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Seasonal Variation and Sources of Elements in Urban Submicron and Fine Aerosol in Brno, Czech Republic

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ABSTRACT

Seasonal variation and sources of elements bound to urban aerosol were studied. Fine ($PM_{2.5}$) and submicron (PM_1) aerosol was collected at 2-week campaigns during four seasons in 2018. The total concentration of 21 elements accounted on average for 3.5% of the total $PM_{2.5}$ mass concentration and 2.0% of the total PM_1 mass concentration. Seasonal differences in the element concentrations in PM_1 and $PM_{2.5}$ were found. Most elements in both aerosol size fractions had a higher concentration in winter or autumn than in summer or spring. Enrichment factor analysis showed extremely highly enriched Cd, Sb and Se in both fractions. Positive matrix factorization (PMF) analysis revealed 3 sources (residential heating, resuspension and anthropogenic background) for PM_1 and 3 sources (residential heating, resuspension and brake abrasion) for $PM_{2.5}$ aerosol in Brno. In addition, health risk assessment of possibly toxic elements was calculated.

Keywords: Particulate matter, Seasonal variation, Sources of elements, ICP-MS, Elements toxicity

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1 INTRODUCTION

Atmospheric aerosol (particulate matter, PM) affects a number of atmospheric processes (for example radiation balance, reduction of visibility and climate changes) and also affects human health (Amit *et al.*, 2013; Zang *et al.*, 2015; Bates *et al.*, 2019; Manousakas *et al.*, 2020). The atmospheric aerosol has been found to be one of the most important ways of spreading elements (Radzi bin Abas *et al.*, 2004; Desboeufs *et al.*, 2005; Amit *et al.*, 2013; Zang *et al.*, 2015; Rushdi *et al.*, 2017). Elements enter to the atmosphere from natural (volcanic eruptions, wind and Earth erosion, forest fires and the sea salts) or anthropogenic (fossil fuels and wood combustion, industrial activities and waste incineration) sources (Allen *et al.*, 2001; Toscano *et al.*, 2005; Qi *et al.*, 2016). They are found in all aerosol size fractions (Finlayson-Pitts and Pitts, 1999; Allen *et al.*, 2001; Ny and Lee, 2011). Their concentration and size distribution are controlled by the nature of emission source (Ottley and Harrison, 1993; Otten *et al.*, 1994; Migon *et al.*, 1997; Gysels and Van Grieken, 1998; Allen *et al.*, 2001; Qi *et al.*, 2016).

The size distribution of particulate matter allows to predict the extent to which elements can be dispersed via atmospheric transport. The potential impact of PM on human is determined by their chemical composition, shape and size (Allen *et al.*, 2001; Galindo *et al.*, 2013). Particle size may have a significant influence on a noxiousness of elements bound to the particle. In general, as particle size decreases, the specific surface increases, which makes elements to be absorbed easier on PM (Zhang *et al.*, 2018a). Also, the degree of respiratory penetration and retention depends on particles sizes, smaller the particle, the more it can penetrate and subsequently deposit into lungs, and stay there for longer time (Espinosa *et al.*, 2001; Chen and Lippmann, 2009; Miguel, 2017).

To evaluate the possible health risks of PM, the research of their chemical composition and size distribution is essential (Espinosa *et al.*, 2001; Wang *et al.*, 2005; Miguel, 2017). Therefore, this study aims to determine the concentration of elements in urban aerosol ($PM_{2.5}$ and PM_1) during four seasons and to estimate their origins. An enrichment factor was determined to evaluate anthropogenic versus natural sources of elements in the particles. Positive Matrix Factorization was used to determine more specific sources and their contribution to the element concentrations. In addition, the health assessment risk of elements (As, Cd, Co, Cr, Mn, Ni, Pb, V and Zn) with confirmed adverse health effects were calculated.
over 387,298 deaths worldwide (WHO, 2020b).

2 METHODS

2.1 $PM_{2.5}$ and PM_1 Samples Collection

Samples of urban aerosol in $PM_{2.5}$ and PM_1 size fraction were collected in Brno (49°12′28.27′′N, 16°36′28.00′′E), the Czech Republic. Brno is the second-largest city in the Czech Republic with 370 thousand citizens. It is an industrial, political and economic centre of Moravia (Křůmal *et al.*, 2010). Sampling was carried out on the balcony of the Institute of Analytical Chemistry on Veveří Street at the height of 8.9 m and the distance of 15.6 m from the road surface (Coufalík *et al.*, 2016). Veveří Street is one of the most critical thoroughfares in Brno, besides heavy traffic in the street, there is an important tram line.

A total amount of 56 urban aerosol samples (28 $PM_{2.5}$ and 28 PM_1 samples) were collected during four 2-week campaigns. Sampling took place in February/March, April/May, July and November 2018. Samples of PM_1 and $PM_{2.5}$ aerosol were collected for 48 hours on nitrocellulose filters (airflow rate of 30 $m^3\ h^{-1}$) in parallel using two high-volume samplers (DHA-80 and DHA-77, Digitel, Switzerland). 14 samples were collected (7 $PM_{2.5}$ and 7 PM_1 samples) in each campaign; 4 samples over the weekend and 10 samples during a working week. Nitrocellulose membrane filters (150 mm diameter, 3 µm porosity, Sartorius, Germany) were before and after sampling equilibrated in a clean climatic room at relative humidity 50% (±2%) and 21°C (±1°C) for 48 hours. Equilibrated filters were weighed on M5P microbalance (±1 µg, Sartorius, Germany) equipped with a special large plate to allow weighing of 150 mm filters (Křůmal *et al.*, 2010). Filters with collected PM after weighting were divided into four equal pieces with the ceramic knife on a PTFE plate. Each piece was individually weighed and then sealed in Petri dishes with Parafilm and stored in a refrigerator at 4°C before analysis. Three unsampled filters were analyzed along with the samples as process blanks for each campaign.

The sampling campaigns were performed under different atmospheric conditions during different seasons of 2018: winter (12 February–1 March), spring (18 April–2 May), summer (9–23 July) and autumn (15–29 November). Meteorological parameters (temperature and relative humidity) were observed for each campaign using commercial sensors (type T3113, Comet System, Czech Republic). The analysis of air masses transported towards the sampling site was performed using the Hysplit model v5.0.0 (Stein *et al.*, 2015; Rolph *et al.*, 2017). One-day backward trajectories calculated for 300, 750, 1500 m above ground level are shown in Supplementary Materials (Fig. S1–S4) separately for each day during all campaigns. The average values of temperature, humidity and predominant wind directions (according to backward trajectories) are shown in Table S1.

2.2 Analytical Procedures

2.2.1 Decomposition of filters

One-quarter of each filter was decomposed in an UltraWAVE microwave mineraliser (MA 149-010, Milestone) in 3 mL of sub-boiling HNO_3 for 25 min. The decomposed filters were transferred quantitatively along with 7 mL of deionized water into polyethylene scintillation vials (Kartel, Italy). Those operations were performed in a clean laboratory with HEPA filters. Increased attention was paid to avoid contamination. Glass and Teflon cups were cleaned in boiling HNO_3 for at least 12 hours and then leached in deionized water (Coufalík *et al.*, 2016).

2.2.2 ICP-QQQMS analysis

Determination of 21 elements was performed using a triple quadrupole inductively coupled plasma mass spectrometer (ICPMS/MS 8800, Agilent Technologies, Germany) with two quadrupoles (Q1, Q2) and an octupole reaction cell (ORC), in different tune modes (collision/reaction, He, O_2, NH_3), working in the on mass mode and mass shift mode used to determine selected elements. Especially for the detection of Cd the reaction with NH_3 in MS/MS mode was necessary to achieve low detection limits. As, Se and Fe were analyzed in O_2 MS/MS mode with a mass shift of +16. The forwarded RF power was 1550 W, carrier gas (Ar) flow rate 1.07 L min^{-1}, integration time per isotope was 0.3 s in all used modes and cell gas flow rates: helium in collision single quadrupole mode: 4 mL min^{-1}, 4th Cell gas (oxygen in reaction MS/MS mode): 0.29 mL min^{-1}, 3rd cell gas (ammonium in reaction MS/MS mode) + helium: 4 mL min^{-1} NH_3 + 1 mL min^{-1} He.

2.2.3 Calibration standards

Calibration standards were prepared in a matrix of 2% HNO_3 from a 1.0000 g L^{-1} stock solution of a single element (Analytika Prague, Czech Republic) mixing into one solution in the concentration range of 0–10 mg L^{-1} for Na, Mg, Ca, K, Mn, Al, Fe and 0 – 100 µg L^{-1} for the rest of elements. Calibration curves of all elements were linear in the whole range (R^2 = 0.9994–1.0000).

Internal standard for elements determination were prepared from commercial 10 mg L^{-1} mixture of Bi, Ge, In, Li, Sc, Tb, Y (Agilent technologies, USA) by dilution in 2% HNO_3 to final concentration 100 µg L^{-1}. The tuning solution were prepared from commercial 10 mg L^{-1} mixture of Li, Co, Y, Ce, Mg and Tl (Agilent technologies, USA) to final concentration 1 µg L^{-1} each element in a matrix of 2% HNO_3.

2.2.4 Quality control

Trueness of the method was evaluated by use of suitable standard reference materials: SLRS-5 River water reference material for trace elements (National Research Council, Canada) and SRM 1640a Trace Elements in Natural Water (National Institute of Standards & Technology, Gaithersburg, USA), which were periodically included in the sample measurement sequence. Furthermore, a control standard (10 µg L^{-1} calibration standard) was measured periodically for instrument stability control. Online monitoring of internal standard solution was used for instrument stability control as well.

SRM 1648a Urban Particulate Matter (National Institute of Standards & Technology, Gaithersburg, USA), ERM -CZ 120 Fine dust (PM_{10}-like) (JRC European Commission, Belgium) were digested and analyzed at the same conditions as samples for the control of the whole determination process from preparation to determination.

2.3 Positive Matrix Factorization

Positive Matrix Factorization (EPA PMF 5.0) was applied to $PM_{2.5}$ and PM_1 elemental composition to obtain source profiles and their contributions. The data matrix was prepared in compliance with the procedure described in Polissar *et al.* (1998). The relative analytical uncertainty ranged between 5 and 20%. The final matrices had 28 samples and 20 species for both PM_1 and $PM_{2.5}$ included as a total variable and modelled separately. Previously, PMF has been successfully applied to datasets with a limited number of samples (Contini *et al.*, 2014; Diapouli *et al.*, 2017; Kozáková *et al.*, 2019). The model was run several times using a different number of factors and different extra modelling uncertainties to obtain the Qtrue closest to Qtheoretical, to determine the most physically meaningful result and the best diagnostics.

2.4 Health Risk Assessment

The corresponding level of cancer risk (CR) or hazard index (HI) for selected element was determined using the lifetime average daily dose (LADD) or average daily dose (ADD). LADD determined the amount of intake per kg of body weight per day of a pollutant suspected of adverse health effects (U.S. EPA, 1992; Liu *et al.*, 2018). Because of the different effects on the respiratory system the subjects were divided into two groups: children (0–6 years) and adults. The LADD (mg kg^{-1} d^{-1}) was calculated according to the following equation:

$$LADD\ (ADD) = C \times \frac{IR \times EF \times ED}{BW \times AT} \tag{1}$$

where C (mg m^{-3}) is the concentration of the element of interest in PM, IR (m^3 d^{-1}) is the inhalation rate, EF (365 d $year^{-1}$) is the exposure frequency, ED (years) is the exposure duration, BW (kg) is the body weight) and AT (days) is the average time (AT_c for carcinogens and AT_n for non-carcinogens). The parameters used in Eq. (1) are listed in Table 1.

Cancer risk is described as the possibility of occurrence of diseases caused by tumors in the whole lifetime due to the impact of compounds that produce carcinogenic effects. The US Environmental Protection Agency (USEPA) recommends that CR values less than 1×10^{-6} are considered as negligible, and values that exceed 1×10^{-4} are probably harmful to human. If hazard index (HI) ≥ 1, there may be concern about possible non-carcinogenic health effects (U.S. EPA, 1989; Cerón Bretón *et al.*, 2019; Adimalla, 2020). Equations are as below:

$$CR = LADD \times CSF \tag{2}$$

$$CSF = IUR \times \frac{BW}{IR} \times 1000 \tag{3}$$

$$HI = \frac{ADD}{RfD} \tag{4}$$

were CSF (kg d mg^{-1}) is cancer slope factor, IUR (m^3 μg^{-1}) is inhalation unit risk a reference value reported in the database of the U.S. EPA (U.S. EPA, 2009) and RfD (mg kg^{-1} d^{-1}) is reference dose of each element considered to have no health effects (Cerón Bretón *et al.*, 2019).

3 RESULTS AND DISCUSSION

3.1 Mass Concentration of Aerosol

Summary of average mass concentrations and their standard deviations for PM_1 and $PM_{2.5}$ aerosol during all campaigns is shown in Table 2. The mass concentration of PM_1 and $PM_{2.5}$ aerosol was two times higher during winter campaign than during summer campaign. Mass concentrations of PM_1 aerosol in spring and summer were on average nearly comparable (Table S2) but in autumn the PM_1 concentrations were slightly higher than in spring or summer. The $PM_{2.5}$ concentrations in autumn were higher than in summer or spring but similar to those in winter (Table S3). The variations in PM mass concentration probably result from the changes in the meteorological situation during the campaigns and also depend on the sources of PM

Table 1. List of parameters used in the exposure calculation.

Parameter	Acronym	Unit	Child	Adult
Inhalation rate	IR	m^3 d^{-1}	12	16
Bodyweight	BW	kg	18	70
Exposure frequency	EF	d $year^{-1}$	365	365
Exposure duration	ED	years	6	30
Average lifetime	ATc	d	25550[a]	25550[a]
	ATn	d	2190[b]	10950[b]

[a] ATc corresponds to the typical life expectancy (70 years) in *days*. [b] ATn corresponds to the relevant ED multiplied by EF.

Table 2. Summary of mass concentration of PM_1 and $PM_{2.5}$ in µg m^{-3}.

	Winter		Spring		Summer		Autumn	
	PM_1	$PM_{2.5}$	PM_1	$PM_{2.5}$	PM_1	$PM_{2.5}$	PM_1	$PM_{2.5}$
average	21.9	30.4	11.5	16.0	9.74	12.4	15.9	24.0
SD	6.25	7.01	2.17	3.06	2.13	2.02	3.13	5.12
min	14.5	19.7	7.92	11.4	7.19	9.34	12.0	17.9
max	32.6	42.0	13.7	20.0	13.7	15.8	20.4	29.8

emissions which likely vary seasonally. The PM_1 and $PM_{2.5}$ mass concentrations show strong negative correlation (correlation coefficient for PM_1 $r = -0.80$ and correlation coefficient for $PM_{2.5}$ $r = -0.87$) with temperature. The temperature changes influence intensity of primary household heating, which probably causes the increased mass concentration of PM_1 and $PM_{2.5}$ aerosol in winter and autumn. Mass concentrations of PM during week-day were not notably different from those of weekends with exceptions of $PM_{2.5}$ aerosol in spring and PM_1 aerosols in summer that are different (Table S4).

To verify representativeness of sampling site, the mass concentrations of $PM_{2.5}$ for all seasons were compared by Kruskal-Wallis test with $PM_{2.5}$ mass concentrations observed by Czech Hydrometeorological Institute at four stations of automatic monitoring system in Brno (i.e., stations Cernopolni (Children's Hospital), Uvoz, Lisen, Turany) (CHMI). The comparison proved that the concentrations of $PM_{2.5}$ aerosol collected on the balcony of the Institute of Analytical Chemistry are comparable with the $PM_{2.5}$ results of other measuring stations in Brno (Fig. S5). The chosen sampling site is therefore representative for Brno in terms of $PM_{2.5}$ concentrations. The annual $PM_{2.5}$ limit value was not exceeded at any measuring station in Brno in 2018 (CHMI).

A comparison of the observed mass concentrations of PM_1 in this study with the results of previous studies from the same sampling site in Brno in 2009 and 2010 (Mikuška *et al.*, 2020) and 2016 (Coufalík *et al.*, 2016) found that there are no significant changes in mass concentrations (Table S5). Slightly increased mass concentrations were observed in the winter campaign of 2010 and the summer campaign of 2009 in comparison with data of this study. Lower mass concentrations were found in the winter campaign of 2009 and 2016 and the summer campaign of 2010 (Coufalík, 2016; Mikuška *et al.*, 2020). It is likely that there has been no significant change of air pollution in Brno over the years in terms of the PM_1 fraction.

Summer mass concentrations of $PM_1$and $PM_{2.5}$ from Brno were similar to summer samples from Elche, Spain (Galindo *et al.*, 2011) but winter mass concentrations of PM_1 and $PM_{2.5}$ in Elche were lower than those found in Brno. Winter and summer concentrations of PM_1 (Rogula-Kozłowska *et al.*, 2019) and $PM_{2.5}$ (Juda-Rezler *et al.*, 2020) from Warsaw, Poland were similar to those found in Brno. Studies in Milan, Italy (Vecchi *et al.*, 2004) and Zabrze, Poland (Rogula-Kozłowska *et al.*, 2012; Rogula-Kozłowska *et al.*, 2019) found higher $PM_{2.5}$ and PM_1 mass concentration in summer and winter than in Brno. Therefore, the PM mass concentrations observed at Brno lie in a similar range to those measured at other European cities.

3.2 Concentrations of Elements in PM_1 and $PM_{2.5}$ Aerosol

The average total concentration of elements in PM_1 and $PM_{2.5}$ samples, maximum and minimum contents, limits of detection (LOD) and limits of quantification (LOQ) are presented in Table 3.

The sum of analyzed elements detected by ICP-MS represents a small part of the mass of the particulate matter for both fractions accounting on average for 2.0% in PM_1 mass and 3.5% in $PM_{2.5}$ mass. The highest contribution of elements to the mass of the PM_1 and $PM_{2.5}$ was observed in spring (3.5% in PM_1 and 5.7% in $PM_{2.5}$). In other seasons, the contributions of elements to the mass of the particles were comparable (1.5% in PM_1 and 2.9% in $PM_{2.5}$ in winter, 1.4% in PM_1 and 3.0% in $PM_{2.5}$ in summer, 1.7% in PM_1 and 2.3% in $PM_{2.5}$ in autumn). The sum of element concentration in both PM_1 and $PM_{2.5}$ shows a slightly similar trend to that of PM mass concentration. The lowest sum of element concentration in PM_1 and $PM_{2.5}$ aerosol was in summer, which corresponded with PM mass concentrations. The highest sum of average elements concentration was in spring, but the highest PM mass concentrations were in winter.

Table 3. Seasonal variations of element concentration (ng m^{-3}) bound to PM_1 and $PM_{2.5}$ aerosol.

Sample			Al	As	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sb	Se	Sn	Ti	V	Zn
winter	PM_1	**average**	**11.5**	**1.01**	**0.37**	**9.65**	**0.32**	**0.04**	**0.33**	**1.97**	**38.9**	**172**	**4.83**	**2.17**	**51.6**	**0.38**	**7.36**	**0.48**	**0.65**	**0.70**	**0.55**	**0.24**	**20.3**
		min	6.62	0.65	0.24	1.39	0.20	0.02	0.10	1.40	25.9	122	3.04	1.26	13.9	0.23	3.46	0.25	0.48	0.39	0.23	0.07	14.6
		max	23.6	1.41	0.56	19.3	0.47	0.05	0.67	2.75	58.6	297	8.06	4.39	147	0.50	10.9	0.78	0.91	1.36	1.21	0.54	38.6
	$PM_{2.5}$	**average**	**97.2**	**1.28**	**2.45**	**101**	**0.48**	**0.08**	**1.02**	**4.53**	**178**	**226**	**35.7**	**5.09**	**176**	**0.62**	**9.90**	**0.75**	**0.84**	**1.15**	**5.07**	**0.53**	**39.9**
		min	63.2	0.90	1.40	46.1	0.21	0.05	0.58	2.85	108	157	18.8	3.25	69.8	0.40	3.94	0.41	0.48	0.76	3.23	0.21	18.3
		max	167	1.67	3.65	182	0.85	0.11	0.34	8.23	260	313	58.7	8.09	528	0.99	14.5	1.38	1.08	2.24	9.16	1.13	82.6
spring	PM_1	**average**	**56.4**	**0.49**	**1.06**	**31.6**	**0.12**	**0.03**	**0.40**	**1.76**	**77.9**	**92.5**	**15.0**	**2.01**	**102**	**0.25**	**3.12**	**0.50**	**0.64**	**0.59**	**1.90**	**0.37**	**13.9**
		min	13.0	0.27	0.63	22.1	0.04	0.02	0.30	1.55	44.2	53.9	6.16	1.40	84.0	0.17	1.05	0.25	0.35	0.36	0.53	0.14	5.64
		max	103	0.98	1.23	37.0	0.28	0.04	0.55	2.34	99.3	160	24.2	2.75	117	0.46	5.72	0.69	1.04	0.89	0.53	0.73	34.0
	$PM_{2.5}$	**average**	**200**	**0.57**	**3.69**	**99.6**	**0.13**	**0.08**	**1.14**	**4.13**	**272**	**134**	**53.2**	**5.32**	**106**	**0.47**	**3.35**	**0.58**	**0.69**	**1.00**	**7.61**	**0.67**	**17.3**
		min	103	0.36	2.46	64.2	0.06	0.05	0.79	3.34	147	87.5	28.5	3.79	84.7	0.26	1.29	0.40	0.42	0.82	3.16	0.33	9.74
		max	347	1.05	4.32	141	0.24	0.14	1.55	5.10	348	191	86.7	7.09	132	0.64	5.70	0.86	0.99	1.41	11.31	1.27	31.9
summer	PM_1	**average**	**13.0**	**0.57**	**0.18**	**19.5**	**0.03**	**0.01**	**0.24**	**0.98**	**33.1**	**31.7**	**4.67**	**1.09**	**18.4**	**0.21**	**1.66**	**0.94**	**0.53**	**0.33**	**0.71**	**0.09**	**3.86**
		min	8.68	0.16	0.06	10.5	0.02	0.01	0.16	0.57	25.6	23.5	3.41	0.73	6.6	0.12	0.91	0.23	0.35	0.25	0.44	0.05	2.82
		max	16.2	1.10	0.51	33.5	0.05	0.01	0.33	1.42	39.4	44.6	6.63	1.98	40.2	0.28	2.52	10.54	0.78	0.48	0.89	0.20	6.25
	$PM_{2.5}$	**average**	**57.6**	**0.62**	**1.12**	**73.5**	**0.03**	**0.03**	**0.53**	**2.54**	**106**	**54.3**	**19.6**	**2.21**	**48.2**	**0.29**	**1.69**	**1.83**	**0.57**	**0.58**	**2.87**	**0.14**	**6.48**
		min	39.4	0.17	0.78	56.2	0.03	0.02	0.35	1.36	86.4	42.2	15.4	1.53	19.4	0.22	0.97	0.30	0.37	0.40	2.06	0.09	4.60
		max	76.9	1.35	1.38	101	0.05	0.03	0.70	3.33	121	71.9	25.6	3.42	123	0.41	3.24	5.89	0.91	0.75	3.79	0.17	11.8
autumn	PM_1	**average**	**4.30**	**0.57**	**0.11**	**10.9**	**0.15**	**0.02**	**0.40**	**1.62**	**28.0**	**126**	**4.30**	**1.36**	**66.3**	**0.18**	**5.34**	**0.52**	**0.45**	**0.60**	**0.18**	**0.08**	**25.8**
		min	0.76	0.35	0.00	5.61	0.08	0.00	0.10	0.94	14.8	88.8	2.46	0.76	30.7	0.14	3.10	0.21	0.27	0.30	0.10	0.07	10.8
		max	6.15	0.82	0.23	14.7	0.20	0.03	0.62	2.76	38.4	192	6.77	1.97	127	0.27	6.48	0.92	0.64	0.89	0.26	0.10	27.1
	$PM_{2.5}$	**average**	**34.3**	**0.92**	**1.19**	**46.8**	**0.25**	**0.04**	**0.98**	**4.39**	**101**	**205**	**11.3**	**3.25**	**80.2**	**0.31**	**7.75**	**0.64**	**0.72**	**1.02**	**1.19**	**0.19**	**44.7**
		min	6.71	0.66	0.64	20.1	0.13	0.01	0.37	3.23	59.9	130	4.55	1.49	43.1	0.22	4.39	0.41	0.02	0.05	0.03	0.14	20.8
		max	75.5	1.59	1.98	83.5	0.60	0.07	1.51	6.73	165	369	19.6	5.04	113	0.42	18.4	0.99	1.26	2.16	2.91	0.39	112
LOD			0.012	0.003	0.0007	0.306	0.0001	0.0001	0.005	0.014	0.038	0.551	0.046	0.002	0.077	0.003	0.001	0.0008	0.004	0.019	0.0006	0.002	0.025
LOQ			0.022	0.007	0.0011	0.732	0.0003	0.0003	0.012	0.022	0.052	1.430	0.071	0.004	0.173	0.008	0.003	0.0021	0.010	0.003	0.0015	0.003	0.053

Elements showed similar seasonal concentration variability, where the concentration of elements originated predominantly from anthropogenic sources (e.g., As, Cd, Cu, K, Pb, Se, Sn and Zn) were higher in winter/autumn than in spring/summer and the concentration of elements originated predominantly from natural sources (e.g., Al, Ba, Ca, Fe, Mg and Ti) were higher in spring/summer than in winter/autumn. Potassium was the most abundant element in both fractions in winter and autumn. The highest level of potassium in these periods is probably due to its primary emission sources - coal combustion and biomass burning (Yu *et al.*, 2018). Emissions from these sources are increasing through the heating season in autumn and winter. The highest average concentrations of As, Cd, Co, Cu, Ni, Pb and Se for both fractions occurred in winter, and the highest levels of Cr and Zn for both fractions occurred in autumn. These elements come mainly from anthropogenic sources like burning fossil fuels, coal combustion, biomass burning, industrial processes, vehicle emissions etc. (Chakraborty and Gupta, 2010; Nordberg *et al.*, 2007). Increased anthropogenic activities (biomass burning, coal combustion, etc.) combined with the low boundary layer, low wind speed and low temperature leading to poor dispersion condition could cause increased concentrations of these elements (Chakraborty and Gupta, 2010).

Increased concentrations of Al, Ba, Ca, Fe, Mg and Ti especially for $PM_{2.5}$ fraction were found, which could indicate resuspension of soil dust in spring. Al, Ba, Fe, Mg and Ti comes predominantly from soil dust, and their emission is increased in warmer periods (Rudnick and Gao, 2003; Titos *et al.*, 2014; Nordberg *et al.*, 2007; Sarti *et al.*, 2015; Cheng *et al.*, 2018). For Al, Ba, Ca, Mg and Ti the lowest concentrations were observed in winter or autumn. The highest concentration of Sb in PM_1 and $PM_{2.5}$ was observed in summer. The observed increase of Sb concentration, especially in PM_1 fraction, could be caused by road dust (Titos *et al.*, 2014). Ca and Fe showed increased concentration also during winter, especially in $PM_{2.5}$ fraction. The concentration of elements originating mainly from the soil dust was probably higher in spring/summer season due to higher wind speed and lower humidity, which resulted in enhanced resuspension of crust.

The comparison of element concentrations found in PM_1 and $PM_{2.5}$ fractions during winter and summer indicated that some elements (As, Cd, Ni, Pb, Se, Zn) with primary anthropogenic source (Chung *et al.*, 2014; Zhang *et al.*, 2018b; Mainka and Zahusz-Zubek, 2019) are more often found in the PM_1 fraction. $PM_1/PM_{2.5}$ ratio of Ni and Se did not exhibit any significant winter/summer seasonal effect but As, Cd, Pb and Zn $PM_1/PM_{2.5}$ ratio changes during the seasons. These changes were probably caused by different elements sources. Potassium concentrations share of PM_1 in $PM_{2.5}$ appear to be 20–40% higher during winter and autumn than in summer, which is probably due to emissions from domestic heating (Yu *et al.*, 2018). Al, Ca, Fe, Mg, Na and Ti were present for about 60–90% in $PM_{2.5}$ fraction and were probably emitted by natural sources (Kegler *et al.*, 2001; EEA, 2012) or by traffic relative sources (Monaci and Bargagli, 1997).

Elements can originate from several different sources (Mikuška *et al.*, 2020). As and Cd are emitted mainly from coal combustion. Various industrial sources, traffic, biomass burning and coal combustion produced Ca, Cu, Fe, Mn, Pb, Sb or Zn. In addition some elements also originate partly from a municipal solid waste incinerator (As, Cd, Cu, Mn, Ni, Pb, Sb, Sn, V and Zn) located at the east end of sampling site in Brno, incineration of waste in households (Cd, Cu, Pb, Sb, Sn, Ti and Zn) or cement plant (As, Ca, Cd, Cu, Fe, Mn, Ni, Pb and Zn) located east of Brno (Hu *et al.*, 2003; Conesa *et al.*, 2011; Gupta *et al.*, 2012; Titos *et al.*, 2014; Arfala *et al.*, 2018; Rovira *et al.*, 2018; Horák *et al.*, 2019; Mikuška *et al.*, 2020).

The concentrations of elements found in PM_1 samples in this study are comparable with those (Table S6) found in other studies in Brno (Coufalík *et al.*, 2016; Mikuška *et al.*, 2020). The concentration of potassium was comparable to the concentrations from 2009 and 2010 for summer samples, in winter the potassium concentration for 2018 was higher than for 2009 but lower than for 2010. The major differences in the concentration were observed for Ba and Pb. Both of these elements had lower concentrations in 2018 than in samples from 2009 and 2010 but lead concentrations in 2018 and 2014/2015 were comparable. The element concentrations in corresponding seasons were similar predominantly with samples from 2010.

The concentrations of most elements in $PM_{2.5}$ and PM_1 samples (Tables S6–S7) are comparable with the concentrations found in Nanjing (Qi *et al.*, 2016), Karachi (Mansha *et al.*, 2012), Milan (Vecchi *et al.*, 2004), Barcelona (Moreno *et al.*, 2011) but lower than those in Nanjing (Wu *et al.*, 2019), Wuhan (Zhang *et al.*, 2015), Kanpur (Chakraborty and Gupta, 2010), Tito Scalo (Caggiano *et al.*, 2010), Katowice (Rogula-Kozłowska, 2015).

3.3 Enrichment Factor

Enrichment factors (EFs) were determined concerning the content of elements in the Earth´s crust to differentiate the anthropogenic and natural origin of the studied elements. EF of an element is based on normalizing an observed element against a crustal concentration of a reference element. The reference element is an element strongly stable in the crust (Rudnick and Gao, 2003). Typical reference elements used in other studies are Al (Zhang *et al.*, 2007; Rogula-Kozłowska *et al.*, 2012; Sudheer and Rengarajan, 2012; Rogula-Kozłowska *et al.*, 2013a; Barbieri, 2016), Fe (Ghrefat *et al.*, 2011; Coufalík *et al.*, 2016; Blanchard *et al.*, 2018), Mn (Barbieri, 2016) and Ce (Coufalík *et al.*, 2016). EF is calculated using the equation:

$$EF_x = (X/Y)_{aerosol}/(X/Y)_{crust} \quad (5)$$

where $(X/Y)_{aerosol}$ are concentrations of the examined (X) and reference (Y) element in the samples and $(X/Y)_{crust}$ are average concentrations of the examined and reference element in the Earth crust. Generally, $EF < 10$ indicates the crustal soil as the predominant origin of the element. Extremely high enrichment by anthropogenic sources is considered at $EF > 40$ (Ghrefat *et al.*, 2011; Barbieri *et al.*, 2016; Coufalík *et al.*, 2016; Blanchard *et al.*, 2018).

The calculated average EFs from element concentrations determined in PM_1 and $PM_{2.5}$ collected in Brno with iron as the reference element are presented in Fig. 1. High values of EFs observed for As, Cd, Cu, Pb, Sb, Se, Sn, and Zn indicate that these elements originate primary from anthropogenic sources. The lowest values of EF were observed for Al, Ba, Ca, Co, Mg, Na and Ti, which indicates negligible contribution of anthropogenic sources and main origin from natural sources.

Although the elements in PM_1 emitted predominantly from anthropogenic sources (As, Cd, Pb, Sb, Se, Sn, Zn) consistently showed higher EFs than same elements in $PM_{2.5}$, they both lie within the error limits. EFs of elements originated predominantly from natural sources (Al, Ba, Ca, Mg and Ti) were higher in $PM_{2.5}$ fraction than in PM_1 fraction. These results indicate the enhanced association of anthropogenic sources of these elements with the submicron fraction (Al-Momani and Shatnawi, 2017; Mohammed *et al.*, 2017).

Comparison of seasonal changes of EF values (Table S8) indicates that for most of the elements (namely As, Cd, Co, Cu, K, Pb, Se, Sn and Zn) in both fractions, EFs were higher in colder periods. Other elements such as Al, Ba, Ca, Mg and Ti with low values of EFs did not show distinct seasonal trend.

3.4 Source Identification

Positive Matric Factorization (PMF) was applied to determine sources contribution to the fine

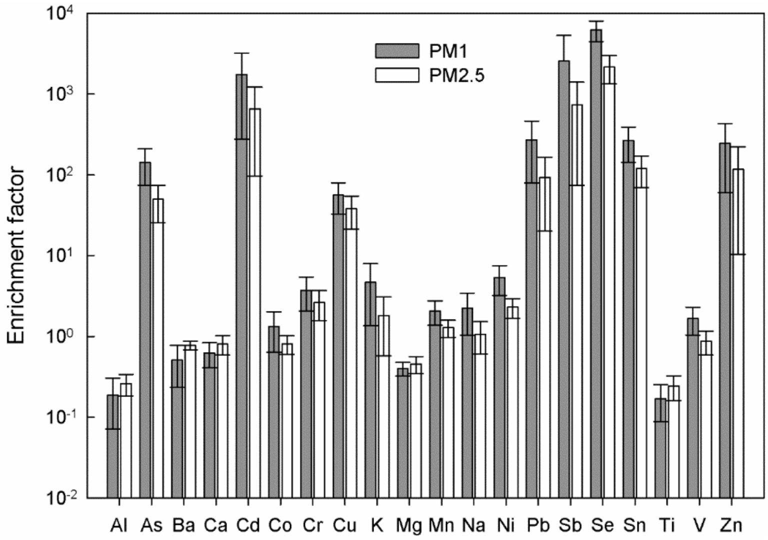

Fig. 1. Average enrichment factors of elements.

and submicron aerosol fraction. The model was run several times with different factor numbers (3–6) to determine the most physically meaningful result and the best model diagnostics (Table S9). The Q values, the resulting source profiles, and the scaled residuals were examined. The most stable solutions were found for 3 factors for both size fractions by extra modelling uncertainties of 3.0 and 9.3% for PM$_{2.5}$ and PM$_1$, respectively. The factor chemical profiles and the time-series plots of the estimated contributions from each factor to the PM$_{2.5}$ and PM$_1$ mass are shown in Figs. 2 and 3.

The first factor common for both fractions, *residential heating*, was associated with high concentrations of K, Zn, Pb and As that originate predominantly from coal and biomass combustion for the domestic heating (Rogula-Kozłowska *et al.*, 2013b; Hovorka *et al.*, 2015; Samek *et al.*, 2017; Pokorná *et al.*, 2018). In Brno, gas or electricity is mainly used for residential heating. Coal and wood combustions are used for heating only in 0.46% of houses in Brno, whereas up to 11% of houses in small villages nearby Brno use coal and wood for residential heating (Křůmal *et al.*, 2010, 2013, 2015; Mikuška *et al.*, 2017). Increased concentrations of K, Zn, Pb and as thus probably result from the regional transport of PM from the villages surrounding Brno. We calculated one-day backward trajectories for all sampling campaigns to test this hypothesis (Figs. S1–S4). Analysis of trajectories proved the transport of air masses from all directions, although during individual campaigns certain wind directions prevailed (Table S1). The elements in the residential heating factor showed strong correlation (Pearson correlation coefficient r = 0.82) and clear seasonal trend with winter maxima in both fractions. Residential heating dominated in the PM$_1$ with an average contribution of 53% (PM$_{2.5}$ 44%).

The second factor common for both sizes was ascribed to *resuspension of road dust* dominated by Earth's crust elements (Mooibroek *et al.*, 2011; Bressi *et al.*, 2014; Diapouli *et al.*, 2017). Re-suspension of dust and road wear related particles are the primary contributors to traffic non-exhaust emissions (Keuken *et al.*, 2010). Since the sampling site is located near the road with tram traffic, the road dust also includes particles related to tram rail wear such as Fe, Ca, Al, Mn, Cu, Ba, Zn etc. (Abbasi *et al.*, 2013; Švábenská *et al.*, 2016; Minguillón *et al.*, 2018; Gonet and Maher, 2019). Road dust consists of primarily coarse-sized particles; hence the factor contributed by 12% to PM$_{2.5}$ and by 1% to PM$_1$ in average, and the clear seasonal trend with high factor contribution in spring is apparent in PM$_{2.5}$. The elements in this factor are strongly correlated (Pearson correlation coefficient r = 0.92).

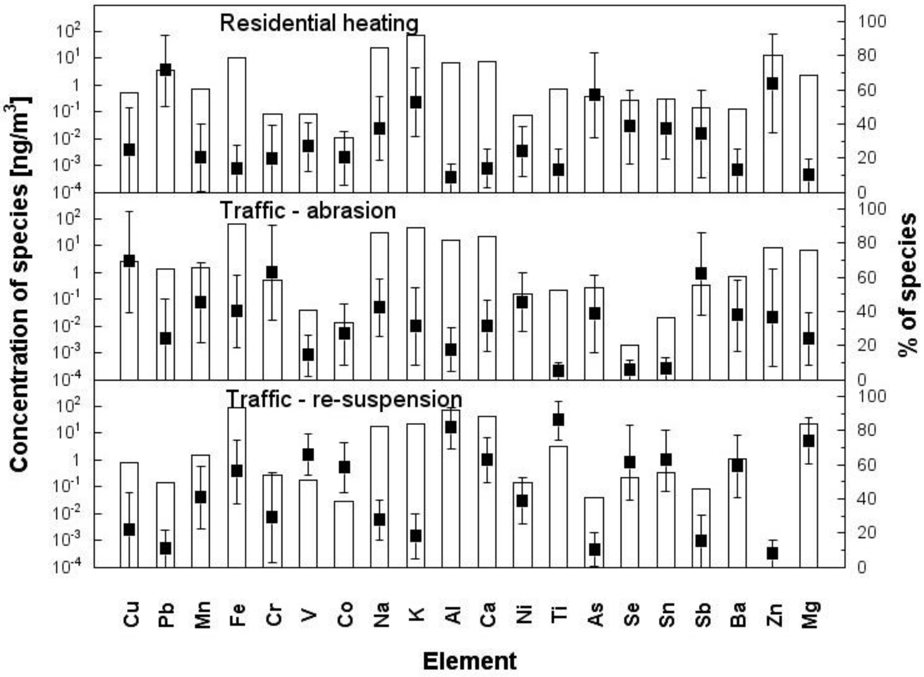

Fig. 2. Factor profiles (concentration of the species – bars, percentage of the species – markers; DISP Average – box, DISP Max and DISP Min – whiskers) for PM$_{2.5}$ (left) and PM$_1$ (right) resolve.

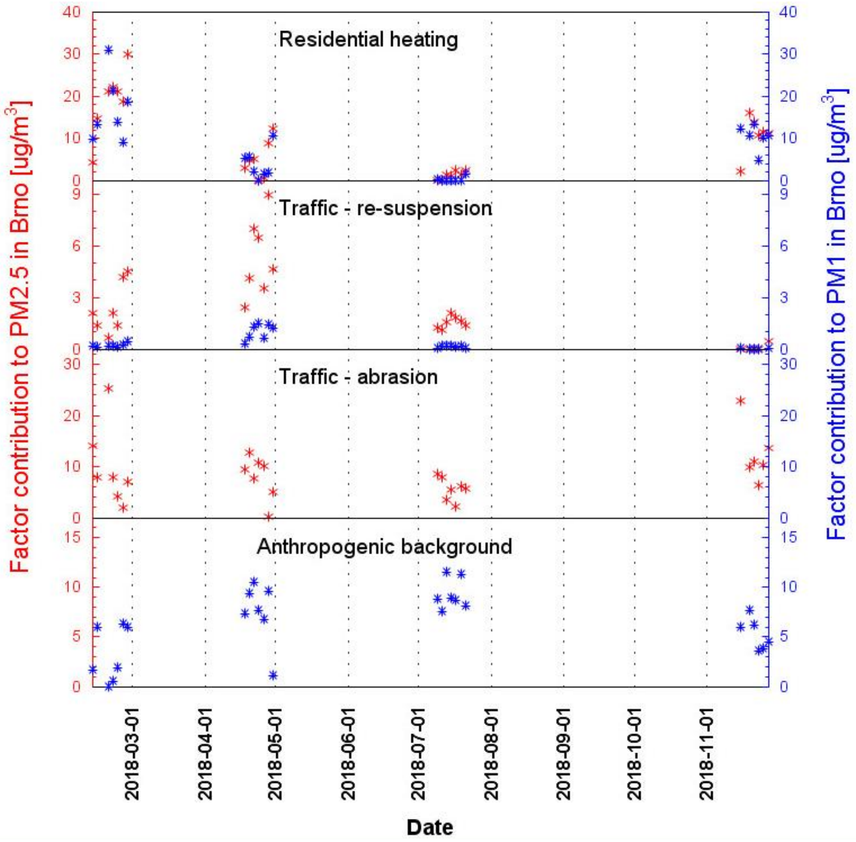

Fig. 3. Factor time series resolved by PMF.

The third factor, *abrasion of brakes*, represented by Cu, Cr, Sb (Wahlin *et al.*, 2006; Ondráček *et al.*, 2011; Varrica *et al.*, 2012) was resolved by the model for $PM_{2.5}$ only with an average contribution of 44%. There is a parking lot under the balcony on which the samplers were placed, also there is a large car sales shop about 40 meters from the sampling point. Moreover, there is a road junction loaded with frequent traffic jams due to high traffic and two large parking lots approximately 100 meters from the sampling point. Brake wear release PM directly into the atmosphere and contribute to the trace element concentration in airborne PM, particularly less than 10 µm (Garg *et al.*, 2000; Wahlin *et al.*, 2006; Gietl *et al.*, 2010; Varrica *et al.*, 2012). The ratio of Cu and Sb (8.0 ± 0.3) as characteristic of brake wear particles was in agreement with study Amato *et al.* (2011) conducted in at roadside in Barcelona. However, the literature reports differences in Cu/Sb ratio attributed to the difference in the brake pad composition and the site characteristics (Pant and Harrison, 2013).

The last factor assigned as *the anthropogenic background* was related with tracers of traffic as well as local (engineering industry, foundry, municipal waste incinerator) or regional (cement factory) industrial emissions and resolved only for PM_1 with the contribution of 45%. Mixed anthropogenic PM_1 sources were also identified by Vecchi *et al.* (2008) in Florence and by Richard *et al.* (2011) in Zürich. The factor shows a clear seasonal trend with high contributions in spring and summer linked to higher wind speed and mixing conditions in the city. One day backward trajectories (Figs. S1–S4) indicate possible local (i.e., municipal waste incinerator – east wind, foundry – south wind) as well as regional (cement factory – east wind) transport of air masses to sampling site.

The Fig. 3 shows factors contribution to PM_1 and $PM_{2.5}$ in Brno during the seasons. The contribution of residential heating shows a significant dependence on the season, in the colder seasons its contribution increases. The contribution of traffic resuspension to $PM_{2.5}$ increased slightly in warmer months when the higher average temperatures causes drier conditions leading to enhanced particles resuspension. The contribution of traffic abrasion and anthropogenic background is almost constant throughout the year.

3.5 Health Risk Assessment

The probability of occurrence of cancer (CR, carcinogenic risk) and hazard index (HI, non-carcinogenic risk) through respiratory system in the exposed human population were calculated for selected elements and two representative groups – children and adults. The lifetime of adults was assumed to 70 years. CR values less than 1×10^{-6} are considered as negligible, and values that exceed 1×10^{-4} are probably harmful to human health. Hazard index values (HI) ≥ 1 may indicate possible non-carcinogenic health effects (U.S. EPA, 1989; Cerón Bretón *et al.*, 2019; Adimalla, 2020). Assessed elements were selected based on their carcinogenic and non-carcinogenic potential for humans (IARC, 1965). The International Agency for Research on Cancer (IARC) classifies substances into several groups. According to this classification, arsenic (As), cadmium (Cd), chromium (Cr), and nickel (Ni) belongs to group 1 (carcinogenic to humans), cobalt (Co) and lead (Pb) belong to group 2B (possibly carcinogenic to humans). For this reason, CR values were calculated only for these elements (IARC, 1965). Moreover, we assessed Mn, V and Zn which induce non-carcinogenic but toxic effects (Zheng *et al.*, 2010; Li *et al.*, 2015).

This study, the CR and HI values (Table 4) were calculated only for inhalation exposure. Carcinogenic and non-carcinogenic health risk of PM_1 for both representative groups peaked in winter and summer. The average values of CR and HI for adults and children were following the order of Cr > As > Pb > Cd > Co > Ni for the CR and Mn > Cr > Co > As > Pb > Cd > Zn > Cu > V

Table 4. Average cancer risk level and hazard index of carcinogenic and element components of PM_1 and $PM_{2.5}$ for adult (A) and children (CH).

				As	Cd	Co	Cr	Mn	Ni	Pb	V	Zn
PM_1	Carcinogenic (CR)	CH	winter	3.7E-07	4.9E-08	2.9E-08	3.38E-07		7.7E-09	5.0E-08		
			spring	1.8E-07	1.9E-08	8.4E-09	4.13E-07		4.3E-09	1.2E-08		
			summer	2.1E-07	5.2E-09	8.4E-09	2.45E-07		4.3E-09	1.2E-08		
			autumn	2.1E-07	2.3E-08	1.5E-08	4.08E-07		3.8E-09	3.7E-08		
		A	winter	6.4E-07	2.5E-07	3.6E-08	5.8E-07		3.9E-08	2.5E-07		
			spring	3.1E-07	9.3E-08	2.6E-08	7.1E-07		2.6E-08	1.1E-07		
			summer	3.6E-07	2.6E-08	1.0E-08	4.2E-07		2.1E-08	5.8E-08		
			autumn	3.6E-07	1.1E-07	1.9E-08	7.0E-07		1.9E-08	1.8E-07		
	Non-carcinogenic (HI)	CH	winter	2.2E-03	2.1E-04	4.4E-03	7.7E-03	1.0E-01	1.2E-05	1.4E-03	2.3E-05	5.7E-05
			spring	1.1E-03	8.0E-05	3.2E-03	9.3E-03	9.6E-02	8.2E-06	5.9E-04	3.5E-05	3.1E-05
			summer	1.3E-03	2.3E-05	1.3E-03	5.6E-03	5.2E-02	6.7E-06	3.2E-04	8.6E-06	8.6E-06
			autumn	1.3E-03	9.8E-05	2.3E-03	9.4E-03	6.5E-02	5.9E-06	1.0E-03	8.1E-06	4.5E-05
		A	winter	7.6E-04	7.3E-05	1.5E-03	2.6E-03	3.5E-02	4.2E-06	4.8E-04	7.7E-06	2.0E-05
			spring	3.7E-04	2.8E-05	1.1E-03	3.2E-03	3.3E-02	2.8E-06	2.0E-04	1.2E-05	1.1E-05
			summer	4.3E-04	7.7E-06	4.4E-04	1.9E-03	1.8E-02	2.3E-06	1.1E-04	2.9E-06	2.9E-06
			autumn	4.3E-04	3.4E-05	7.9E-04	3.2E-03	2.2E-02	2.0E-06	3.5E-04	2.8E-06	1.5E-05
$PM_{2.5}$	Carcinogenic (CR)	CH	winter	4.7E-07	7.5E-08	6.3E-08	1.0E-06		1.3E-08	6.8E-08		
			spring	2.1E-07	2.0E-08	6.5E-08	1.2E-06		9.7E-09	2.3E-08		
			summer	2.3E-07	5.3E-09	8.4E-09	5.5E-07		4.3E-09	1.2E-08		
			autumn	3.4E-07	3.9E-08	3.1E-08	1.0E-06		6.5E-09	5.3E-08		
		A	winter	8.1E-07	3.7E-07	7.8E-08	1.8E-06		6.3E-08	3.4E-07		
			spring	3.6E-07	9.9E-08	8.1E-08	2.0E-06		4.9E-08	1.1E-07		
			summer	3.9E-07	2.6E-08	2.6E-08	9.4E-07		3.0E-08	5.7E-08		
			autumn	5.8E-07	2.0E-07	3.9E-08	1.7E-06		3.2E-08	2.7E-07		
	Non-carcinogenic (HI)	CH	winter	2.8E-03	3.2E-04	9.5E-03	2.4E-02	2.4E-01	2.0E-05	1.9E-03	5.0E-05	8.9E-05
			spring	1.3E-03	8.6E-05	9.8E-03	2.7E-02	2.5E-01	1.5E-05	6.3E-04	6.4E-05	3.9E-05
			summer	1.4E-03	2.3E-05	3.1E-03	1.2E-02	1.1E-01	9.4E-06	3.1E-04	1.3E-05	1.4E-05
			autumn	2.0E-03	1.7E-04	4.7E-03	2.3E-02	1.5E-01	1.0E-05	1.5E-03	1.8E-05	9.9E-05
		A	winter	9.7E-04	1.1E-04	3.3E-03	8.1E-03	8.3E-02	6.8E-06	6.4E-04	1.7E-05	3.0E-05
			spring	4.4E-04	2.9E-05	3.4E-03	9.1E-03	8.7E-02	5.2E-06	2.2E-04	2.2E-05	1.3E-05
			summer	4.7E-04	7.8E-06	1.1E-03	4.2E-03	3.6E-02	3.2E-06	1.1E-04	4.5E-06	4.9E-06
			autumn	7.0E-04	5.8E-05	1.6E-03	7.9E-03	5.3E-02	3.5E-06	5.0E-04	6.3E-06	3.4E-05

> Ni for the HI. Carcinogenic and non-carcinogenic health risk of $PM_{2.5}$ for both representative groups showed similar seasonal trend as PM_1. The average values of Cr and HI for adults and children were following the order of Cr > Co > As > Pb > Cd > Ni for the CR and Mn > Cr > Co > As > Pb > Cd > Cu > Zn > V > Ni for the HI. As a result of individual differences (body weight, respiratory rate, exposure duration and average lifetime), the carcinogenic and non-carcinogenic risks to adults and children were different. For both aerosol fraction, the average carcinogenic risks of selected elements were higher for adults than for children but average non-carcinogenic risks were higher for children.

In any season, the upper limit value of CR or HI evaluated in $PM_{2.5}$ or PM_1 wasn't exceeded. All CR values were below value of 1×10^{-6}, so their effect on human health via inhalation in terms of cancer was negligible. The highest values of HI were achieved for Mn (in PM_1 HI = 0.08 for children and HI = 0.03 for adults, in $PM_{2.5}$ HI = 0.19 for children and HI = 0.06 for adults), but even these values are lower than the limit value of 1.00. The obtained results indicate no significant health risk for both representative groups from inhalation exposure to studied elements.

$PM_{2.5}$ HI and CR values of As, Cd, Co, Cr, Mn, Ni, Pb, V and Zn obtained in urban and rural site of Shandong province (Zhang *et al.*, 2018c), Nanjing (Hu *et al.*, 2012) and Chengdu (Li *et al.*, 2016) were higher than those in Brno. $PM_{2.5}$ HI values of Pb and Zn found in Shangai (Huang *et al.*, 2018) and CR values of As, Cd, Ni found in Tianjin (Chen *et al.*, 2015) were comparable to values in Brno but HI of Mn (Chen *et al.*, 2015; Huang *et al.*, 2018), Pb and Zn (Huang *et al.*, 2018) were lower than in Brno.

4 CONCLUSIONS

The average mass concentrations of $PM_{2.5}$ and PM_1 aerosol collected in Brno during four seasons within 2-week campaigns were 20.8 ± 8.1 µg m^{-3} and 14.8 ± 5.4 µg m^{-3}. The aerosol mass concentrations were the highest in winter and lowest in summer. The comparison of observed $PM_{2.5}$ mass concentrations with results from another measuring stations in Brno proved the chosen sampling site as representative for Brno. In 2018, the annual $PM_{2.5}$ limit value was not exceeded at any measuring station in Brno.

High values of enrichment factors (EF > 40) were observed for As, Cd, Pb, Sb, Se, Sn and Zn. Therefore, the primary sources of these elements were considered to be anthropogenic. The lowest values of EFs observed for Al, Ba, Ca, Mg and Ti indicated that these elements originated primarily from soil resuspension. In both fractions, the highest average annual concentration was found for K and the lowest average concentration for Co. The dependence of element concentrations in PM_1 and $PM_{2.5}$ on season was proved except for Se and Ni, whose concentrations were similar during all campaigns 2018. The highest concentrations of elements originated mostly from anthropogenic sources were found in winter and autumn and elements which probably originate from resuspension of Earth's crust peaked in spring and summer.

PMF resolved three factors for both PM fractions with two common sources identified as residential heating and road dust resuspension. The high contribution of residential heating to PM_1 and $PM_{2.5}$ proved the significant influence of wood and coal combustion in small villages surrounding Brno on air pollution in the sampled locality. The second main source of PM pollution was traffic resuspension which was a dominant source of $PM_{2.5}$ aerosol. The abrasion of brakes was identified as the third source for the $PM_{2.5}$ fraction and the anthropogenic background was identified as the third source for the PM_1 fraction. The health risk assessment values of cancer risk and health index didn't overcome a critical value, which indicates that the concentrations of As, Cd, Co, Cr, Mn, Ni, Pb, V and Zn were at safe level during all campaigns in 2018.

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DISCLAIMER

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at https://doi.org/10.4209/aaqr.2020.09.0556

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