Geochemistry of urban street sediments of Bratislava, Slovakia

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Abstract. Street dust samples in the inner as well as outer parts of the city agglomeration Bratislava were collected to acquire the baseline pattern of their granulometric, mineralogical and chemical composition and its spatial variation.

Grain-fraction analysis pointed out relatively small rate, ca 1–4 %, of the finest, clay fraction (<2μm). The highest amount, ca 85–90 %, was documented for the sand fraction in all samples.

From the point of view of mineralogical composition analysis of clayey fraction the presence of following minerals in street dust samples was observed: illite, chlorite, quartz and feldspar. Other minerals like kaolinite, smectite and calcite were found variably distributed in collected samples. No significant differences were documented when comparing the inner and outer city samples.

Chemical analyses of 2 size fractions (<1 mm and <0.125 mm) revealed significant concentration variations as a function of particle size mainly for Cu, Fe, Mn and Zn which are characterized with the higher content levels in finer fraction. On the other hand, As and Cd feature the very similar concentrations in both analysed fractions.

Comparison of chemical composition of inner and outer city samples did not show any substantial differences in chemical content levels. Markedly higher mean concentrations in inner city samples have been found in the case of Cr (>100 $mg.kg^{-1}$) and Cu (>200 $mg.kg^{-1}$). Following inner city sampling locations are characterized by the higher contents of some metals: Kamenné námestie (Cr, Fe, Ni and Mn), Hodžovo námestie (Cu and Zn) and Nový most (Pb and Zn). Based on concentration data correlations the significant correlation coefficients were determined for Cr and Ni, Cd and Zn, Fe and Mn with Cr and Ni.

Key words: street sediments, chemical composition, mineralogy, Bratislava, Slovakia

Introduction

Street sediments represent specific component of urban environment which chemical composition reflects the character of anthropogenic activities and geogenic processes proceeding in a particular city agglomeration. Thus the urban street sediment geochemistry represents one of the needful investigation fields concerning assessment of urban environment pollution.

Present studies realized worldwide are focused on analysis of metal concentration levels mainly in the finer street sediment fraction (dust particles) and their potential mobility and availability in urban environment (aquatic systems, Charlesworth & Lee, 1999; Li et al., 2001; Ordóñez et al., 2003; Varrica et al., 2003 and others). Their purpose is to acquire basic information on input and distribution pattern of potentially toxic elements in urban ambient in relation to associated potential health and environmental risks.

In the Slovak Republic a regional and local continual monitoring of air quality and pollution, performed by the Slovak Hydrometeorological Institute, is done (SHMU, 2005). It is focused mainly on quantitative analysis of total suspended particles (TSP), particulate matters <10 μm (PM_{10}) and <2.5 μm ($PM_{2.5}$) and to a lesser extent on concentration determination for some metals in TSP (mainly Cd and Pb). No analogous study in relation to geochemistry of road-deposited sediments, representing potential source of contaminated airborne and fine respirable particles, was until realized.

The main aim of this study is to provide the primary basic characteristics of street sediments in one of the busiest industrial cities of Slovakia, a capital Bratislava. It includes granulometric analysis, mineralogical characteristics and chemical content level determination regarding the different particle size as well as the particles spatial distribution (inner / outer samples). The first impulse for performing such a study was a British research project, realized in summer 2003 in Bratislava. Within its frame the chemical composition analyses of some street sediment samples from the major city crossroads were acquired (Robertson, 2004). A relative consistency of both studies (selection of sampling sites, similar range of chemical analyses) allows to achieve the basic information on possible distribution variability of chemical concentration levels at respective sampling locations.

Presented study represents a contribution to the field of urban pollution characterization and assessment based

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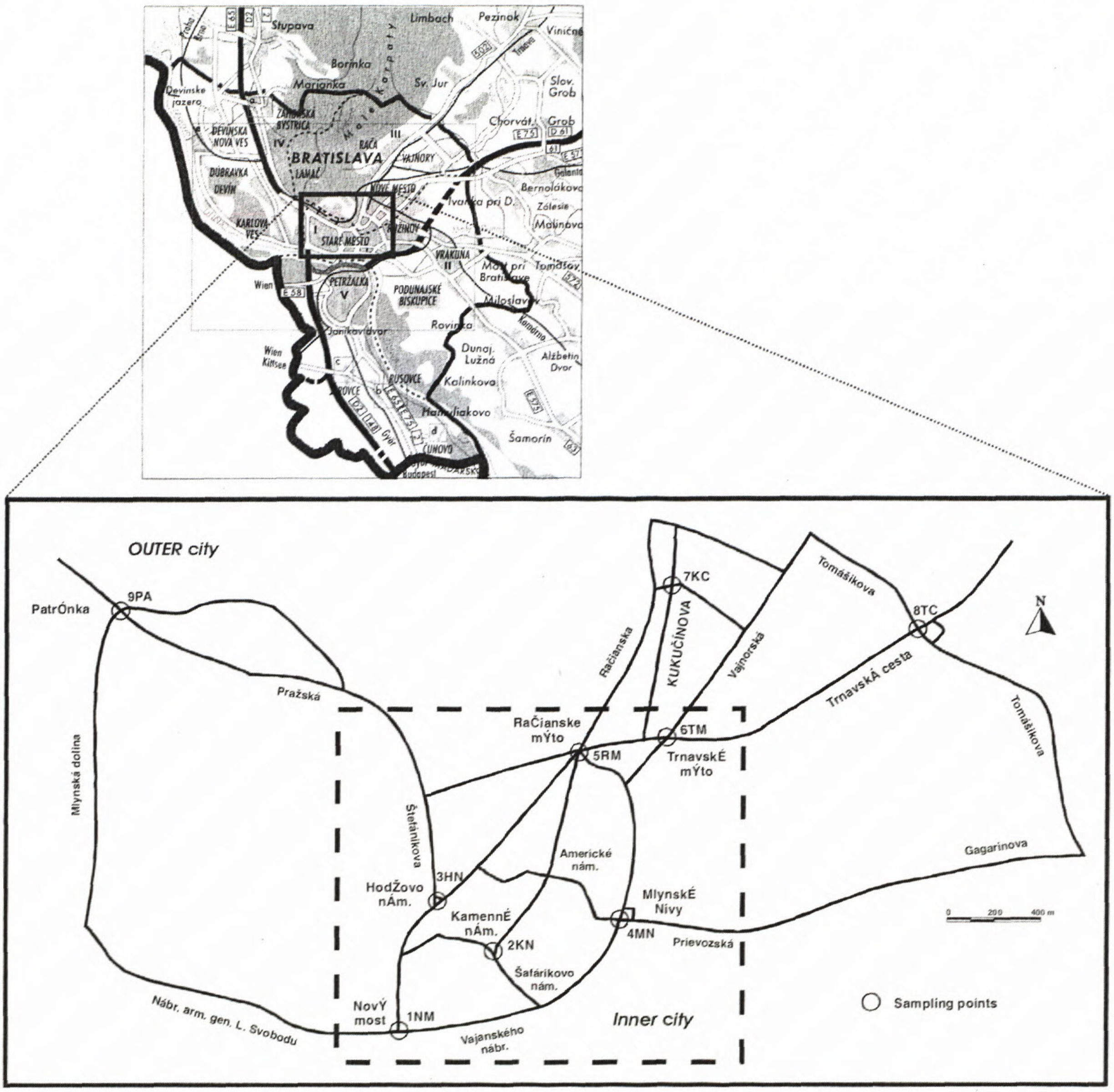

Fig. 1. Map of studied area with sampling locations.

on identification of metal content levels and their spatial distribution in the street sediment samples of one of the most industrialized Slovak cities, Bratislava.

Description of study area

Bratislava is a capital city of the Slovak Republic, situated in the southwestern part of the country. It is spread on both banks on the river Danube on the area ca 370 km^2, at the foot of the Malé Karpaty Mts. and on the boundary of the Danube and Borská lowlands. Bratislava represents the most significant economic and transport centre in Slovakia with population density more than 1,200 inhabitants per km^2 (cca 450,000 inhabitants). Since it is the most industrialized city and the significant traffic junction, it is predisposed to be one of the most polluted regions in the Slovak Republic. Chemical industry, power generation and traffic are defined as the main sources of atmospheric pollution. From the administrative point of view Bratislava is divided to 17 town parts comprising old historical centre, defined in this study as the inner city, and numerous outer city districts.

Materials and methods

Street sediment sampling was carried out in the summer-time (August 2004), in a long-term rainless period. Sampling collection was focused on some major crossroads with the high traffic density in the inner city as well as its outer parts. Overall, 9 samples were collected by the street sweeping at traffic islands on some of the busiest inner/outer city crossroads. The setting of study area and location of sampling sites are shown in Fig. 1. Six of nine samples were located in the central part of the city and three in the outer zone. Inner city samples caught the busiest Bratislava traffic junctions (samples 1KN, 3HN, 4MN, 5RM, 6TM) and marginally also pedestrian zone (2KN). The outer city samples

(7KC, 8TC and 9PA) were situated closely to trunk roads with the big traffic flows.

A plastic dustpan and brush and self-sealing polyethylene bags were used to avoid sample contamination. Sample quantity ranged from 100 to 200 g, depending on the sediment amount accumulated at the site of concern. Collected samples were air-dried at room temperature and screened through a 1 mm aperture metal sieve to exclude large particles, including stones and impurities (e.g. glass, debris). Subsequently, respective sub-samples were acquired in compliance with requirements of further analyses.

Following analytical methods were used within a frame of urban sediment composition analysis:

- Grain-fraction analysis,
- X-ray analysis,
- Chemical analysis.

Grain-fraction analysis

Sub-samples <1 mm of about 60 g undergone the granulometric analysis to provide basic characteristics of collected samples, regarding the composition rate of various grain fractions. Based on using sieves with the different aperture sizes a percentual content of distinct size fractions, sandy (1–0.060 mm), loamy (0.060–0.002 mm) and clayey (<0.002 mm), was determined.

X-ray analysis

The X-ray powder diffraction method was used to provide general information on mineralogical composition of the street sediment samples. A finest, clayey fraction <0.002 mm was analysed since it plays an important role in sorption processes of potentially toxic elements.

Sub-samples <0.002 mm were obtained from suspension consisting of <1 mm fraction and distilled water. Subsequently, the oriented specimens were prepared, both untreated (air-dry) as well as treated with ethylene glycol (EG), and were used for identification of phyllosilicates with expandable layers (e.g. smectite). The X-ray analysis was performed using the DRON – 3 diffractometer (Co anticatode with Fe filter, 15 mA current and 30 kV voltages).

Chemical analysis

Two size fraction chemical composition analyses were carried out for the purpose of chemical content level determination as well as their distribution as a function of particle size. Samples were divided in 2 sub-samples, <1 mm and <0.125 mm, of about 5 g, respectively. The total contents of 10 chemical elements (As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb and Zn) were determined for both particle size fractions, using standard laboratory procedures and analytical methods (see Tab. 1).

Results and discussion

Grain-fraction composition

All samples have similar particle-size composition without any marked differences regarding the inner/outer city sampling site location (see Fig. 2). Sandy fraction predominates with percentage rate corresponding approximately to 85–90 %. Further loamy and clayey fractions are present proportionally in minor quantities, cca 9–11 % and 1–4 %, respectively. Despite of low amount of the finest clay fraction, this still represents, since respirable and relevantly involved in potentially toxic element retention, the most significant particle size fraction with respect to associated potential human health risk.

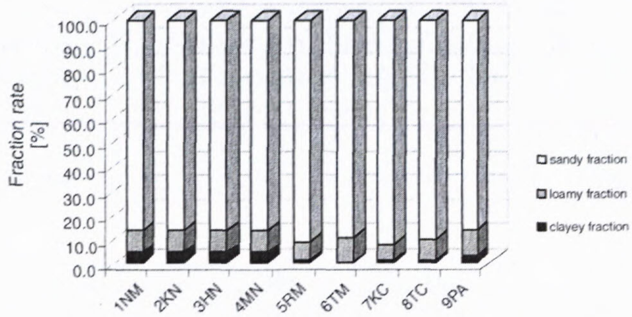

Fig. 2. Percentage rate of grain-size fractions in collected street samples.

Mineralogical composition

In general, the mineralogical composition of clayey sample fraction seems to be rather complex but without any significant differences between inner/outer city sampling sites. Following minerals were determined in all street sediment samples (see Tab. 2): illite, chlorite, quartz and feldspar. Other minerals including kaolinite, smectite and calcite were found to be characterized with variable distribution in collected samples.

Chemical composition

Review of chemical contents, determined in the urban street sediment samples of Bratislava inner and outer city, is shown in Tabs. 3 and 4.

In all of the Bratislava street sediment samples (see Tab. 5) the lowest concentration levels were, in the case of <1 mm fraction, found for As and Cd, ranging from 2.6 to 3.8 $mg.kg^{-1}$ (mean concentration: 3.1 $mg.kg^{-1}$) and 0.6 to 1.2 $mg.kg^{-1}$ (mean concentration: 0.9 $mg.kg^{-1}$). On the other hand, Fe and Mn show the highest concentration levels in a range 19.0-30.6 % (mean concentration: 23.1 %) and 360.8-577.7 $mg.kg^{-1}$ (mean concentration: 428.4 $mg.kg^{-1}$), respectively. Similar trend in chemical content levels was determined also in the case of <0.125 mm fraction.

Generally, data on metal concentration levels in street dust and urban sediments, acquired in analogous studies realized worldwide, are rather variable and often each other inconsistent (different analysed fraction, decomposition methods etc.). Comparing our results with some of existing concentration data on analysed chemical elements (see Tab. 5) we can state similar content range determined for Cd and Zn (Ahmed & Al-Swaidan, 1993; De Miguel et al., 1997; Charlesworth et al., 2003). The significantly lower contents are documented for Pb whereas Fe and Mn concentrations are found to be substantially higher (Robertson et al., 2003; Loredo et al., 2003). Cr,

Tab. 1 Review of analytical techniques and detection limits

Chemical element	Way of decomposition	Analytical method	Analytical instrument	Detection limit	Units
As	HNO_3, HCl	HG-AAS	PE 3100 FIAS 100	0.1	$mg.kg^{-1}$
Cd	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	0.1	$mg.kg^{-1}$
Cr	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	5	$mg.kg^{-1}$
Cu	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	1	$mg.kg^{-1}$
Fe	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	0.01	%
Hg	-	AAS	TMA-254	0.03	$mg.kg^{-1}$
Mn	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	10	$mg.kg^{-1}$
Ni	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	1	$mg.kg^{-1}$
Pb	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	5	$mg.kg^{-1}$
Zn	HF, $HClO_4$, H_3BO_3, HCl	F-AAS	PE 3030B	1	$mg.kg^{-1}$

Tab. 2 Mineral composition of clayey street sediment sample fraction

Sample	Illite	Kaolinite	Chlorite	Smectite	Quartz	Calcite	Feldspar
1 NM	+	+	+	+	+	+	+
2 KN	+	+	+	–	+	–	+
3 HN	+	+	+	–	+	+	+
4 MN	+	+	+	–	+	+	+
5 RM	+	–	+	+	+	+	+
6 TM	+	+	+	+	+	+	+
7 KC	+	+	+	–	+	–	+
8 TC	+	+	+	+	+	?	+
9 PA	+	–	+	–	+	+	+

Cu a Ni seem to be the most variable parameters with markedly varying content levels in the street sediment samples (Banerjee, 2003; Charlesworth et al., 2003; Loredo et al., 2003).

The wide variability of chemical concentrations in urban street sediments was revealed and documented also by a comparison of our results with the previously realized British study (Robertson, 2004, Fig. 3). In general, the metal concentration data in the street sediment samples of presented study are higher. The most significant differences can be observed in the case of Fe and Mn whereas the slightliest are in the case of Pb and Cu. One of the possible explanations of observed variations is supposed to be the different way of sample decomposition performed in our study using the strong mixed acid digestion in contrast to foreign study proceeding using the weak 10 % nitric acid digestion.

From the point of view of chemical content levels as a function of particle size we can state that higher concentrations are found in finer fraction <0.125 mm. The most significant variations in chemical contents between 2 analysed fractions can be observed in the case of Cu, Fe, Mn and Zn. On the contrary, As and Cd feature very similar concentration levels in both fractions.

Comparing inner and outer city samples, in general no substantial variations can be noticed. Main differences are observed for Cr and Cu, which contents are markedly higher in the inner city samples, exceeding 100 and 200 $mg.kg^{-1}$, respectively. Mean concentration variations of other chemicals result mainly from their increased content levels, determined locally in single inner (Ni, Pb, Fe) and also outer city samples (Zn, see Tabs. 3 and 4). Interpreted spatial trend in chemical content variations is figured in Fig. 4. From the inner city part, following sampling sites are characterized by the higher metal contents: Kamenné námestie (Cr, Fe, Ni and Mn), Hodžovo námestie (Cu and Zn) and Nový Most (Pb and Zn). On the other hand the lowest concentrations of chemicals were determined in the case of Račianske mýto crossroad. From the outer city part sampling locations, Kukučínova is characterized by the slightiest differences in content levels of analysed chemicals (mainly Cr, Ni and Pb) in comparison with inner city samples.

The fact that localities with potentially highest traffic are relatively "clean" (e.g. Račianske mýto) and on the other hand those considered to be "cleaner" (e.g. Kukučínova) are characterized with higher metal contents, points out various genesis and manifold sources of urban street sediment material.

Based on linear correlation of chemical concentration data on urban street sediment samples of fraction <0.125 mm (see Tab. 6), the significant correlation coefficients were determined for following elements: Cr-Ni, Cd-Zn and Fe-Mn with Cr-Ni. No correlations at evaluated significance levels (<0.01 and <0.001) are observed in the case of As, Cu, Hg and Pb.

From the point of view of spatial distribution of analysed chemicals no significant differences in their content levels in the inner and outer city samples were documented. Additional and extended sample collection and analysis of street sediments, but also of other media like urban soils, are required to confirm introduced spatial trends in chemical distribution as well as to catch their concentration variations in time (continual research). Performance of such a research could contribute to identification of the main urban contaminants and possible detection and interpretation of their sources (anthropogenic/geogenic).

Conclusions

Street sediment samples collected in inner as well as outer city parts were submitted to granulometric, mineralogical and chemical composition analyses.

Grain-fraction analysis has revealed the predominance of sandy fraction and on the other hand relatively low rate of the finest, clay particles.

Tab. 3 Chemical concentration data for urban street samples of Bratislava – INNER CITY

Sample	Particle size	As	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn
1NM	<1mm	3.2	0.9	55.2	138.7	21.1	<0.03	378.2	27.5	276.5	366.5
	<0.125mm	3.1	1.4	103.8	215.5	27.5	0.03	593.4	40.5	272.8	367.3
2KN	<1mm	3.8	1.2	341.9	204.2	30.6	<0.03	577.7	150.1	34.4	222.4
	<0.125mm	3.7	1.1	416.2	267.2	42.1	0.05	773.1	185.5	48.0	302.4
3HN	<1mm	2.6	0.9	69.2	214.6	25.6	0.03	461.6	21.9	29.3	322.6
	<0.125mm	3.0	1.4	104.7	401.8	30.0	0.05	604.5	36.3	63.1	463.2
4MN	<1mm	3.0	0.9	51.3	201.2	26.9	<0.03	428.6	21.2	33.6	297.2
	<0.125mm	3.6	1.5	86.8	368.5	29.9	0.05	607.6	32.7	84.4	369.5
5RM	<1mm	3.0	0.6	48.5	278.9	21.8	<0.03	360.8	18.4	25.6	203.1
	<0.125mm	3.5	1.0	86.2	103.3	28.5	<0.03	564.1	32.3	52.7	342.9
6TM	<1mm	3.6	1.0	46.4	143.8	20.0	0.04	373.2	20.7	52.1	241.1
	<0.125mm	3.7	1.5	86.9	218.4	29.4	0.08	571.5	32.0	61.1	377.3
Mean	<1mm	3.2	0.9	102.1	167.6	24.3	0.02	430.0	43.3	75.2	275.5
	<0.125mm	3.4	1.3	147.4	291.7	31.2	0.05	619.0	59.9	97.0	370.4

Note: Concentration units in $mg.kg^{-1}$, for Fe in %.

Tab. 4 Chemical concentration data for urban street samples of Bratislava – OUTER CITY

Sample	Particle size	As	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn
7KC	<1mm	3.1	0.9	61.4	100.4	23.1	0.1	443.4	23.5	84.0	364.3
	<0.125mm	3.9	1.5	80.6	177.2	29.4	0.1	692.1	38.9	98.6	462.6
8TC	<1mm	2.8	1.0	50.0	88.5	19.0	<0.03	378.6	16.8	32.1	313.8
	<0.125mm	3.2	1.9	76.0	265.7	30.8	0.04	600.3	31.4	72.1	832.4
9PA	<1mm	3.0	1.0	60.1	130.0	19.6	<0.03	453.5	20.5	36.2	313.5
	<0.125mm	4.1	1.2	68.8	146.0	22.0	0.03	572.8	24.7	44.4	265.9
Mean	<1mm	3.0	1.0	57.1	106.3	20.5	0.05	425.2	20.3	50.8	330.5
	<0.125mm	3.7	1.5	75.1	196.3	27.4	0.06	621.7	31.7	71.7	520.3

Note: Concentration units in $mg.kg^{-1}$, for Fe in %.

Tab. 5 Review of concentration data on urban sediment samples from different studies

Study		Bratislava[1]		Riyadh[2]	Oslo[3]	Madrid[3]	Coventry[4]	Birmingham[4]	Manchester[5]	Mieres[6]
Analysed fraction		< 1mm	< 0.125mm	< 0.5mm	< 0.1mm	< 0.1mm	< 1mm	< 1mm	< 1mm	< 0.147mm
Analytical method		FAAS	FAAS	FAAS	ICP-MS	ICP-MS	AAS	AAS	FAAS	ICP-AES
As	Mean	3.1	3.5							68.8
	Min – Max	2.6 – 3.8	3.0 – 4.1							34.0 – 202.0
Cd	Mean	0.9	1.4	1.7	1.4		0.9	1.6		1.6
	Min – Max	0.6 – 1.2	1.0 – 1.9				0.0 – 8.9	0.0 – 13.1		0.7 – 2.3
Cr	Mean	87.1	123.3							41.1
	Min – Max	46.4 – 341.9	68.8 – 416.2							24.0 – 97.0
Cu	Mean	147.2	259.9		123.0	188.0	226.4	466.9	113.0	112.4
	Min – Max	88.5 – 214.6	146.0 – 401.8				49.3 – 815.0	16.4 – 6688.4	32.0 – 283.0	50.0 – 211.0
Fe	Mean	23.1	29.9						0.9	3.2
	Min – Max	19.0 – 30.6	22.0 – 42.1						0.3 – 1.7	1.8 – 6.4
Hg	Mean	0.03	0.05							3.1
	Min – Max	0.02 – 0.12	0.02 – 0.12							0.5 – 9.0
Mn	Mean	428.4	619.9		833.0	362.0			282.0	495.9
	Min – Max	360.8 – 577.7	564.1 – 773.1						49.0 – 433.0	421.0 – 547.0
Ni	Mean	35.6	50.5		41.0	44.0	129.7	41.1		25.8
	Min – Max	16.8 – 150.1	24.7 – 185.5				6.2 – 233.5	0.0 – 636.2		20.0 – 32.0
Pb	Mean	67.1	88.6	257.0			47.1	48.0	265.0	317.5
	Min – Max	25.6 – 276.5	44.4 – 272.8				0.0 – 199.4	0.0 – 146.3	25.0 – 645.0	143.0 – 618.0
Zn	Mean	293.8	420.4		412.0	476.0	385.7	534.0	653.0	420.4
	Min - Max	203.1 – 366.5	265.9 – 832.4				93.0 – 3038.2	81.3 – 3164.8	172.0 – 2183	197.0 – 1077.0

[1]Present study; [2]Ahmed & Al-Swaidan, 1993; [3]De Miguel et al., 1997; [4]Charlesworth et al., 2003; [5]Robertson et al., 2003; [6]Loredo et al., 2003.
Note: Concentration units in $mg.kg^{-1}$, for Fe in %.

Tab. 6 Correlations for chemical contents in Bratislava street sediment samples < 0.125 mm

	As	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn
As	1.000									
Cd	-0.289	1.000								
Cr	0.041	-0.394	1.000							
Cu	-0.606	0.073	0.090	1.000						
Fe	-0.159	-0.091	0.883*	0.330	1.000					
Hg	0.385	0.388	-0.030	-0.219	0.149	1.000				
Mn	0.182	-0.134	0.829*	-0.022	0.814*	0.418	1.000			
Ni	0.073	-0.372	0.997**	0.050	0.888*	0.014	0.860*	1.000		
Pb	-0.464	0.126	-0.155	-0.167	-0.184	-0.084	-0.107	-0.151	1.000	
Zn	-0.481	0.804*	-0.276	0.168	0.054	0.105	-0.087	-0.254	-0.015	1.000

Note:
* Correlation significance at $p < 0.01$.
** Correlation significance at $p < 0.001$.

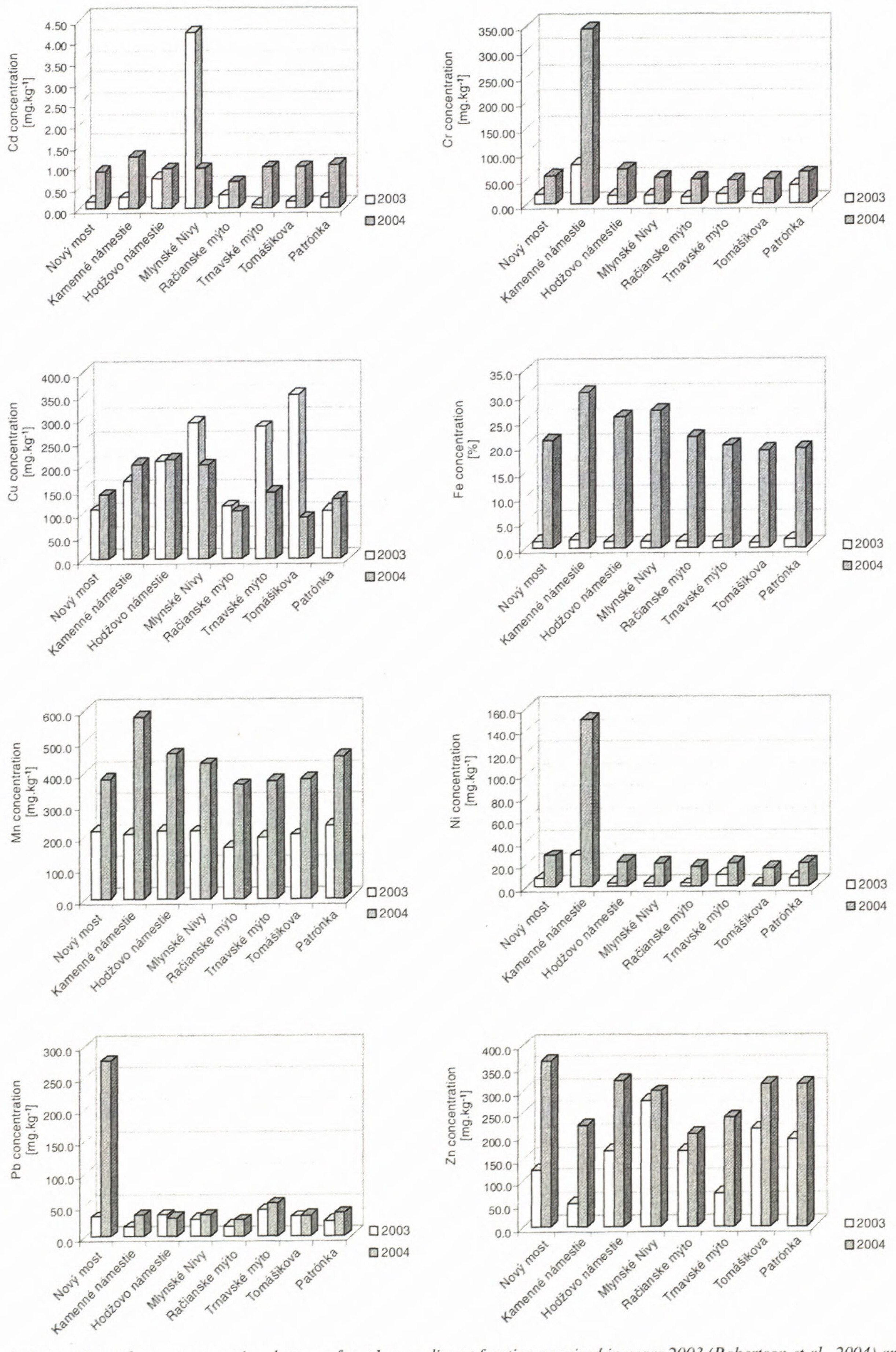

Fig. 3. Comparison of two concentration data sets for <1mm sediment fraction acquired in years 2003 (Robertson et al., 2004) and 2004 (presented study).

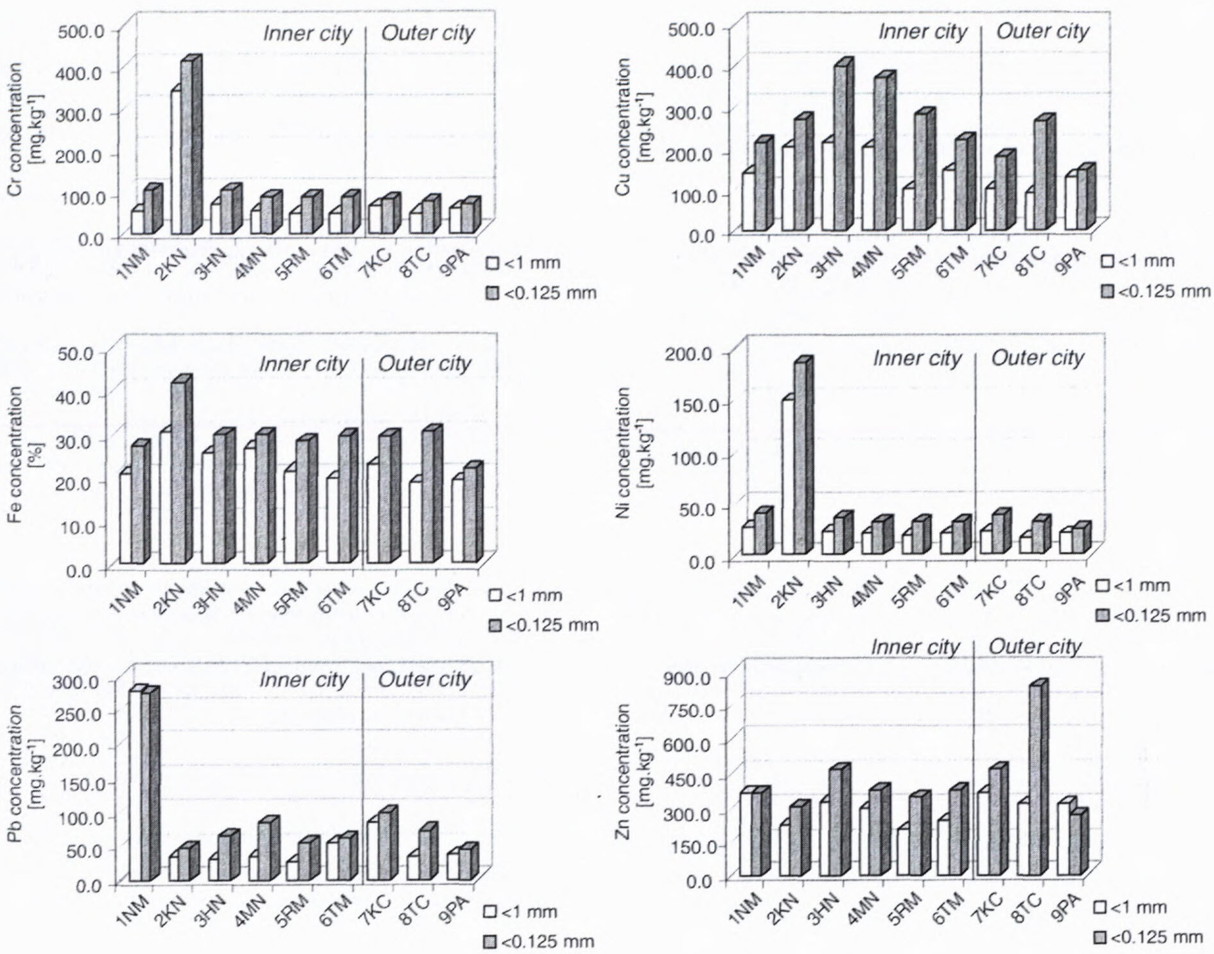

Fig. 4. Concentration variations of selected chemicals in inner / outer city samples.

Based on using X-ray powder diffraction method the following minerals were determined in the clayey fraction of collected samples: illite, chlorite, quartz and feldspar. The presence of other minerals like kaolinite, smectite and calcite is rather variable. No significant differences between inner/outer city samples were observed.

Chemical composition analysis has revealed the significantly lower Pb concentrations and on the other hand higher Fe and Mn contents in comparison with some similar foreign studies.

Higher chemical content levels were found in the finer analysed sediment fraction (<0.125 mm) with the most marked increase in the case of Cu, Fe, Mn and Zn.

Based on comparison of chemical composition determined in the inner and outer city samples, generally no substantial differences were documented. The higher concentrations in the inner city samples have been found mainly in the case of Cr and Cu. From the point of view of sampling locations the following inner city sites are characterized by the higher contents of some metals: Kamenné námestie (Cr, Fe, Ni and Mn), Hodžovo námestie (Cu and Zn) and Nový most (Pb and Zn).

The statistical concentration data evaluation has revealed the significant correlation of Cr with Ni, Cd with Zn and Fe and Mn with Cr and Ni. The least significant correlation has been documented for As, Cu, Hg and Pb.

Urban street sediments in Bratislava show increased concentration levels of some metals, e.g. Fe and Mn, together with relatively high variability in concentration levels of various potentially toxic elements (e.g. Cr, Cu). In the areas, at first sight looking as those most environmentally loaded, such as big junctions with the high traffic density, the highest metal concentrations were not always found there. On the other hand the relatively "clean" areas near pedestrian zone or in the outer city parts show the higher metal contents. This fact reveals a genesis variability of street sample material including local, regional and long-distance sources. Acquired first results point out those contents of potentially toxic elements in the urban street sediments in Bratislava which are not so high in comparison with those found in some other industrial centres and cities (e.g. Manchester).

The urban street sediments still should be the object of future study since they represent an important medium for assessment of quality of urban environment.

Analysis of their chemical composition provides basic information on character and level of urban pollution. However, from the point of view of related potential health risk assessment and its objective interpretation, the chemical analyses of fine, airborne and respirable particles, being potentially present in the road deposited sediments, are necessary.

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