise a complex mixture that contains toxic metals and persistent organic compounds such as polycyclic aromatic hydrocarbons (PAH) and dioxins are considered to be particularly dangerous (Dallarosa et al., 2008). There are many studies conducted on particulate air pollution as respirable particles have a significant impact on the environmental and human health (Dongarra et al., 2007; Toledo et al. 2008; Cheng et al., 2015). During these studies, it is especially necessary to conduct detailed researches regarding the chemical properties of atmospheric PM in order to determine the toxicity of the particles (Hueglin et al., 2005).

Diffusion and transportation of the atmospheric particles change too, depending on their sources, sizes, meteorological variables and topographic properties of the region. Therefore, the size, concentration and composition of the atmospheric particulate matters show significant differences in terms of time and location. Thus, air pollution studies are exclusive to the regions. In order to control the air pollution in a given area, air pollution monitoring studies should be conducted for the particular area. To this respect, PM₁₀ samples have been taken simultaneously from two points in Duzce, one of which is an urban location and the other, a semi-urban location. As a result, information on the level of the particulate air pollution in the given province has been obtained as well as the source and content of such pollution by determining the trace elements concentrations.

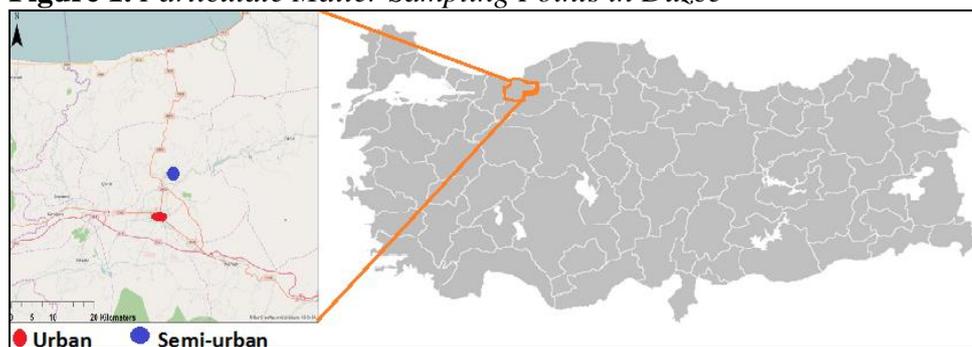
Materials and Methods

Study Area

Duzce is located at $40^{\circ} 49. 8'$ north latitude, $31^{\circ} 10. 2'$ east longitude, in the north-west of Turkey. The topography of the province is described as mountainous and hilly (approximately 85%). The surface area of the province is $2,574 \text{ km}^2$, and there are 355,545 inhabitants residing in the province. The population is rather low compared to the province's surface area. The dominant flow of the wind in the downtown of Duzce is prevented by the mountains and hills circumventing the province. Therefore, pollutants cannot distribute in the downtown of the province and exposition takes place at higher levels. The weather in Duzce is misty for approximately six months from October to March due to temperature inversion. Low inversion layer can be observed over the downtown both in winter and summer.

Particulate matter samples (PM_{10}) have been taken simultaneously in Duzce University Vocational School located in the downtown (at $40^{\circ} 50. 703'$ north latitude, $31^{\circ} 08. 913'$ east longitude) and in Konuralp Campus of Duzce University which can be classified as a semi-urban location (at $40^{\circ} 54. 170'$ north latitude, $31^{\circ} 10. 529'$ east longitude). The locations of the sampling points are shown in Figure 1. The selection of the sampling points was made in consideration of the possible sources (traffic, industrial facilities, domestic heating) and a comparison was made by picking two points one of which is an urban location and the other, a semi-urban location. Urban sampling point is affected mainly by traffic and domestic heating emissions. However, the semi-urban sampling point is located in a relatively remote area of the city center and main roads. For this reason, it is expected that this point will be less affected by heating and traffic-related emissions.

Figure 1. Particulate Matter Sampling Points in Duzce



Sampling Period

The sampling of particulate matters (PM_{10}) was carried out by taking samples from each point in three consecutive days, and then taking a two days break and taking more samples in the following three days. Hereby, The number of samples taken during the sampling period is approximately the same

for each weekday and weekend day. Totally, 35 samples have been taken from each sampling point during the sampling period. Winter sampling was made between 28 January-25 March 2015 and summer sampling was made between 22 June-16 August 2015.

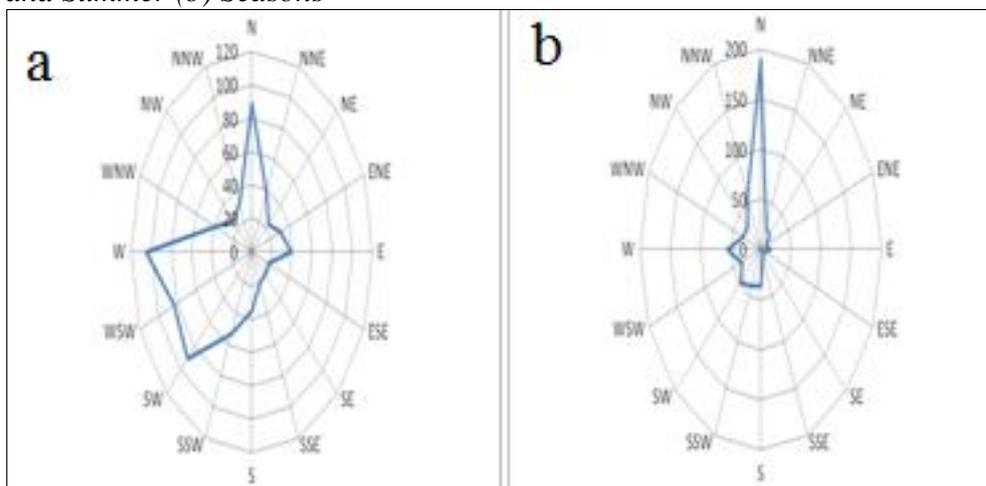
Meteorological Parameters

Meteorological parameters, which are determined during the sampling periods, are given in Table 1. These data are obtained from the Meteorology Directorate of Duzce. The prevailing wind directions were determined to be north, east, south-east for winter and north sectors for summer sampling period (Figure 2).

Table 1. Meteorological Parameters for Sampling Periods

Sampling Campaign	Average Relative Humidity (%)	Average Wind Speed (m/s)	Maximum Wind Speed (m/s)	Temperature (C°)	Average Rainfall (mm)	Total Rainfall (mm)	Mixing Height (m)	Pressure (hPa)
28.01-26.03/2015	76.4	1.08	5.4	7.2	3.1	109	404	1000
22.07-16.08/2015	68.5	1.23	6.5	24	1.6	54.3	654	995.5

Figure 2. Wind Rose Graphic of the Sampling Period during the Winter (a) and Summer (b) Seasons



Preparation and Collection of Filters

In this study, Thermo brand high volume air samplers are used. PM₁₀ samples were collected on quartz filters. The sampling duration was 24 hours and air flow rate during the sampling was regulated to 1.13 m³/min. Before and after sampling, the filters were stored in desiccators for 24 h prior to an initial weighing using a precision balance under a controlled temperature and relative humidity conditions. After the sampling, the quartz filters are carried into the laboratories in a suitable condition and kept for 24 hours in desiccators, and

their final weight has been measured. Before the analysis, the samples are divided into four equal parts and one piece has been spared for metal analysis.

Extraction and Analysis

Filter which is cut for the extraction is weighed (~1 g) and divided into smaller parts, and transferred into polytetrafluoroethylene (PTFE) caps, 4 ml of Hydrofluoric acid (HF), 2 ml of Hydrogen peroxide (H₂O₂) and 8 ml of Nitric acid (HNO₃) added on them and held for 20 minutes, then extracted in 1200 W microwave oven for 20 minutes in 200°C. Samples taken out from the microwave oven are cooled for 45 minutes and in order to reach a %2 HNO₃ concentration, ultra-pure water has been added until 50 ml, and the solution became ready for analysis. These phases are applied to the blank samples in the same manner. The element concentrations of the PM₁₀ samples were determined via inductively coupled plasma mass spectrometer (ICP-MS) (Thermo Scientific X Series 2). Quality control and quality assurance data for elemental analyses are given in Table 2.

Table 2. *Quality Control and Quality Assurance Data for Elemental Analysis*

Element	SRM (1648-A) Conc. ^a (ppb)	Exp. Conc. ^b (ppb)	Blank Conc. ^c (ppb)	MDL ^d (ppb)	MQL ^e (ppb)	RCV ^f (%)	RPT ^g (%)
Al	34300	33990	0.180	3	1	100.91	99.84
Fe	39200	37980	0.293	3	1	103.21	99.95
Sc		11500	0.000	84	28		99.97
V	127000	117100	0.001	12	4	108.45	99.98
Cr	402000	402600	0.022	297	99	99.85	98.92
Mn	790000	796000	0.006	609	203	99.25	99.95
Co	17930	16500	0.000	3	1	105.04	99.83
Ni	81100	88500	0.016	150	50	91.64	99.54
Cu	610000	648500	0.006	237	79	94.06	99.04
Zn	4800000	4725000	0.006	1527	509	101.59	99.89
As	115500	110400	0.001	195	65	104.61	99.77
Se		65000	0.000	966	322		99.70
Mo		191100	0.006	726	242		99.89
Ag		517900	0.004	3	1		99.33
Cd	73700	72600	0.000	3	1	101.51	99.46
Sn		7192000	0.043	105	35		99.86
Sb	45400	43200	0.003	117	39	105.09	98.32
Ba		327700	0.007	477	159		99.78
Pb	6550	6736	0.023	351	117	97.24	99.69
Bi		208700	0.001	3	1		97.44

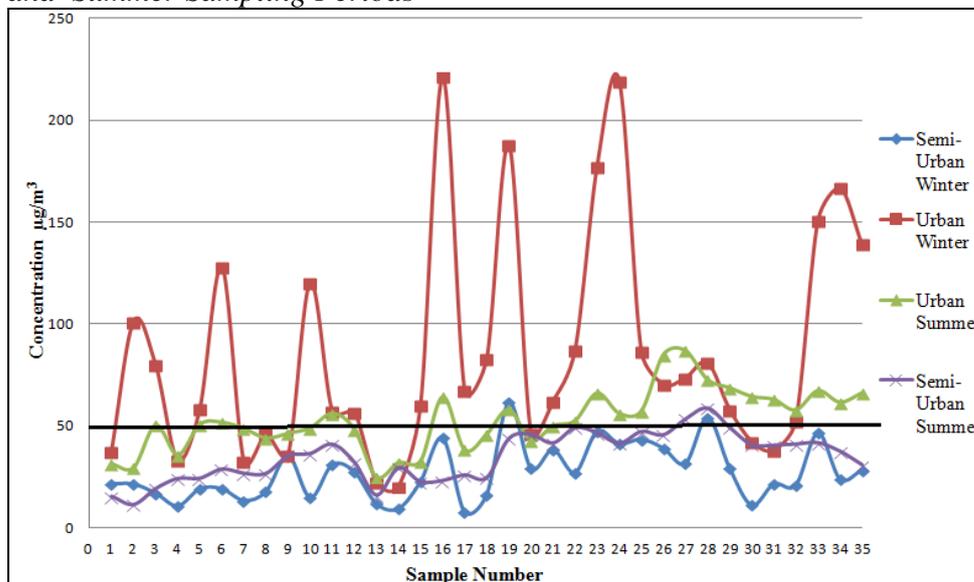
^aSRM (1648-A) Conc. standard reference materials, SRM 1648-A Concentration ^bExp. Conc Experimental Concentration, ^cBlank Conc. Blank Concentration, ^dMDL Method Detection Limit, ^eMQL Method Qualification Limit, ^fRCV Recovery, ^gRPT Repeatability

Results and Discussion

Particulate Matter (PM_{10}) Concentrations

Particulate matter concentrations measured during the winter and summer sampling periods are given in Figure 3. When Figure 3 is evaluated, it is observed that the daily PM_{10} values reach critical levels during the winter sampling period in an urban sampling point which is near the traffic flow, and it is noted that as a result of the 25 days in 35 day sampling period, the daily values exceed $50 \mu\text{g}/\text{m}^3$ which is the daily limit value set by EU regulations. This limit value is defined under the relevant regulation as the limit which should not be exceeded for more than 35 times in a year. On the other hand, the values obtained from the semi-urban sampling point are below the limit values except for only two days. The average mass of PM_{10} concentrations calculated in the urban sampling point during the winter sampling period is $85.67 \mu\text{g}/\text{m}^3$ whereas the average PM_{10} in semi-urban sampling point is determined as $27.13 \mu\text{g}/\text{m}^3$. When Figure 3 is evaluated in terms of summer sampling period, it is observed that the PM_{10} mass concentrations are below than those observed in winter sampling period. However, the limit value of $50 \mu\text{g}/\text{m}^3$ was exceeded in the summer sampling period in most of the days as is the case with the winter sampling period in the urban sampling point. The average mass concentrations of PM_{10} is determined during the summer sampling period $53.16 \mu\text{g}/\text{m}^3$ in the urban station whereas $34.70 \mu\text{g}/\text{m}^3$ in the semi-urban station. The limit value has been exceeded in 19 days in urban sampling point as the limit value is exceeded only for one day in semi-urban sampling point.

Figure 3. Measured Concentrations of Particulate Matter during the Winter and Summer Sampling Periods



The Effect of Meteorological Factors on Particulate Matters

It is determined that the particulate matter concentrations are interacting with the meteorological variables such as the wind speed, rain, wind direction, vertical mixture height and ventilation coefficient. The relationship between the particulate matter concentration and the meteorological parameters is shown in Table 3 by determining the Pearson correlation coefficients. Among the most important effects of wind speed are to carry PM away from the sources, to determine the mixture height, and to dilute the pollutants. On the other hand, high wind speeds increase the rate of PM recirculation from the soil surface and roads. At low speeds, concentrations can reach significant levels because they cannot move away from PM sources. According to the Table 3, particulate matter concentration shows a negative correlation with wind speed in winter sampling period and hence as the wind speed increases, the particulate matter concentration decreases. This is probably the result of high speeds increasing the dilution effect. In the summer sample, no meaningful correlation was found between wind speed and pollutant concentrations. The reason for this situation can be explained as the effect of PM transport from the soil is greater than the dilution effect. A similar relationship, although weaker, has been observed in rainy weather and the particulate matter concentration is observed to be around $22.59 \pm 8.28 \mu\text{g}/\text{m}^3$ in winter sampling period whereas it is determined to be $27.59 \pm 10.93 \mu\text{g}/\text{m}^3$ in summer sampling period during the rainy weather in semi-urban location. These values are determined to be around $64.92 \pm 15.94 \mu\text{g}/\text{m}^3$ in the winter sampling period and $49.54 \pm 14.63 \mu\text{g}/\text{m}^3$ in the summer sampling period in an urban location. In rainless days, the average particulate matter concentrations in semi-urban locations are determined as $30.96 \pm 31.48 \mu\text{g}/\text{m}^3$ and $37.95 \pm 10.79 \mu\text{g}/\text{m}^3$ during the winter and summer sampling periods, respectively. In the urban sampling point, this value is determined to be $103.14 \pm 65.27 \mu\text{g}/\text{m}^3$ in the winter sampling period and $54.82 \pm 14.78 \mu\text{g}/\text{m}^3$ in the summer sampling period. The most important reason for the reduction of PM concentrations by rainfall is the removal of particles from the atmosphere by washing. Also, the wetting of the soil with rainfall causes the reduction of air mixing of the particles with the wind effect. According to Table 3, there is a weak positive correlation between the particulate matter concentration and temperature. Especially, more meaningful values are observed in the summer sampling. This correlation can be explained based on the following; as the temperature increases, the dried soil can circulate better and therefore, the particulate matter increases in the air. When the correlation between the PM_{10} mass concentrations with the vertical mixture height is evaluated, it is observed that there is a negative correlation between these in the urban sampling point in the winter sampling period. To this regard, it is observed that as the height of the mixture decreases, the pollutant concentration increases. Another parameter is the ventilation coefficient which is one of the parameters used in the literature for reviewing the changes in pollutant concentrations. Ventilation coefficient is achieved by multiplying the mixture height with the wind speed (Krishnan and Kunhikrishnan, 2004). In

our study, it is found that there is a meaningful relationship between the ventilation coefficient and PM₁₀ concentrations only in urban sampling point. According to results, the PM concentration decreases as the ventilation coefficient increases. This is due to the dilution of the atmospheric PM concentrations by increasing the wind speed and mixture height parameters, which are elements of the ventilation coefficient.

Wind direction is a meteorological factor affecting the pollutant concentrations and types. As can be seen in Figure 2, the dominant wind sectors during the winter sampling period are N, W, and SW. When we consider the location of the sampling points in the city, the main roads where there is a major traffic flow is on the north side of the points whereas the residences are majorly on the west side of the points. When we consider the wind direction, the pollutants have an impact on both of the sampling points due to the domestic heating and the traffic activity in winter. When the wind rose graphic in summer sampling point is evaluated, almost the only dominant wind direction is N sector. Thus, we can say that the traffic activity had a continuous impact throughout the sampling period.

Table 3. *The Relationship between Meteorological Factors and the Concentration of PM₁₀*

		MWS ^a	AWS ^b	Rainfall	T ^c	MH ^d	VC ^e	Urban
PM₁₀ WINTER	Semi-Urban	-0.61**	-0.48**	-0.21	0.22	0.01	-0.29	0.58**
	Urban	-0.51**	-0.47**	-0.23	0.36*	-0.36*	-0.55**	1
PM₁₀ SUMMER.	Semi-Urban	0.00	0.03	-0.34*	0.73**	0.06	0.04	0.74**
	Urban	0.07	0.05	-0.19	0.52**	-0.01	-0.03	1

** P < 0.01, * P < 0.05

^a Diurnal Maximum wind speed, ^b Diurnal Average wind speed, ^c Temperature, ^d Mixing Height, ^e Ventilation Coefficient

Elemental Concentrations

Statistical results for elemental concentrations measured in urban and semi-urban points in winter and summer periods are seen in Figure 4. When Figure 4 is evaluated, it is observed that the elemental concentrations are generally higher in urban points than those determined in semi-urban points. This is due to the closeness of the downtown to sources, the low altitude (150 m in urban sampling point, and 230 m in semi-urban sampling point) and the pollutants not being able to distribute due to the location being mountainous and hilly. When the trace element content is analyzed, the highest concentrations for the winter are found to be Al and Fe and the lowest concentrations are found to be Ag, Bi, Cd, Co, Se. The highest concentrations are found to be Al and Fe and the lowest concentrations are found to be Sc, Co, Bi, Ag, Cd for the summer, as in the winter. The rates of the concentrations for urban and semi-urban points are calculated for each point separately and given in Figure 4 as an average value. When the rates are evaluated, the highest rate

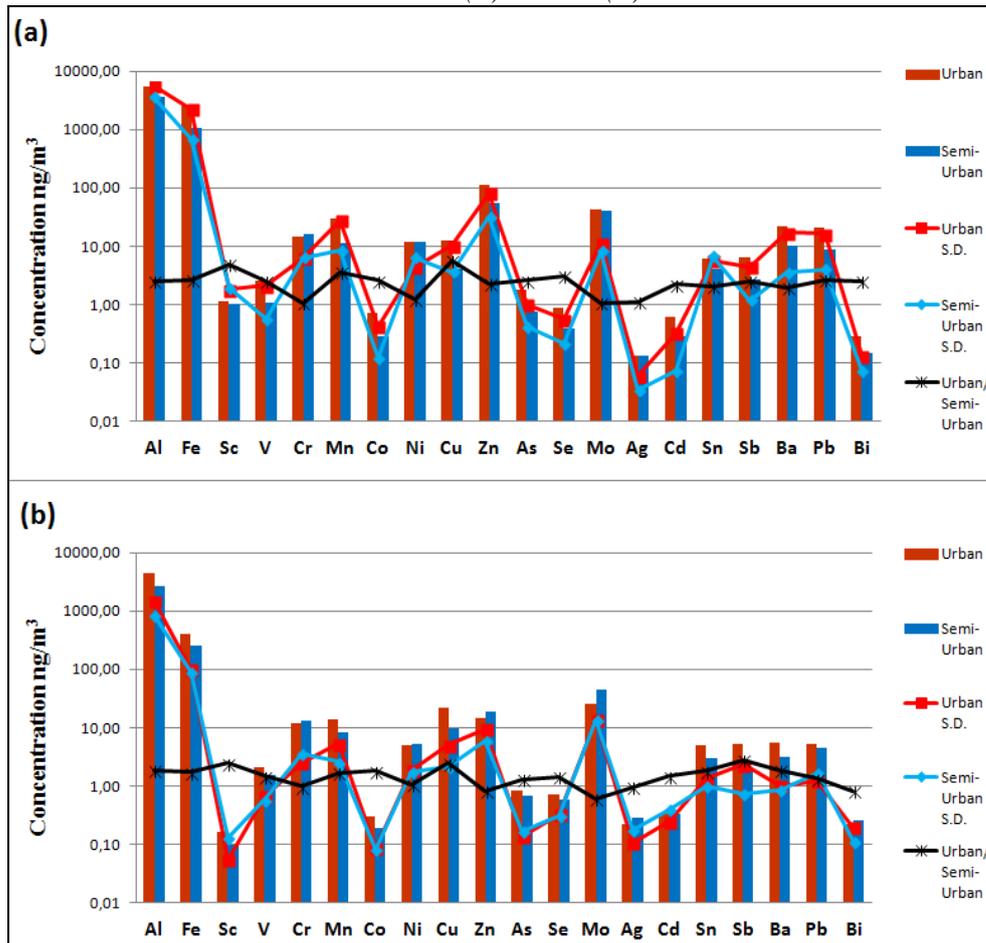
for the winter contains Cu, Sc, Mn, Sb, and Co elements whereas for the summer, Sc, Sb, Cu, Al, Ba, Sn and Co elements were found to be dominant. These elements are among the traffic and heating based elements which are among the most significant sources for a city center (Baez et al., 2015; Manousakas et al., 2015).

The relationship between the elements which are evaluated within the scope of this study is analyzed by Pearson correlation analysis. It is possible to think that the pollutants which have meaningful correlation have similar common sources and are held subject to similar atmospheric conditions. When the relationship between the anthropogenic elements in the urban sampling point in winter is evaluated, it is observed that V, Mn, and Co elements related with each other on a bilateral basis (Pearson correlation coefficients are 0.6-0.7 $p < 0.01$). These elements come out as a result of oil combustion (Bove et al., 2016; Hsu et al., 2016; Qi et al., 2016). A similar situation is observed for Sb, Mn, Zn, Cu, Ba, Pb elements. The Pearson correlation coefficients of these elements ($p < 0.01$) varying between 0.5 and 0.7 are calculated. These elements are related to vehicle emissions (Liu et al., 2014). To this respect, we may think that these elements emit from a common source which may be the traffic activity. Another relation is observed between Co, As, Se, Cd, Sn and Sb elements (0.5-0.7 $p < 0.01$). These elements also come out as a result of coal combustion (Qi et al., 2016, Hsu et al., 2016). This source may be meaningful wherefore coal is still used for heating purposes in the some residential or industrial areas in Duzce. The correlation coefficient between Al and Fe, which are among the soil based elements, is calculated as 0.8 ($p < 0.01$). Furthermore, a meaningful relation between the mentioned elements and the traffic based Mn, Cu, Zn, Sb, Ba and Pb elements (0.5-0.9 $p < 0.01$) is observed. This may be due to the re-suspension of the polluted soil (road dust) or the vehicle emissions. Particularly, friction and abrasion in the metal components of the vehicles cause Fe emissions (Bernardoni et al., 2011; Minguillon, 2014).

When the interactions between the anthropogenic elements are evaluated, generally lower coefficients are calculated in semi-urban sampling point compared to the calculations observed in urban sampling point. In addition, similar relationship patterns are observed in both of the sampling points. The relation between V, Mn, Co elements, between Sb, Mn, Zn, Cu, Ba, Pb elements and between Co, As, Se, Cd, Sn, Sb elements in the semi-urban sampling point is observed to be similar (0.4-0.6 $p < 0.01$) as it is observed in urban sampling point. The relation between Al and Fe elements are found to be similar to the observation made in urban sampling point (0.4 $p < 0.01$). However, the interaction observed between the traffic based and soil based elements in urban sampling point was not observed in semi-urban sampling point, especially regarding Al element. However, in related iron elements, weaker interaction is observed between V, Mn, Zn, Mo, and Ba elements in semi-urban sampling point compared to the observations made in urban sampling point. This may be because of the relatively long distance of the sampling point to the traffic activities.

Almost all of the anthropogenic elements show a meaningful and powerful interaction among themselves in urban sampling points in the summer period, compared to the observations made in the winter period. Therefore, the main source of pollution for the elements observed in the summer sampling period may be thought as the traffic and hence the correlation coefficients increased. Less interaction is observed between the elements in semi-urban sampling point since the sampling location is relatively farther from the traffic activities. Both Al and Fe elements reflect meaningful interaction both among themselves and with traffic based elements.

Figure 4. Mean, Standard Deviation and Urban/semi-urban Rates of Measured Element Concentrations (a) Winter (b) Summer



Conclusions

When the results of the study are evaluated, it is observed that the PM_{10} mass concentrations are higher in winter compared to summer in urban sampling point and close to each other in semi-urban sampling point. For elemental concentrations, the observed values are higher in winter compared to

summer in both of the sampling points. Furthermore, seasonal changes are more significant in urban sampling point. Almost all rates for urban to semi-urban PM₁₀ mass and elemental concentrations are found to be more than 1 for both of the seasons. The reason why the urban concentrations are higher compared to semi-urban concentrations and the seasonal changes are more significant in urban sampling point is because the urban sampling point is closer to pollution sources. However, the reason for the significance of the seasonal change in the urban sampling point is not merely due to the pollution sources. This is also due to the meteorological factors causing elevation of pollutant concentrations and the pollutants not being able to distribute due to the topography of the location. PM₁₀ mass concentrations have been found lower in rainy days due to the decrease of airborne particles. Also, it is determined that the wind speed and direction are also effective on concentrations. Based on the height of the vertical mix and the decrease of the ventilation coefficient, the particulate matter concentrations have been found higher in winter compared to summer. The relationship between the elements is determined by correlation coefficients. According to the correlation analyses, the elemental sources for urban and semi-urban sampling points were identified as combustion and traffic.

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