Human Health Risk Assessment of Heavy Metals Bound on Particulate Matter

Drahoslava ZMIJKOVÁ[1)], Miroslav KOLIBA[1)], Konstantin RACLAVSKÝ[2)]

[1)] VSB – Technical University of Ostrava, Faculty of Mining and Geology, Institute of Environmental Engineering, 17. listopadu Street 15, 708 33 Ostrava–Poruba, Czech Republic; email: drahoslava.zmijkova.fmmi@vsb.cz, miroslav.koliba@vsb.cz
[2)] VSB – Technical University of Ostrava, Faculty of Mining and Geology, Centre ENET, 17. listopadu Street 15, 708 33 Ostrava–Poruba, Czech Republic; email: konstantin. raclavsky@vsb.cz

http://doi.org/ 10.29227/IM-2018-01-15

Abstract

The samples of particulate matter (PM_{10}) were collected at five sampling sites of various character situated in the Moravian-Silesian Region (Czech Republic). Concentrations of heavy metals bound to particulate matter were determined by the method of ICP – optical emission spectroscopy. The contamination of heavy metals was expressed as the pollution load index – PLI. Human health risk assessment was performed by U.S. EPA method – using the hazard quotient – HQ, hazard index – HI, and excess lifetime cancer risk – ELCR. The pollution and resulting carcinogenic risks increase during the winter season. The potential non--carcinogenic risks are not significant.

Keywords: air pollution, particulate matter PM 10, heavy metals, hazard quotient, hazard index, cancer risk, pollution load index

Introduction

Particulate matter (PM) represents a significant factor of the human environment from many points of view. One of these aspects is that these particles contain many harmful substances in their structure. These components of PM can be associated with many negative effects on human health. Noxious substances bound on PM very often include heavy metals (Blažek et al., 2012; Borm and Donaldson, 2012).

The toxic effects of this group of noxious chemicals can be divided into two main categories – i. e. carcinogenic impacts and non-carcinogenic results. These two types of heavy metal toxic effects were studied in a case of heavy metals contained in PM_{10} collected in the Moravian-Silesian Region. The main reason of this investigation is a high degree of inhalation exposure of the Moravian-Silesian population to the heavy metals present in the structure of PM_{10} and its potential adverse impacts on morbidity and mortality within the Moravian-Silesian Region (Čupr et al. 2013).

The Moravian-Silesian Region belongs among the zones with the highest rate of atmospheric dusty pollution in the European Union. In this region, the air pollution is a result of a combination of many factors. The major part of ambient air PM_{10} in the Moravian-Silesian Region originates from metallurgy, thermal power plants, heat stations using fossil fuels and from chemical industry. The significant part of PM_{10} is also released from the surfaces of the dumps and piles, generally from the surface of burning dumps. Another very significant source of total suspended particles is road traffic. Car transport release the primary particles and at the same time, it is a rich source of resuspended dust. Other important source of PM are local boilers. Besides the facts discussed above, it is necessary to mention also a contribution of cigarette smoke. All earlier mentioned sources of PM are simultaneously principal contributors of heavy metal atmospheric contamination or pollution and this is the reason why suspended particles sampled in the Moravian-Silesian Region usually contain high amount of heavy metals in their structure. On the top of it, the dusty pollution can be aggravated due to bad meteorological conditions like windless weather, lack of rainfall or snowfall and western or south-western winds. The geographical position influences, together with atmospheric contamination, influences a predisposition to smog formation in the winter period with deteriorated dispersion of contaminants and pollutants including PM_{10} (Blažek et al., 2008). For these reasons, it is necessary to monitor and study air contamination and its potential health effects.

Materials and methods

The estimation of contamination and human health risk assessment of heavy metals bound on PM_{10} had three parts: 1. work in field – i.e. sampling procedure; 2. laboratory work – i.e. chemical analysis; 3. assessment of laboratory data – i.e. the calculation of the pollution load index and health risks.

Sampling sites, ambient air particle sampling and sample preparation

The area of investigation – i.e. Moravian-Silesian Region – is situated in the east part of the Czech Republic. It is surrounded by mountains in the west, in the east and in the south. The Moravian-Silesian Region

Fig. 1. Geographical location of sampling sites in the Moravian-Silesian Region (1. Ostrava-Radvanice, Nad Obcí 2859/1; 4. Ostrava-Poruba, Opavská 60/121; 24. Frýdek-Místek, Street K. H. Máchy; 31. Čeladná, Mountain Hotel Čeladenka; 36. Hradec nad Moravicí, Street Opavská)

Rys. 1. Lokalizacja geograficzna miejsc pobierania próbek w Regionie Morawsko-Śląskim (1. Ostrava-Radvanice, Nad Obcí 2859/1; 4. Ostrava-Poruba, Opavská 60/121; 24. Frýdek-Místek, Street K. H. Máchy; 31. Čeladná, Mountain Hotel Čeladenka; 36. Hradec nad Moravicí, Street Opavská)

has a moderate climate with predisposition to form inversions in the winter period of the year. As discussed above, the Moravian-Silesian Region is a typical industrial area with predominance of metallurgy. In the concerned region, one tenth of population of Czech Republic lives. In the area of the Moravian-Silesian Region, five sampling sites of different nature were chosen (Fig.1).

The first one – i.e. Ostrava-Radvanice – is an industrial area with very significant load of metallurgy as a result of its proximity to the metallurgical plant ArcelorMittal Ostrava, a.s. The second site – i.e. Ostrava-Poruba – represents a zone with a high intensity of traffic load; this sampling site is situated close to a road with traffic density of approximately 5,000 vehicles per day. The third sampling site – that is Hradec nad Moravicí – was chosen with regard to predominance of influence of many local boilers on the ambient air quality. The last but one locality of interest Frýdek-Místek was selected because of its ambiguous character. In Frýdek-Místek, it is necessary to count with combination influence of transport, local burning processes and industry, but at the same time, none of them is dominant. The last sampling area – Čeladná - represents a site without all influences discussed earlier. This sampling point is situated in the vicinity of the Mountain Hotel Čeladenka. At each sampling site, the sampling process was realized during the summer and winter period of the year 2014 in order to distinguish the contributions of industry and local heating processes to negative environmental impacts and human health risks.

For the purpose of collecting size-segregated PM – i.e. PM_{10} – in all the sampling sites mentioned above, the High-Volume Cascade Sampler (Diggital MD 05) was used. The chosen size fraction of PM was sampled at nitrocellulose filters. Before initial and final weighing, all sampling mediums were stabilized in a chamber with constant temperature and humidity. During transport, the filters were wrapped in alumina foil.

Chemical analysis

Before chemical analysis, each of the samples was weighed. The used nitrocellulose filters with collected samples were cut into fragments. The filter fragments were decomposed in the mixture of HCl, HNO_3, HF and H_2O_2. The concentration of the selected heavy metals (As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, V, Zn) was measured using the method of ICP optical emission spectrometry.

Environmental contamination assessment

The heavy metal contamination of ambient air in the Moravian-Silesian Region was estimated using a modification of the method according to Cai at al., 2015 and Liu at al., 2015. This method is based on calculating the pollution load index (PLI). This technique is dependent on ambient air volume concentrations of heavy metals. The following system of equations was applied to obtain the values of PLI in the atmosphere.

$$CF_i = c_i / c_{0i} \quad (1)$$

$$PLI = (CF_1 \text{ x } CF_2 \text{ x } ... \text{x } CF_n)^{1/n} \quad (2)$$

where CFi is a contamination factor for metal i, ci and c0i represent the heavy metal concentration in the ambient air of observed locality and in the atmosphere of background site. In the case of this study, the sampling site Čeladná was a reference background. The results were compared to the benchmark levels. The values of PLI bellow 1.0 are considered as contamination or no pollution, while PLI exceeding 1.0 is seen as pollution with three levels of intensity (PLI from 1.0 to 2.0 is considered as slight pollution, PLI between 2.0 and 3.0 is scored as moderate pollution, and PLI above three represents a heavy pollution) (Cai et al., 2015; Liu et al., 2015).

Exposure assessment (respiratory pathway)

The U.S. EPA evaluation method (U.S. EPA, 2000) was applied to estimate the rate of human heavy metal exposure through the main exposure pathway – i.e. inhalation. The exposure assessment takes the miscellaneous exposure scenarios into account. In this case, four following scenarios of exposure to heavy metals were evaluated – i.e. adult resident, adult tourist, child resident and child tourist.

According to this method, the chronic daily intake of heavy metals in PM_{10} via the respiratory tract (CDI_{inh}, [mg/kg/day]) is defined through the following equation:

$$CDI_{inh} = (CA \text{ x } IR \text{ x } EF \text{ x } ED \text{ x } ET) / (BW \text{ x } AT) \quad (3)$$

where CA [mg/m³] is an air concentration of heavy metal i in locality i, IR [m³/day] is a rate of inhalation, EF [day/year] is an exposure frequency, ED is an exposure duration [years], ET [hours/day] is an exposure time, BW is a body weight [kg] and AT [days] is an averaging time (U. S. EPA, 2014).

In the case of non-carcinogens, the chronic daily intake through the inhalation (CDI_{inh}) is called the average daily exposure dose (ADD_{inh}). For carcinogens, a term the lifetime average daily dose ($LADD_{inh}$) is used. The same units as for CDI_{inh} are used for $LADD_{inh}$ and ADD_{inh} (Chen et al., 2016; Hu et al., 2012).

Human health risk assessment

The human health risk assessment through inhalation is based on exposure estimation at all sampling sites. For the estimation of non-cancer risk of heavy metals bound in PM_{10}, the hazard quotient (HQ_{inh}) and hazard index (HI_{inh}) were used. The hazard quotient (HQ_{inh}) can be applied only for risk evaluation of respiratory exposure to one heavy metal, while the hazard index (HI_{inh}) as a sum of risks of multiple heavy metals reflects the negative health impacts of all heavy metals bound in the structure of PM_{10} for one sampling site (U.S. EPA, 1986; U.S. EPA, 2000; U.S. EPA, 1992; U.S. EPA, 2007; U.S. EPA, 2006; U.S. EPA, 2014).

The possibility of carcinogenesis was identified by means of the excess lifetime cancer risk ($ELCR_{inh}$). The equivalent of hazard index (HI) for carcinogens is called the total excess lifetime cancer risk ($ELCR_{inh,tot}$). The hazard quotient (HQ_{inh}), hazard index (HI_{inh}) and excess lifetime cancer risk ($ELCR_{inh}$) were quantified by the following group of equations.

$$HQ_{inh} = ADD_{inh} / RfD_{inh} \quad (4)$$

$$HI_{inh} = HQ_1 + HQ_2 + \ldots + HQ_i \quad (5)$$

$$ELCR_{inh} = LADD_{inh} \text{ x } CSF_{inh} \quad (6)$$

$$ELCR_{inh,tot} = ELCR_1 + ELCR_2 + \ldots + ELCR_i \quad (7)$$

where ADD_{inh} is an average daily dose for inhalation for heavy metal i, RfD_{inh} is a reference dose for heavy metal i through respiratory pathway, HQ_i is the hazard quotient for heavy metal i, $LADD_{inh}$ is the lifetime cancer risk for inhalation and for heavy metal i, CSF_{inh} is the cancer slope factor for heavy metal i through inhalation and $ELCR_i$ is the excess lifetime cancer risk for heavy metal i (U. S. EPA, 2014).

Results and discussion

Environmental contamination estimation

The values of pollution load index (PLI) ranged from 6.42 to 24.80 in the winter season, and from 0.84 to 2.74 in the summer period. After comparison with benchmark levels (Li et al., 2015; Tomlinson et al., 1980), it was found that in the Moravian–Silesian Region there is a heavy ambient air pollution during winter. In the summer period, the situation is better – i.e. no pollution (locality Hradec nad Moravicí), slight air pollution (Ostrava-Poruba) and moderate atmospheric pollution (localities Frýdek-Místek and Ostrava-Radvanice) was determined. The high values of the pollution load index (PLI) are probably connected with the beginning of the heating season and with the bad dispersion of air contaminants and pollutants. The highest values of PLI were determined in the industrial locality Ostrava-Radvanice in the winter season (PLI = 24.79). The values of PLI detected for the locality with high density of traffic (Ostrava-Poruba) and for the sampling site with ambiguous character (Frýdek-Místek) were very similar during winter season (PLI = 7.18 and PLI = 7.49), while in summer, there was a higher variation between their values of PLI (PLI = 2.74 versus PLI = 1.03). High PLI values were also calculated in the case of Hradec nad Moravicí (PLI = 6.41 for the winter period).

Human health risk assessment

Substantial differences were found between the values of hazard index for respiratory pathway (HI_{inh})

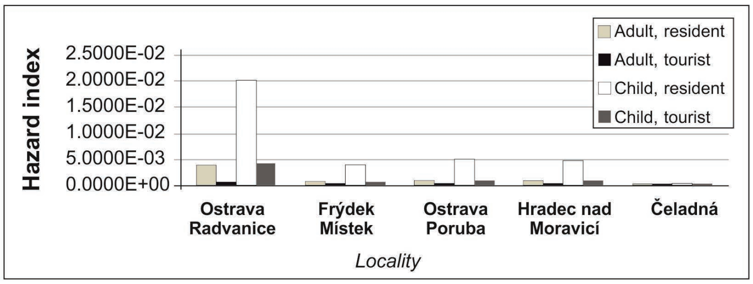

Fig. 2. Hazard index for all sampling sites during the winter season

Rys. 2. Indeks zagrożenia dla wszystkich próbek w sezonie zimowym

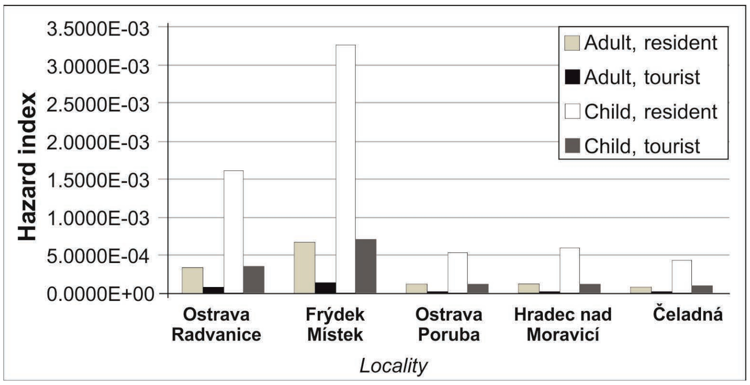

Fig. 3. Hazard index for all sampling sites during the summer season

Rys. 3. Indeks zagrożenia dla wszystkich próbek w sezonie letnim

depending on various exposure scenarios and the time of sampling. In the case of scenario adult, resident, the numbers of HIinh ranged in an interval from 4.12E-03 (Ostrava-Radvanice, winter season) to 8.80E-05 (Čeladná, summer season). Approximately ten times lower values of HI_{inh} were detected for exposure model adult, tourist. In comparison with exposure scenarios for adults, slightly higher numbers of HI_{inh} were calculated for both exposure scenarios concerning children as a population category with higher sensitivity to external environmental noxes (Figures 2 and 3). It is also clear from both mentioned figures that the health risks tend to deteriorate during the winter period.

In comparison with the study realized in Changsha, China (Liu et al., 2015), the numbers of the hazard index for all sampling sites of the Moravian-Silesian Region are lower (exposure scenario adult, resident) or comparable (exposure scenario child, resident); higher HI value (2.01) was calculated only for exposure scenario child, resident in locality Ostrava-Radvanice in the winter season. On the contrary, ultrafine size fraction PM_1 collected in Kanpur, India (Singh and Gupta, 2016) were enriched in heavy metals. As it was to be expected, the identified HI numbers were higher than HI values found out for indoor dust obtained in Istanbul (Kurt-Karakus, 2012). More information is shown in Table 1.

With respect to the tested heavy metals, the highest risk (HQ_{inh} = 5.97E-03) was identified for lead (locality Ostrava-Radvanice, winter season, exposure scenario child, resident). On the contrary, the lowest health risk (HQ_{inh} = 1.26E-07) was found for vanadium in the summer season, at sampling site Ostrava-Poruba, for exposure scenario adult, tourist.

Concerning the health risks of evaluated carcinogenic heavy metals (As, Cd, Co, hexavalent Cr, Ni, Pb), some differences depending on time period and exposure scenarios were also determined. These variations of $ELCR_{tot,inh}$ values were very similar as in the case of HI. The highest rate of potential carcinogenic risk (ELCRtot, inh = 3.43E-05) was calculated for the industrial area Ostrava-Radvanice (winter period, exposure scenario adult, resident); the lowest danger of the potential carcinogenesis (ELCRtot, inh = 9.3E-08) was identified – a little unexpectedly – at the sampling site with high traffic load – i.e. in Ostrava-Poruba. From the point of view of monitoring heavy metals, the worst situation ($ELCR_{inh}$ = 1.66E-05) was identified for hexavalent chromium (site Ostrava-Radvanice, winter season, exposure scenario adult, resident); the lowest risk ($ELCR_{inh}$ = 4.8E-10) was determined for cobalt (sampling site Hradec nad Moravicí, summer time, exposure scenario child, tourist).

Tab. 1. Non-cancer risk characterization of various size fractions of PM

Tab. 1. Ryzyko nie rakotwórcze dla różnych frakcji PM (pyłu zawieszonego)

Locality	HI_{inh} (adult, resident)	HI_{inh} (child, resident)
Ostrava-Radvanice (PM_{10})	2.23E-03	1.09E-02
Frýdek-Místek (PM_{10})	7.60E-04	3.70E-03
Ostrava-Poruba (PM_{10})	5.76E-04	2.81E-03
Hradec nad Moravicí (PM_{10})	5.52E-04	2.69E-03
Čeladná (PM_{10})	8.80E-05	4.28E-04
Changsha, China (TSP)	6.95E-03	9.53E-03
Kanpur, India (PM_1)	2.13E-00	1.25E-00
Istanbul, Turkey (indoor dust)	3.58E-06	4.91E-06

Conclusions

The environmental contamination estimation and human health risk assessment were performed for heavy metals present in the structure of particulate matter PM_{10}. The samples of PM_{10} were collected by the High-Volume Cascade Sampler (DiggitalMD05) during two time periods at five sampling sites with various characteristics in the Moravian-Silesian Region. In all obtained samples, the heavy metal contents were determined by the method of ICP optical emission spectroscopy.

The environmental heavy metal contamination was assessed by the pollution load index (PLI); the human health risks connected with the inhalation exposure to heavy metals contained in PM_{10} were derived from calculating the hazard quotient (HQ_{inh}), the hazard index (HI_{inh}) and the excess lifetime cancer risk ($ELCR_{inh}$). In the comparison with background area, the level of environmental contamination was enormous especially during winter months. The potential non-cancer risks are not significant. The values of ELCR were considered to be tolerable or acceptable.

The limitations of this study lie in a small number of samples. Moreover, it is inevitable to get a more overall research with a view to the other PM_{10} noxious components because of their potential synergic effects onto human health (Reddy et al., 2012).

Acknowledgements

This paper was supported by the research projects of the Ministry of Education, Youth and Sport of the Czech Republic: The National Programme for Sustainability LO1404 – TUCENET, and SP2017/116 Research of various kinds of materials, their properties and application potential.

Literatura – References

1. BLAŽEK, Zdeněk; ČERNIKOVSKÝ, Libor; KREJČÍ Blanka; VOLNÁ, Vladimíra. The atmospheric particle contamination in region of Ostrava and Karviná. Prague: Czech Hydrometeorological Institute, 2008. 75pp.
2. BORM, Paul and DONALDSON, Ken.. Particle Toxicology. New York: CRC Press, 2007. 434 pp.
3. ČUPR, Pavel; FLEGROVÁ, Zuzana; FRANCŮ, Juraj; KLÁNOVÁ, Jana; LANDLOVÁ, Linda. Mineralogical, chemical and toxicological characterization of urban air particles. Environment International, 2013, vol. 54, p.26 - 34.
4. CAI, Li-Mei; XU, Zhen-Cheng; QI, Jian-Ying, FENG,Zhi-Zhou, XIANG,Ting-Sheng. Assessment of exposure to heavy metals and health risks among residents near Tonglushan mine in Hubei, China. Chemosphere, 2015, vol. 127, p. 127 – 135.

5. CHEN, Haiyang, TENG, Yanguo, LU, Sijin, WANG, Yeyao, WU,Jin, WANG, Jinsheng. Source apportionment and health risk assessment of trace metals in surface soils of Beijing metropolitan, China. Chemosphere, 2016, vol. 144, p. 1002 - 1011.
6. HU, Xin, ZHANG, Yun, DING, Zhuhong, WANG, Tijian, LIAN, Hongzhen Lian, SUN, Yuanyuan, WU, Jichun. Bioaccessibility and health risk of arsenic and heavy metals (Cd, Co, Cr, Cu, Ni, Pb, Zn and Mn) in TSP and PM2.5 in Nanjing. Atmospheric Environment, 2012, vol. 57, p. 146 – 152.
7. KURT-KARAKUS, Perihan Binnur. Determination of heavy metals in indoor dust from Istanbul, Turkey: Estimation of the health risk. Environment International, 2012, vol. 50, p. 47 - 55.
8. LIU, Xiaoting, ZHAI, Yunbo, ZHU, Yun, LIU, Yani, CHEN, Hongmei, LI, Ping Li, PENG, Chuan, XU, Bibo, LI, Caiting Li, ZENG, Guangming. Mass concentration and health risk assessment of heavy metals in size-segregated airborne particulate matter in Changsha. Science of the Total Environment, 2015, vol. 517, p. 215 – 221.
9. REDDY, R. Gottipolu; FLORA, S.J.S.; BASHA, Riaz. Environmental Pollution: Ecology and Human Health. Kolkota: Narosa, 2012. 371 pp.
10. SINGH, Dharmendra Kumar and GUPTA, Tarun. Source apportionment and risk assessment of PM1 bound trace metals collected during foggy and non-foggy episodes at a representative site in the Indo-Gangetic plain. Science of the Total Environment, 2016, vol. 550, p. 80 – 94.
11. TOMLINSON, D.L., WILSON, J.G., HARRIS, C.R., JEFFREY, D.W., 1980. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. Helgoländer Meeresun. 33, 566–575.
12. U. S. Environmental Protection Agency. (December 2000) Risk Characterization Handbook. Office of Science Policy, Office of Research and Development, Washington, DC. EPA 100-B-00-002.
13. U. S. Environmental Protection Agency. (May 1992) Guidelines for Exposure Assessment. Federal Register 57 (104): 22888-22938.
14. U. S. Environmental Protection Agency. (March 2007) Frame for Metals Risk Assessment. Office of the Science Advisor, Washington, DC. EPA 630/P-03/001F.
15. U. S. Environmental Protection Agency. (September 2006) A Framework for Assessing Health Risks of Environmental Exposures to Children. Office of Research and Development, Washington, DC. EPA/600/R-05/093F.
16. U. S. Environmental Protection Agency. (September 1986) Guidelines for Health Risk Assessment of Chemical Mixtures. Federal Register 51 (185): 34014-34025.
17. U. S. Environmental Protection Agency. (January 2014) Region 4 Human Health Risk Assessment. Supplement Guidance. Technical Services Section, Superfund Division. EPA Region 4.

Ocena ryzyka dla zdrowia ludzkiego w odniesieniu do metali ciężkich związanych z materią cząstek stałych

Próbki pyłu zawieszonego (PM10) zebrano w pięciu miejscach o różnym charakterze w regionie Mo-rawskośląskim (Republika Czeska). Stężenie metali ciężkich związanych z cząstkami stałymi oznaczano metodą ICP - optycznej spektroskopii emisyjnej. Zanieczyszczenie metalami ciężkimi zostało wyrażone jako wskaźnik obciążenia zanieczyszczeniem - PLI. Ocenę ryzyka dla zdrowia ludzkiego przeprowadzono metodą US EPA - z wykorzystaniem ilorazu zagrożenia - HQ, wskaźnika zagrożenia - HI i zwiększenia ryzyka wystąpienia raka w całym okresie życia - ELCR. Zanieczyszczenie i związane z tym ryzyko rako-twórczości zwiększa się w sezonie zimowym. Potencjalne zagrożenie nierakotwórcze nie jest znaczące.

Słowa kluczowe: zanieczyszczenie powietrza, pył zawieszony PM 10, metale ciężkie, wielkość zagrożenia, indeks zagrożeń