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Trace Metal Concentrations in Size-Fractionated Urban Atmospheric Particles of Sarajevo, Bosnia and Herzegovina

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ABSTRACT: A study of the atmospheric particulate size distribution of total suspended particulate matter (TSPM) and associated heavy metals concentrations has been carried out for the urban part of Sarajevo city, Bosnia and Herzegovina. Urban particles (n=150) were collected using a high volume air sampler equipped with a 6-stage impactor. Apart from Fe which has been determined by flame atomic absorption spectrometry (FAAS), all measurements (Co, Cu, Mn) have been carried out by graphite furnace atomic absorption spectrometry (GFAAS). The average concentrations of particulate matter are 37%, 18%, 15%, 8%, 15% and 6% (averaged over all the observations) of total suspended particulate for $PM_{< 0.49}$, $PM_{0.95 - 0.49}$, $PM_{1.5 -0.95}$, $PM_{3.0 - 1.5}$, $PM_{7.2 - 3.0}$ and $PM_{> 7.2}$, respectively. Metal concentrations in size-fractionated urban particles ranged from 0.01-3.83 ng/m^3 for Co, 6.30-179.20 ng/m^3 for Cu, 5.00-208.70 ng/m^3 for Mn and 0.35-6.82 g/m^3 for Fe. Major concentrations of investigated metals are associated with the PM 7.2-3.0 μm. Overall, the decreasing trend of average trace element concentrations (24-h) in the particulates revealed the following order: Fe>Mn>Cu>Co.

Key words: Size-fractionated particulate matter, Size distribution, Urban particles, Metals, Sarajevo

INTRODUCTION

Particulate matter (PM) is the general term used for a mixture of solid particles and liquid droplets found in the air. These particles come in a wide range of sizes and they are originated from natural sources as well as from many different anthropogenic sources (Buccolieri *et al.,* 2005). From a toxicological point of view, the most important particles are those with a diameter <10 μm (PM10), so-called respirable fraction, which penetrate the human respiratory system deeply. It is well established that fine particles (smaller than 2.5 μm) penetrate the pulmonary region and tend to deposit in alveoli (WHO, 2000) causing adverse health effects leading to pulmonary and respiratory diseases, outbreaks of cancer and even mortality rates (Dockery and Pope, 1994; Espinosa *et al.,* 2001; Shah *et al.,* 2006). Early studies were aimed at assessment of the general pollution levels in urban areas like estimation of the amount of the total suspendend particulate matter (TSPM), and determination of metal concentrations in it. Natural emissions (crustal minerals, forest fires and oceans) and industrial emissions (combustion of fossil fuel and industrial metallurgical processes) are the principal sources of heavy metals in the ambient air (Park *et al.,* 2008; Xue *et al.,* 2010; Cheung *et al.,* 2011). Traffic emissions also represent potential sources of trace heavy metals, including combustion products from fuel and oil, road construction materials, road dust and wear products from tires, brake linings and bearings (El- Fadel *et al.,* 2001; Kothai *et al.,* 2008). Depending on emission sources, rates of wet and dry deposition, and physical/chemical transformation, concentration and size distributions of metal atmospheric particles will vary. Long-range transport of atmospheric particles will also affect concentration and size distribution of metals (Xue *et al.,* 2010). Typically, particles in the accumulation mode have a long residence time and can be transported over a long distance affecting remote regions from sources.

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Therefore, size distribution of particles and associated metal concentration will provide information on the toxicity level of metals, as well as on transport behavior in the ambient atmosphere and on inhalation characteristics of the human respiratory system (Oberdörster, 2000; Buccolieri *et al.,* 2005).

The aim of this study was to determine the size distribution of TSPM and metal concentrations associated with each size fraction (> 7.2µm, 7.2 – 3.0 µm, 3.0 – 1.5 µm, 1.5 -0.95 µm, 0.95 – 0.49 µm and < 0.49 µm) of urban atmospheric particles of Sarajevo. Monthly variations of size-fractionated particles and trace metal levels in urban PM were also determined. Obtained results of PM and trace metals are also compared with the international guidelines as well as with the results from other sites around the world. In the context of urban environment in Bosnia and Herzegovina, there are no studies on size distribution of TSPM and associated metal concentrations.

MATERIALS & METHODS

Atmospheric particulate matter was collected from August to December 2008, in the city of Sarajevo using a high volume air sampling system equipped with a six-stage cascade impactor (Tisch Environmental, Inc). Sarajevo is the capital city and largest urban center of Bosnia and Herzegovina, with a population of about 700.000 people. It is situated around the Miljacka River and about 500 meters above sea level and lies in the Sarajevo valley, in the middle of the Dinaric Alps. Sarajevo has a mild continental climate, lying between the climates zones of Central Europe to the north and the Mediterranean to the south. The cascade impactor system was placed on the roof of the Department of Chemistry building (Faculty of Science, University of Sarajevo) at a height of 6 m from ground and 20 m from the nearest busy street intersection. Sources of anthropogenic emissions surrounding the sampling site include road side dust, combustion of fossil fuels and emis-sions from gasoline fueled road vehicles. The samples were collected on pre-weighed cellulose filters (Whatman 41) at a flow rate of ~1.16/m^3/min. The particle size range collected from impactor stages are >7.2, 7.2–3.0, 3.0–1.5, 1.5–0.95, 0.95–0.49 and < 0.49 ìm. A total of 25 sets were sampled. Meteorological conditions at the sampling site are given in Table 1. Filters were kept in a nitrogen glove box (AtmosBags, Aldrich) for 24 h before and 48 h after weighing in order to reach equilibrium with the laboratory environment and conditions in the box (Cincinelli *et al.,* 2007). Then, the filters were weighted three times per three days on an analytical balance (± 0.01 mg) (Ohaus, model Discovery DV214C, Switzerland) at a constant relative humidity of 46 ± 3 %.

Atmospheric particles collected on cellulose filters were digested in the microwave oven (Anton Paar, model 3000, Austria) with 6 mL nitric acid, 3 mL hydrogen peroxide and 3 mL hydrofluoric acid (Buccolieri, 2005). PM concentrations were determined by gravimetric analysis using the weight difference of the filters before and after the sampling procedure. Trace metal analyses of PM samples were performed by atomic absorption spectroscopy using a graphite furnace (Varian AA240Z) for Co, Cu and Mn, and flame for Fe (Varian AA240FS). All samples were analyzed in triplicate. A series of blanks were prepared using the same digestion method to avoid the matrix effect. High purity reagent-grade (s.p.) chemicals obtained from Merck were used for all procedures. The accuracy of the method was evaluated using the standard reference materials: Fine Fly Ash (CTA–FFA–1, Institute of Nuclear Chemistry and Technology, Poland) and Fly Ash from pulverised coal (BCR-038, Institute of reference materials and measurements- IRMM, Belgium). The obtained results were in the range of the reference materials. The limits of detection (LODs) were determined as three times the standard deviation of the procedural blank and were found to be 0.008, 2.10, 28.5 and 1.47 ng/m^3 for Co, Cu, Fe and Mn, respectively.

Table 1. Summary of meteorological conditions during the sampling period (august-december 2008)

Meteorological parameters	August	September	October	November	December
Temperature (^{o}C)	22.8 - 38.0	10.0 – 31.0	15.0 - 26.0	-4.0 – 20.2	4.0 -13.0
Pressure (Pa)	1003 - 1012	1009 - 1020	1013 - 1026	1003 - 1023	1009 - 1020
Rainfall (mm)	0	80.9	1.3	10.3	8.0
Wind direction	E, SE	E, N	E, SE	E, W	E, SE
Wind speed (m/s)	5.5 – 10.9	5.5 – 16.4	5.5 – 16.4	5.5 – 13.6	2.7 – 24.5

Table 2. The average, maximum and minimum concentrations of size-fractioned urban atmospheric particles of Sarajevo, collected from August to December 2008

Concentration of PM (μg/m^3)							
n = 150		> 7.2μm	7.2 – 3.0 μm	3.0 – 1.5 μm	1.5 -0.95 μm	0.95 – 0.49 μm	< 0.49 μm
August	Min	0.1	3.4	0.4	0.6	1.3	8.3
	Max	2.7	4.6	2.7	3.3	3.5	10.6
	Avg	1.5	3.8	1.7	2.2	2.6	9.2
September	Min	0.4	0.6	0.6	1.7	1.8	6.4
	Max	9.1	4.0	3.9	7.9	6.2	10.8
	Avg	2.9	2.8	1.8	4.0	4.0	9.4
October	Min	1.1	4.2	2.6	4.3	4.9	8.4
	Max	2.4	6.9	4.6	7.0	8.4	12.0
	Avg	1.8	5.5	3.2	5.7	7.1	10.3
November	Min	0.1	1.8	0.7	1.3	2.0	1.8
	Max	1.9	9.1	4.9	10.3	16.4	36.9
	Avg	1.0	4.9	2.4	5.2	7.1	13.3
December	Min	0.8	3.0	1.9	3.5	2.6	7.7
	Max	2.5	4.9	3.2	6.3	8.7	13.5
	Avg	1.4	4.2	2.5	4.6	5.3	10.3

RESULTS & DISCUSSION

The concentrations of atmospheric particulate matter in six size fractions and associated metals were determined in the urban part of Sarajevo city. Table 2 shows the average, maximum and minimum concentrations of size-fractionated urban atmospheric particles, collected from August to December 2008. From the results obtained in this study, it was observed that the highest average concentration of PM was obtained in fraction <0.49 μm with the value of 13.3 μg/m^3. Lowest average level of PM i.e. 1.0 μg/m^3 was obtained in fraction >7.2 μm. From the minimum and maximum values in Table 3 it was observed that the concentrations of PM varies most in fraction <0.49 μm, from 1.8 μg/m^3 to 36.9 μg/m^3 in November. For the other fractions the concentration of PM varies between 0.1-9.1μg/m^3, 0.6-9.1 μg/m^3, 0.4-4.9 μg/m^3, 0.6-10.3 μg/m^3 and 1.3-16.4 μg/m^3 for fractions > 7.2μm, 7.2 – 3.0 μm, 3.0 – 1.5 μm, 1.5 -0.95 μm and 0.95 – 0.49 μm, respectively. For each month, except August, the highest average concentrations of PM were observed in fraction <0.49 μm followed by fraction 0.95-0.49 μm. Apart from the fractions >7.2 μm and 7.2-3.0 μm, all other fraction had the lowest average PM levels in August. One of fine particle size, 0.95-0.49 μm, 1.5-0.95 μm and 3.0-1.5 μm (alveo-bronchinal and bronchinal regions) had the highest average PM concentrations in October. Maximum concentrations of this fine particle size were observed in November with the values of 16.4 μg/m^3, 10.3 μg/m^3 and 4.9 μg/m^3 for particle size 0.95-0.49 μm, 1.5-0.95 μm and 3.0-1.5 μm, respectively. The average concentrations of particulate matter are 37%, 18%, 15%, 8%, 15% and 6% (averaged over all the observations) of total suspended particulate for $PM_{< 0.49}$, $PM_{0.95 – 0.49}$, $PM_{1.5 -0.95}$, $PM_{3.0 – 1.5}$, $PM_{7.2 – 3.0}$ and $PM_{> 7.2}$, respectively.

As shown in Table 1, highest variations in air pressure and temperature appear in November. Unstable weather conditions are the cause of transferring and flow of particles from afar by wich the wet deposition of particles can lead to higher concentration of particulate matter on the local character (Pillai *et al.,* 2002; Glavas *et al.,* 2008). During the winter month of November, more stagnation and less rain scavenging occurs, causing aerosol particles to remain suspended over the city for an extended period of time. Also, the lower solar insulation rates during the winter months lead to lower atmospheric inversion layers where pollutants become trapped close to the ground, further increasing particle concentrations (Gummeneni *et al.,* 2011).

Based on the obtained results it can be said that there is a similar trend of metal distribution per fractions of particulate matter (Fig. 1). Among the analyzed metal

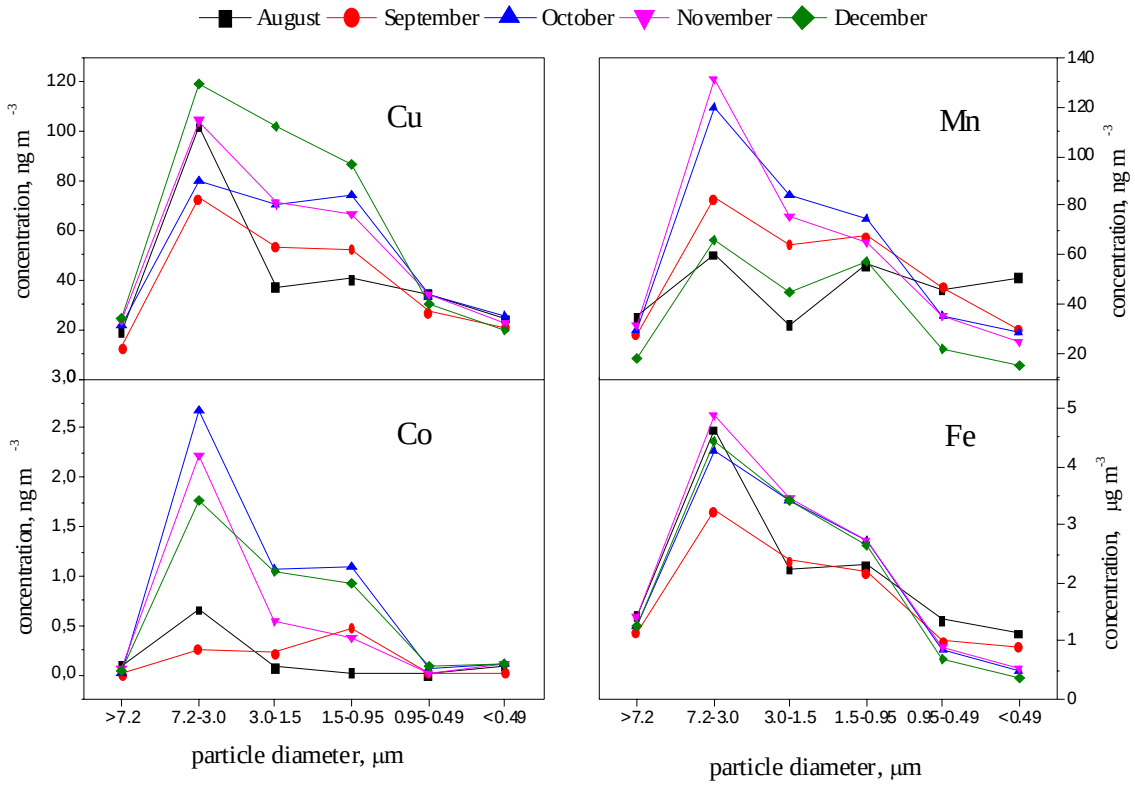

Fig. 1. Mean metal concentration in size fractinated PM for each month

concentrations, Fe showed the highest concentration and Co the lowest one. Maximum concentrations of all analyzed metals are found in the PM 7.2 –3.0 μm with the values of 3.83 ng/m^3 for Co, 179.20 ng/m^3 for Cu, 6.82 μg/m^3 for Fe and 208.70 ng/m^3 for Mn. Also imortant to note that highest variations in metal concentrations of particulate matter appear in fraction PM 7.2 –3.0 μm, then slightly less in fractions PM 3.0 –1.5 μm and PM 1.5 –0.95 μm. This could be the reason of various emission sourses as well as winds of moderate speed which could bring a good amount of dust from the adjoining areas of Sarajevo. Another finding is that fractions PM >7.2, PM 0.95-0.49 μm and PM < 0.49 μm at least contribute to the total mass concentration of Co, Cu, Fe and Mn. Furthemore, these three fractions show to be almost constant during the whole investigation period. Minimum concentration of 0.01 ng/m^3, 6.30 ng/m^3, 0.35 μg/m^3 and 5.00 ng/m^3 were recorded for Co, Cu, Fe and Mn, respectively. During the sampling period winds of moderate speed were mainly observed. This fact in conjunction with the Sarajevo topography leads to poor ventilation of the valley and indicates the significance of local sources rather than long-range transport. Combustion of fossil fuels and emis-sions from gasoline fueled road vehicles as well as road dust are the principal anthropogenic source of metals in the urban air particulate matter of Sarajevo. Considering the above and the traffic density at the sampling site we focus the discussion on specific source categories such as vehicle emissions, road dust, industry and soil. To compare obtained results with previously published results of TSPM, PM_{10} and PM2.5 and heavy metal concentration (Co, Cu, Fe and Mn) in TSPM a new classification of results was used (Table 3).

The results of TSPM concentrations obtained for 6 size fractions were grouped into $PM_{3.0}$ (the sum for first 4 separated fractions: < 3.0 μm) and PM_{10} (the sum for 5 fractions: < 7.2 μm). The 24–h TSPM, PM_{10} and $PM_{3.0}$ levels in urban particulate matter appear in the range 9.9-77.1 ng/m^3, 9.5-75.2 ng/m^3 and 8.3-67.3 ng/m^3, respectively. Among the trace elements in atmospheric particulate samples, major contribution was noted for Fe. Overall, the decreasing trend of average trace element concentrations (24-h) in the particulates revealed the following order: Fe>Mn>Cu>Co. The present data on atmospheric particulate matter and trace elements are compared with the international guidelines as well as with the

Table 3. Statistical summary of TSPM, PM_{10} and PM_3 (µg/m³) and element concentrations (ng/m³) in TSPM and their comparison with other urban areas around the world and guidelines of regulatory agencies

		Particulate Matter (µg/m³)			Metal concentration (ng/m³) in TSPM			
		$PM_{3.0}$	PM_{10}	TSP	Co	Cu	Fe	Mn
August	Min	12.9	16.3	16.4	0.45	227.7	10 140	167.9
	Max	17.8	22.4	25.1	1.45	313.6	16 850	395.5
	Avg	15.6	19.4	20.8	0.95	257.0	13 097	280.6
September	Min	8.3	9.5	9.9	0.73	139.2	5 830	141.8
	Max	23.6	27.6	30.6	1.29	426.6	18 390	604.7
	Avg	15.2	17.9	20.8	1.01	230.1	10 493	307.5
October	Min	21.6	26.2	27.6	4.28	223.8	9 920	255.3
	Max	31.0	37.9	40.3	5.75	392.8	17 630	481.3
	Avg	26.3	31.7	33.5	5.02	305.1	12 970	373.3
November	Min	10.7	13.6	14.0	0.75	210.7	5 530	100.7
	Max	67.3	75.2	77.1	5.94	504.5	21 830	601.6
	Avg	27.9	32.9	33.9	3.35	321.6	13 865	363.7
December	Min	17.0	21.9	22.8	1.81	203.9	11 200	150.5
	Max	30.4	35.0	35.8	7.00	509.8	16 530	370.8
	Avg	22.6	26.8	28.2	4.02	382.0	12 788	223.9
Regulatory Agencies								
		$PM_{2.5}$	PM_{10}	TSP				
WHO (WHO, 2000)		25	50	80				150
USEPA (ATSDR, 2002)				60				500
Air Quality Regulations (AQG,(BiH), 2003)			50	150				2 000
Urban Areas								
Pristina, Kosovo (Arditsoglou and Samara, 2005)				109	6	179	1 725	49
Tito Scalo, Italy (Ragosta *et al.,* 2002)				60		58	521	27
Delft, Netherlands (Wang *et al.,* 2001)				56		139		
Athens, Greece (Valvanidis *et al.,* 2006)				133		700	10 200	
Bilbao, Spain (Aranguiz *et al.,* 2002)				171		200	4 400	260
Delhi, India (Shridhar *et al.,* 2010)				546		3 690	16 430	740
Yokohama, Japan (Khan *et al.,* 2010)				34.2		47.7		32.5
Bursa, Turkey (Samura *et al.,* 2003)							41 400	992
Santa Cruz, Brazil (Quiterio *et al.,* 2004)				87	1.1	335	38 903	1 216

counterpart data from other sites around the world as shown in Table 3. The 24-h TSP and PM_{10} levels measured during the current study were found to be once exceeded the prescribed values by WHO and USEPA (WHO, 2000; ATSDR, 2002). The 24-h $PM_{3.0}$ are compared with the $PM_{2.5}$ prescribed by WHO (WHO, 2000). It can be noted that the obtained values for $PM_{3.0}$ are several times higher than WHO standard values. Among the atmospheric trace elements Mn levels (24-h) in the present study are in most cases higher compared with the guideline values. A number of health related problems may thus be associated with the elevated TSP and Mn concentrations in the atmosphere (Razos and Christides, 2010). Average TSP levels in the atmosphere of Sarajevo were substantially lower than those reported from Delhi (Shridhar *et al.,* 2010), Bilbao (Aranguiz *et al.,* 2002), Athens (Valvanidis *et al.,* 2006) and Pristina (Arditsoglou and Samara, 2005). However, the TSP levels reported from Tito Scalo (Ragosta *et al.,* 2002), Delft (Wang *et al.,* 2001), Yokohama (Khan *et al.,* 2010) and Santa Cruz (Quiterio *et al.,* 2004) were almost comparable with the levels measured in the present study.

The atmospheric Co levels pertaining to the present study were comparable to those reported for Pristina (Arditsoglou and Samara, 2005) and Santa Cruz (Quiterio *et al.,* 2004). The average levels of Cu in the atmospheric particulates of Sarajevo were lower compared with Tito Scalo (Ragosta *et al.,* 2002) and Yokohama (Khan *et al.,* 2010), but comparable with those reported from Pristina (Arditsoglou and Samara, 2005), Delft (Wang *et al.,* 2001), Bilbao (Aranguiz *et al.,* 2002) and Santa Cruz (Quiterio *et al.,* 2004).Only high polluted metropolitan city of Delhi (Shridhar *et al.,* 2010) has significant higher values for atmospheric Cu. Average levels of airborne Mn is comparable to Bilbao (Aranguiz *et al.,* 2002) and Delhi (Shridhar *et al.,* 2010) but higher to Pristina (Arditsoglou and Samara, 2005), Tito Scalo (Ragosta *et al.,* 2002) and Yokohama (Khan *et al.,* 2010). Present Fe levels in the particulates were found to be comparable with those reported from Athens (Valvanidis *et al.,* 2006) and Delhi (Shridhar *et al.,* 2010). Average Fe levels in the atmosphere of Sarajevo were substantially lower than those reported for Santa Cruz (Quiterio *et al.,* 2004) and Bursa (Samura *et al.,* 2003), but higher than that reported for Pristina (Arditsoglou and Samara, 2005), Tito Scalo (Ragosta *et al.,* 2002) and Bilbao (Aranguiz *et al.,* 2002). In conclusion, the trace element value in the urban atmosphere of Sarajevo in comparison with some European urban sites were many times higher.

CONCLUSION

An important finding of the present study is that a major portion of TSPM concentration is in the form of PM 0.49 (i.e. < 0.49 μm). The concentration of TSPM varies between 9.9 and 77.1μm/m^3. Average concentrations of particulate matter are 37%, 18%, 15%, 8%, 15% and 6% (averaged over all the observations) of total suspended particulate for $PM_{< 0.49}$, $PM_{0.95-0.49}$, $PM_{1.5-0.95}$, $PM_{3.0-1.5}$, $PM_{7.2-3.0}$ and $PM_{> 7.2}$, respectively. Results for TSPM were only tree times exceeded the limit value of 40 μm/m^3 (annual average) for PM10 which was set by the European Commission (European Communities Council Directive 1999/30/EC). Another important finding is that major concentrations of four investigated metals (Cu, Co, Mn and Fe) are associated with the PM 7.2-3.0 μm. Metal concentrations in size-fractionated urban particles ranged from 0.01-3.83 ng/m^3 for Co, 6.30-179.20 ng/m^3 for Cu, 5.00-208.70 ng/m^3 for Mn and 0.35-6.82 μg/m^3 for Fe. Overall, the decreasing trend of average trace element concentrations (24-h) in the particulates revealed the following order: Fe>Mn>Cu>Co. There is a similar distribution of metals in fractions of particulate matter, and similar behavior during the same weather conditions for most of the sets for Cu, Co, Fe, and Mn.

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