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AIR POLLUTION STUDIES IN SLOVAKIA USING AEROSOL FILTERS AND BIOMONITORING TECHNIQUE

JANA MEREŠOVÁ[1], MATEJ FLOREK[1], KAROL HOLÝ[1], MIROSLAV JEŠKOVSKÝ[1], IVAN SÝKORA[1], CYRIL BURDA[2], TEREZIA MELICHEROVÁ[2], BLANKA MAŇKOVSKÁ[3], JÚLIUS OSZLÁNYI[3], MARINA V. FRONTASYEVA[4], SERGEY S. PAVLOV[4]

[1] Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University, Mlynská dolina, 842 48 Bratislava, Slovak Republic; e-mail: j.meresova@gmail.com
[2] Slovak Hydrometeorological Institute, Jeséniova 17, 833 15 Bratislava, Slovak Republic
[3] Institute for Landscape Ecology, Slovak Academy Sciences, Štefánikova 3, 814 99 Bratislava, Slovak Republic
[4] Department of Neutron Activation Analysis, Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, str. Joliot-Curie 6, 141980 Dubna, Moscow Region, Russia

Abstract

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Instrumental neutron activation analysis (INAA) and atomic absorption spectrometry (AAS) were employed in order to evaluate the concentrations of up to 36 chemical elements (heavy metals, rare earths, and actinides) in the atmospheric aerosols. Two sampling sites in Bratislava were examined. The first site, Líščie údolie, is quite a pristine location with low traffic density. The second sampling site is close to the crude oil processing plant Slovnaft. The influence of the steel industry in Veľká Ida and thermal power plant in Prievidza were investigated. The most heavily contaminated sampling site, in the vicinity of the Tušimice surface coal mine in the Czech Republic, was also included in this study. The levels of pollutant concentrations were compared to those in the atmosphere of another five European sites: Krakow (Poland); Budapest (Hungary); Ispra, Ponzone and Milan (Italy). Terrestrial moss *Pleurozium schreberi* and *Hylocomium splendens* were collected in the environs of the Slovnaft oil plant to monitor heavy metal atmospheric deposition. The elemental concentrations in moss samples were compared to the Slovakian median values and the Norwegian ones.

Key words: atmospheric aerosol particles, elemental composition, crustal enrichment factor, contamination factor

Introduction

Elemental concentrations of airborne particulate matter can provide important information on the degree of atmospheric pollution and further evaluation of the potential health risk to the population.

For this reason, it is necessary to know their chemical composition and physical characteristics in order to understand their behaviour and impact.

Among many pollutants heavy metals are the most toxic components for all living organisms. Some heavy metals play an important role in the nutrition of plants, animals and humans (Mn, V, Cr, Ni, Cu, Zn), but if they occur in excess, they may produce certain toxic effects. Such elements as Cd, Hg, Pb are toxic even in very low quantities. Heavy metals are present in the atmosphere in organic and also in inorganic compounds. They can be transported to large distances from the source and where they fall out they produce a very negative impact on the environment.

Heavy metals are released into the environment from a great number of sources. Combustion of fossil fuels is the principal anthropogenic source of Ni, V, Cd, As, and Zn (Pacyna, 1986). Pb, Sb, Br, Cr, and V are elements associated with automotive exhaust products and domestic heating. Non-ferrous smelters are the sources of Cu, Zn, Cd, and Pb. The largest source of airborne Cd in the environment is the burning of fossil fuels such as coal or oil, and incineration of municipal waste materials. Cd may also be emitted into the air from zinc, lead, or copper smelters. The current anthropogenic metal emissions are up to several orders of magnitude higher than natural contents (Chmielewska, Spiegel, 2003). Resuspended soil particles, volcanic aerosols and forest fires contribute to natural emissions of trace elements such as Cr, Mn, V, Cu, Mo, Ni, and Zn (Pacyna, 1998). It has to be noted, that Th and U are elements relatively abundant in the Earth's crust, and hence their concentrations in oil are significant. The data on elemental concentration in the Slovakian atmospheric aerosol particles are very scarce and limited to a few elements (Burda et al., 2006).

For more than three decades mosses have been used as biomonitors. This technique is presently widely accepted as a method to assess the atmospheric deposition of metals (Rühling, Steinnes, 1998; Smodis et al., 2004). In the Slovak Republic, extensive studies in this direction have been done by Maňkovská from year 1991 (Maňkovská, 1997). Mosses have only a rudimentary root system and readily take up elements from the atmosphere. Results from moss surveys are regularly published (every five years) in the Atlas of Heavy Metal Atmospheric Deposition in Europe by the UNECE ICP Vegetation (Buse et al., 2003; Harmens, Norris, 2008).

Instrumental neutron activation analysis (INAA) and atomic absorption spectrometry (AAS) are widely used methods to determine the contents of trace elements in airborne particulate matter and in mosses. Currently INAA is considered to be the most suitable and appropriate analytical technique for multi-elemental analyses of aerosol samples (Dams, 1992; Frontasyeva, 2006). The main advantages of this method are high precision, high selectivity and sensitivity, small sample quantity needed, direct non-destructive method, etc.

Material and methods

The main objective of this study was to determine the maximum possible number of elemental concentrations, including heavy metals, in the atmospheric aerosol particles at five sites with different anthropogenic impact. Two sampling sites are located in Bratislava: Líščie údolie and Slovnaft. The other two, Prievidza, and Veľká Ida, are in the polluted areas of Slovakia; the last one, Tušimice, is in the Czech Republic. In the vicinity of the refinery Slovnaft, spatial and temporal trends in heavy metal deposition were also studied using the moss biomonitoring technique.

Description of sampling sites

Bratislava is a city with a population of approximately 500, 000. The chemical industry, technical glasswork, building industry, incineration plant, and car industry are located in the larger city area. Neither a ferrous or non-ferrous smelter, nor a power plant is in the vicinity. The refinery and petrochemical company Slovnaft, one of the biggest in Central Europe, allocates in the area of 5.2 km^2 at the south-eastern border of the capital of SR Bratislava. Annually, Slovnaft processes approximately 5 million tons of crude oil supplied mainly from the Russian Federation. Slovnaft delivers to the market a complete range of refinery, petrochemical products and plastics.

The Prievidza station (48°46' N, 18°37' E) is located in the town centre, close to four–storey residential houses and buildings of similar height. Near the station, passes slight traffic. Close to the city of Prievidza is the Nováky mine, with annual coal production of 1900 thousand tons. Coal is predominantly burned (85%) in the Nováky thermal power plant, with 518 MW of power capacity. Also a chemical industry is located in the town of Nováky.

The Veľká Ida station (48°35'N, 21°10' E) is located in the southeastern part of Veľká Ida municipality, in a relatively open area. In the vicinity of the station are located family houses, gardens, railway stations and waste dumps of slag, which are not fully grassed. Close to the town of Veľká Ida is the largest ferrous metallurgy complex in Slovakia – U.S. Steel Košice.

The Czech sampling site Tušimice is located in the meteorological station of the Czech Hydrometeorological Institute (50°22' N; 13°20' E) on open land with low building density. The area is affected by operation of the surface coal mines. In 1998, the Tušimice-I thermal power plant was terminated. The Tušimice-II power plant is working with a power capacity of 4x200 MW.

Aerosol and moss sampling

Sampling of atmospheric aerosol particles was performed in the period 2004–2006. Nitro-cellulose membrane filters PRAGOPOR were used in the Líščie údolie and Slovnaft sites (collection efficiency ~ 100%). About 3000 m3 of air was pumped through one individual sample. In other sampling stations, glass-fibre filters MILLIPORE were used and the volume of air sampled was about 55 m^3. Therefore, the uncertainties of the results for these particular sites are quite high.

The terrestrial moss samples of Pleurozium schreberi and Hylocomium splendens were collected in the environs of the Slovnaft oil plant to monitor the atmospheric deposition of pollutants. Sampling was carried out according to guidelines of the UNECE ICP Vegetation survey in 2000 and 2006 (Harmens et al., 2008).

Measurement of elemental concentrations

Instrumental neutron activation analysis (INAA) at the IBR-2 reactor in the Joint Institute for Nuclear Research, Dubna was used for the determination of elemental concentration in aerosol filters and moss samples. The irradiation facility is described elsewhere (Frontasyeva et al., 2006). The contents of some environmentally meaningful elements such as Ni, Cu, Zn, Cd, and Pb, not detectable by INAA, were determined using AAS at the Institute of Geology, Comenius University (Medveď et al., 2003). The concentrations of Hg were measured by the Trace Mercury Analyzer TMA-254.

Results on elemental concentrations in atmospheric aerosol

Atmospheric concentration levels for four Slovakian and one Czech sampling sites are presented in Table 1. For most elements the results were obtained for the first time. Our results may be considered as the representative and unique data on elemental concentrations in the atmospheric aerosol for the investigated locations.

Crustal enrichment factors

The concept of enrichment factors (EF) was introduced by Rahn (1971) to detect contributions of non-crustal sources on observed concentrations of elements. EF compares the ratio of the concentration of element *c(x)* in question to that of a selected reference element *c(Al)* in a sample, and the corresponding ratio in the average composition of the Earth's crust.

$$EF = \frac{\left(c(x) / c(Al) \right)_{Sample}}{\left(c(x) / c(Al) \right)_{Crust}}$$

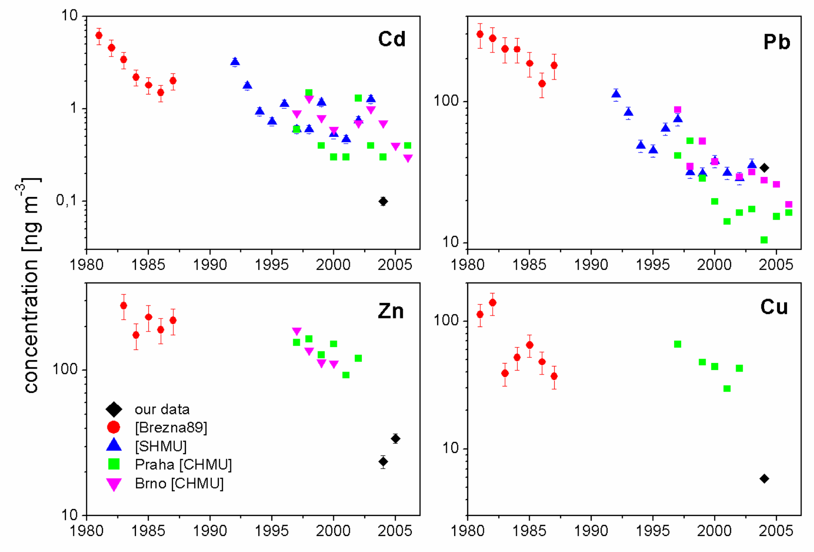

Fig. 1. Temporal variations of atmospheric concentrations of Cd, Pb, Zn, and Cu in Bratislava; circle – (Brežná, Závodský, 1989), triangle – (SHMU), diamond – our data. For comparison, elemental concentrations in Prague (square) and Brno are also shown (reverse triangle) (CHMU); ($ng \cdot m^{-3}$).

Aluminium (Al) was used as a soil (crust) reference element. Also Ti is sometimes used as reference element. The average elemental concentrations in humus horizon of soil for Slovakia are used in the EF calculations (Čurlík, Šefčík, 1999).

Enrichment indicates natural volatilization, marine sources and anthropogenic activities. Rahn (1976) suggested classification criteria of EF: if EF < 7, then air particulate is of crustal

T a b l e 1. Atmospheric concentrations AC and enrichment factors EF for four sampling sites in Slovakia and one in Czech Republic; (ng m^{-3}).

Location	**Líščie údolie**		**Slovnaft**		**Velká Ida**	**Prievidza**	**Tušimice**
	AC	**EF**	**AC**	**EF**	**AC**		
Na	104	3.8					
Al	189	1	184	1			
Cl	49						
K	195	3.6					
Ca	179	9.1					
Sc	0.0320						4.6
Ti	7.8						
V	0.83						
Cr	1.1	4.2	2.8	**11**	1.1	1,2	150
Mn	4.9	2.2					13
Fe	252	3.0			643	435	
Co			0.3		0.42	0.19	1.37
Ni	0.45	5.6	5.1	**65**	1.6*	1.0*	4,4
Cu	8.0	**150**	41	**770**	18	21	9.0
Zn	28	**150**			1294	1515	26122
Ga	0.02	0.52					
As	0.3	13			1.7*	7.9*	1.5
Se	0.42	**400**				0.18	
Sr			3.1				480
Br	3.5						2.5
Cd	0.11	**120**	2.2	**2350**	1.1*	0.4*	7.1
In	0.001				0.045	0.042	
Sb	1.0	**460**			1.37	5.5	2.4
I	0.66					0.44	
Cs	0.045	2.8			0.028	0.032	1.9
Sm	0.012				0.84	0.89	0.5
Dy	0.01						
Tm	0.1					0.016	
W	0.22						
Hg	0.064	**250**					
Pb	22	**350**	42	**670**	90	29	45
Th	0.042	1.4					1.4
U	0.012	1.2					0.2

* Data taken from Burda et al. (2006)

origin and if EF > 10, then it is of the anthropogenic one. Unfortunately, neither Al, nor Ti was measured in aerosol samples, except the Líščie údolie and Slovnaft sites in Bratislava. EF's were calculated for these two locations, considering Al as the reference element. Results are presented in Table 1. The values of EF for the Líščie údolie site indicate the soil origin of Mn, Fe, Cs, Th and U. Following Rahn's criteria, one can conclude that Cu, Zn, As, Se, Cd, Hg, and Pb are of anthropogenic origin.

Fig. 1 shows a decreasing trend of air pollution by heavy metals in Bratislava since 1981. The emissions of Pb have decreased, reflecting the shift from leaded to unleaded gasoline. A further explanation of this decreasing trend is the declination of industry in the Slovak Republic after 1989, since the fuel burning processes in thermal power plants and industry factories are a major source of atmospheric pollution with heavy metals. The emissions of pollutants were reduced also via utilization of more rigid requirements in the environmental legislation and employment of new more effective filtration techniques.

For several environmentally significant elements (Cr, Fe, Ni, Cu, Zn, As, Cd, and Pb) our results are compared to relevant data from some European cities, as can be seen in Figs 2 and 3. Ponzone (Rizzio et al., 2001) is a small town where the major part of wool industries is settled, and Ispra (Rizzio et al., 1999) is a residential settlement in northern Italy. Milan (Gallorini et al., 1999) is the industrial centre of northern Italy; thus concentrations of almost all elements are obviously the highest in relevant aerosols. The main objective of the Krakow research study (Wróbel et al., 2000) was to determine the contribution of traffic to the particulate air pollution, and to characterize the transport of aerosols in an urban area

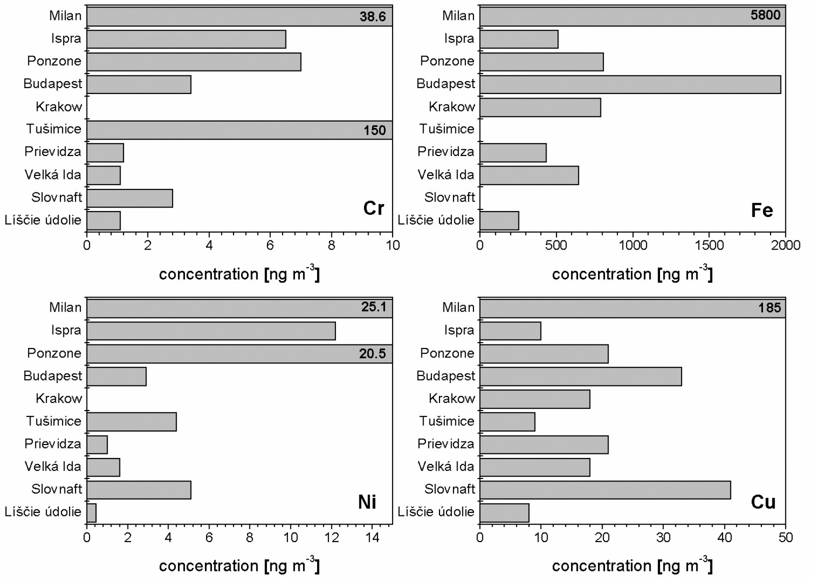

Fig. 2. Atmospheric concentrations of Cr, Fe, Ni, and Cu for 10 European sampling sites (ng·m^{-3}).

close (5 m) to the main road. Also, the location of Széna square in Budapest (Salma et al., 2002) has a more closed downtown character and is affected by heavy traffic.

The concentrations of almost all elements are lower in Bratislava – Líščie údolie compared to the other localities, or comparable to the Italian sites of Ponzone and Ispra, which are typically low pollution areas. The low-level atmospheric pollution in Bratislava may be caused by a small number of pollution sources, and in particular by the high number of windy days per year, typical for this location. This statement is supported by the negative correlation found between the wind velocity and the elemental concentrations in our samples (Merešová et al., 2008). Moreover, in this article, seasonal variations were discussed.

On the contrary, the Slovnaft site is affected by the oil processing plant. The concentrations of elements typical for oil combustion like Ni and Cd are elevated and Cr, Pb, and Cu reach high levels, as well. Unfortunately, we have no results for V, since increased concentrations would also be expected.

The comparison of two sampling sites, Tušimice and Prievidza, is very interesting since both are affected by the mining and combustion of coal in the power plants. The Tušimice site is the most heavily polluted area in this study. Mining operations in the surface coal mines raise atmospheric concentrations of Cr, Zn, and Cd. The concentration of U is one order of magnitude higher in Tušimice than in Bratislava. The coal mines near Prievidza are underground; therefore the concentrations of pollutants are lower compared to the Tušimice data, but still are higher than in Bratislava. Particularly high concentration was measured for As. Results of Keegan et al. (2002) showed that the mean concentration of As in coal burnt at the Nováky power plant is 518 $\mu g \cdot g^{-1}$; the highest observed concentration

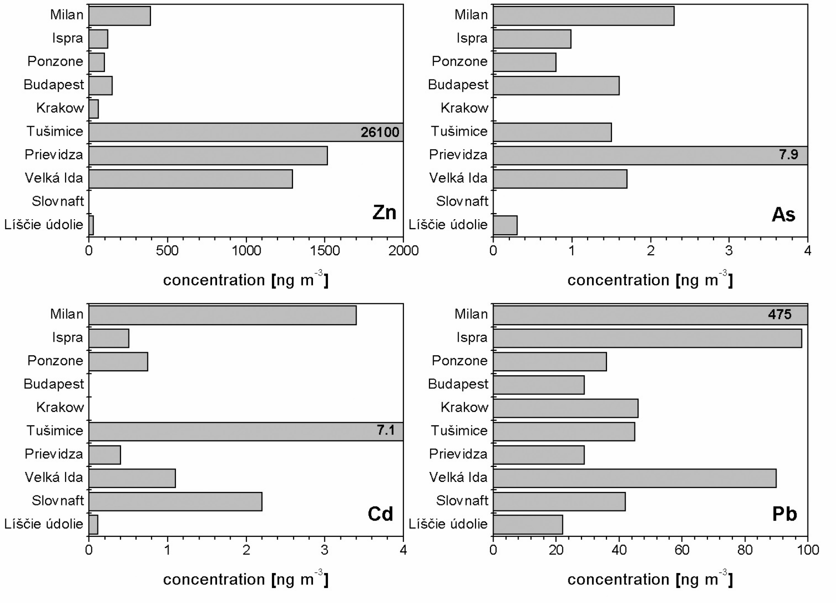

Fig. 3. Atmospheric concentrations of Zn, As, Cd, and Pb ($ng \cdot m^{-3}$) for 10 European sampling sites ($ng \cdot m^{-3}$).

was 1540 μg·g^{-1}. This compares to the normal range of As in British black coal of 2-10 μg·g^{-1}, or US coal, where action was taken if the As concentration exceeds 5 μg·g^{-1} (Swaine, 1994; Goldhaber et al., 2000).

In the data-set from the Veľká Ida site, we observed high concentrations of Fe, Pb, and Zn, which are the elements emitted into the atmosphere during smelting processes. Thus the impact of nearby U.S. Steel is obvious.

Results on biomonitoring survey

The concentrations of 41 elements were determined in moss samples collected in 2000 and 2006 from several locations in the vicinity of the refinery Slovnaft. The spatial and temporal trends in elemental deposition were studied. Results include heavy metals, non-ferrous-metals, halogens, and rare-earths, as well. Five locations with different distance from the refinery were investigated in 2000, and three more sites in 2006.

Contamination factor

In order to evaluate the level of pollution in the vicinity of the Slovnaft refinery, the concentrations of several elements were compared with representative background concentrations. Usually the median concentrations from Norwegian locations are used as reference background values corresponding to a pristine undisturbed environment (Steinnes et al., 2001). In Fig. 4 the contamination factors for each particular site (labelled by its distance from the refinery; km) are presented. The contamination factor is calculated as a ratio of Slovak to Norwegian concentration. For most of the elements at all the sampling sites the contamination factors are higher than one. Moreover, concentrations of some elements measured in Slovak mosses (Al, V, Cr, Fe, Co, Sr, Cd, Sb and U) are 10-times higher compared to background values.

In the UNECE ICP Vegetation survey (UNECE ICP 2003), Slovakia is classified as a rather polluted European country. The main sources of heavy metal emissions are the industrial centers of Hornonitrianska kotlina basin, Žiarská kotlina basin, povodie Váhu (Váh valley) in the west, and Central Spiš, towards the east (UNECE ICP 2003). The previous moss biomonitoring study of 86 Slovak sites (Maňkovská et al., 2003; Florek et al., 2007) revealed areas known by previous or present mining and processing of non-ferrous metals (Volovské vrchy in the Slovenské rudohorie Mts, Kremnické and Štiavnické vrchy Mts). In the north-northwest border areas of Slovakia, the elevated concentration of Hg was determined. Most probably, it reflects the long-range transport form the Katowice-Ostrava region also known as the 2nd Black Triangle. A huge amount of coal is mined there and, accompanied by metallurgical, chemical industry, and mechanical engineering, significantly influence the environment.

The course of prevailing winds in Bratislava mostly determines the effect of the Slovnaft refinery. The east-southeast areas are the most affected by operation of the refinery. In remote sites (Záhorie, Jabloňové, Rusovce and Čunovo), the contamination factor is usually markedly lower as the ratio of medians (Fig. 4.) indicating lower pollution. Concentrations of Al, Cr, Fe, As, Sb, and Th in moss samples collected on these sites are close to the Slovak median values. Only the values of V, Co, and U demonstrate distinct increase.

Low concentrations of a specific group of heavy metals (Cu, In, Cd, and Sb) in the close vicinity of the refinery are naturally lower, as the particulates containing these elements are easily transported with wind outside the territory of the plant. Their deposition at the territory of the refinery is comparable to the Slovak median values. Moreover, pollution with toxic elements, like Hg and Pb, is lower than the Slovak median.

Total annual deposition

Since 1990, the moss biomonitoring method is widely used in Europe for deposition evaluation purposes. Suchara and Sucharová (1999) proposed the method for total annual deposition D (mg m^{-2} y^{-1}) calculation. Table 2 presents the assessment of the total annual deposition D for the Rusovce sampling site. Also minimum and maximum depositions, determined for 86 Slovak sites in the previous study (Florek et al., 2007), are given in Table 2. The elemental depositions at the Rusovce site are within the range of the Slovak values, except the U, where the deposition in Rusovce is three times higher than the maximum deposition of U in Slovakia.

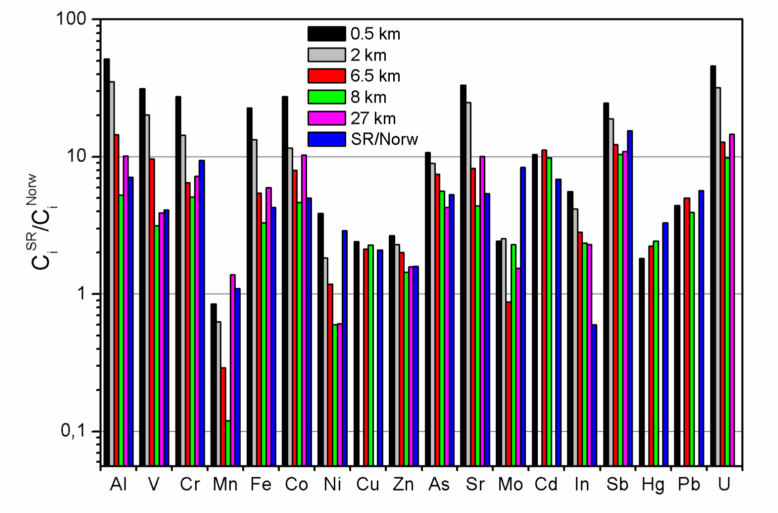

Fig. 4. Contamination factors of some elements in moss samples from vicinity of the SLOVNAFT refinery in 2000. The Norwegian concentrations are taken as reference background values. The sampling sites are labelled by their distance from the refinery (km). The last bars correspond to the concentration ratio of Slovak median and background (C_i^{SR}/C_i^{Norw}).

Comparison of pollution in 2000 and 2006

The same five sampling sites were studied and three more sites were included in the investigation in 2006. The new sites are more distant from the refinery. Fig. 5 shows relative concentrations (C_i^{2006}/C_i^{2000}) for one neighbouring site (~ 0.5 km) and the Rusovce site (~ 6.5 km).

The ratio for most of the elements is less than one, indicating the decline of pollution. The concentrations decreased for approximately 40%, except for Sc, Ni, Se, and the lanthanide group (La, Ce, Sm, Tb), where the double concentration in 2006 in close vicinity of the refinery was observed. The concentrations of lanthanides depend strongly on the oilfield's location. There is a distinct variance between the oil from the Russian Federation and from the Middle East.

T a b l e 2. Assessment of total annual depositions D (mg m^{-2} y^{-1}) for certain elements at the Rusovce site and minimal and maximal depositions from previous study (Florek et al., 2007).

	Rusovce	Slovakia			Rusovce	Slovakia	
Element	**D**	**Dmin**	**Dmax**	**Element**	**D**	**Dmin**	**Dmax**
Al	937	140	3200	**Ni**	0.33	0.18	3.3
As	0.37	0.12	800	**Pb**	3.24	1.2	14
Cd	0.21	0.02	0.3	**S**	1280	500	1400
Co	0.35	0.08	2.1	**V**	3.37	0.45	7.8
Cr	0.84	0.2	7.9	**Th**	0.13	0.03	0.9
Cu	2.9	1.3	12	**U**	0.33	0.005	0.1
Fe	426	92	2900	**Zn**	13.7	4	31

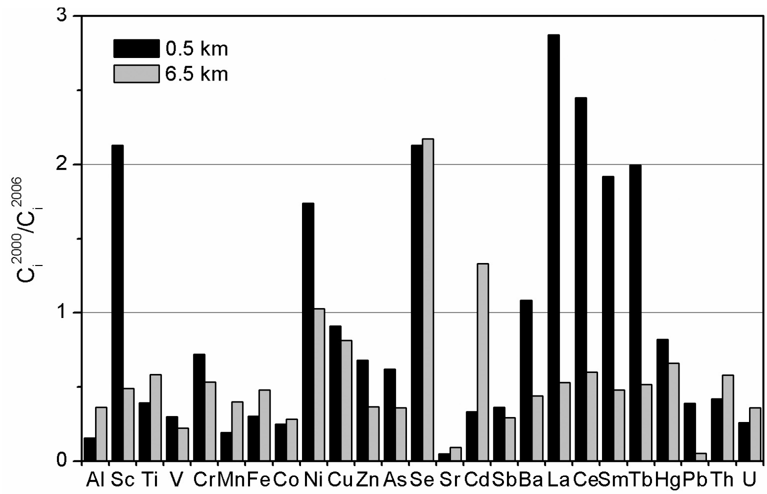

Fig. 5. Comparison of pollution in 2000 and 2006 in vicinity of the Slovnaft oil refinery based on moss biomonitoring method.

Conclusion

Data on the atmospheric concentrations of a large set of elements including heavy metals, halogens, rare earths, uranium, and thorium in particulate matter were obtained by INAA and AAS methods. Four Slovakian and one Czech sampling sites were investigated in this study. The following points can be concluded:

1. Decreasing trend of air pollution by heavy metals (Cd, Pb, Zn and Cu) in Bratislava since the 1981 was established.
2. Atmospheric concentrations of elements typical for oil combustion, such as Ni and Cd are elevated in close vicinity of the Slovnaft refinery. Concentrations of Cr, Pb and Cu are also increased. It is a clear consequence of the oil processing.
3. Crustal enrichment factors calculated for Cu, Zn, As, Se, Cd, Hg and Pb at sites in Líščie údolie and Slovnaft indicate anthropogenic impact.
4. High concentration of As was measured in Prievidza, corresponding to extremely high content of As in coal mined and burned in this area.
5. Impact of nearby U.S. Steel smelters in site Veľká Ida is obvious, since, elevated atmospheric concentrations of Fe, Pb and Zn were measured.
6. Significantly increased airborne pollution (Cr, Zn, Cd and U) was observed in the area of Tušimice affected by the operation of surface mine and combustion of coal in the Tušimice-II thermal power plant.

The following conclusions can be drawn based on the results of moss biomonitoring in the vicinity of the Slovnaft refinery:

1. Significant excess of pollutants in close vicinity of refinery was observed. Concentrations of some elements (Al, V, Cr, Fe, Co, As, Sr, Cd, In, Sb, Sc, Cs, Th and U) in moss samples are 10-times higher in comparison to the Norwegian background values. Concentrations of Al, V, Cr, Fe, Co, Sr, Sc, Th and U are higher than the Slovak median.
2. Low concentrations of some heavy metals (Cu, In, Cd and Sb), comparable to Slovak medians, were determined in samples from the closest site.
3. Comparison of results from 2000 and 2006 indicates 40 % decrease of pollution for most of the elements.
4. For the Rusovce site total annual depositions of several elements were estimated according to concentrations of relevant elements in moss samples. Except for U, depositions of all elements are within the range of typical elemental depositions in Slovakia.

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