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MERCURY CONTENT AND DISTRIBUTION IN HOUSEHOLD DUST AND SOIL IN THE TOWN OF ŠID

ABSTRACT: Mercury (Hg) and potentially toxic elements (PTEs) are components of household dust and are a risk for human health. The aim of this study was to determine the concentrations of Hg and PTEs in household dust in individual housing facilities in the town of Šid, Serbia and their correlation to the content of the elements found in the surrounding garden soil. Total of 64 samples of household dust were collected from 16 locations of individual housing facilities on 4 occasions. Samples of surrounding yard and garden soil were simultaneously collected. None of the 64 analysed soil samples exceeded the threshold limit (TL) prescribed by law for non-agricultural soil, which is 0.3 mg kg^{-1} DM. Content of Hg in household dust was much higher than in the surrounding soil and ranged from 0.005 to 1.566 mg kg^{-1} DM. The resulting values of PTEs (As, B, Co, Cr, Cu, Ni, Pb, and Zn) in household dust had a significantly higher range than in the soil. Contents of Hg, B, Cu, and Zn were much higher in household dust than in the soil. According to the results of correlations of the analysed elements in household dust and surrounding soil, and according to the analysed locations, direct effect of Hg and PTEs contents on the content of the same elements in household dust was not confirmed.

KEYWORDS: mercury, potentially toxic elements PTEs, household dust, soil, Šid

INTRODUCTION

Mercury (Hg) is the only metal that easily changes state of aggregation, which allows it to migrate throughout the environment and enter organic and inorganic compounds with various levels of toxicity. There are studies on

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harmful materials in household dust which indicate health issues related to the dust (Casley et al., 2018), and studies on mercury risk assessment and bioavailability (Rasmussen et al., 2008; 2011; 2013; MacLean et al., 2011; Cukrowska, 2018). However, there are scarce studies on the origins of such contamination. It is assumed that Hg comes from different household appliances, from air (from coal burning products in rural areas), or indirectly from the atmospheric accumulation in the surrounding soil and consequent transfer into household dust. According to some studies, Hg in household dust may also originate from the construction materials of the facilities (Hagan et al., 2013).

The Minamata Convention on Mercury was approved and signed in Geneva in 2013 and entered into force in 2017. It is an international treaty designed to protect human health and the environment from the harmful effects of mercury. Control and monitoring of anthropogenically introduced Hg throughout its life cycle is the key factor in fulfilling the obligations set by the Convention, and the Republic of Serbia is a signatory country to the treaty. Sources of Hg are mostly historical pollution from chlorine production, Hg fluorescent lamps, gold and silver extraction and processing, application of dental Hg amalgam and instruments for measuring various physical quantities (thermometers, barometers). Other sources may include formerly used wood preservatives, seed protection chemicals, pesticide and paint industry, etc. (Adamov, 1984).

Potentially toxic elements (PTEs) are a group of elements in the environment which could be harmful to biota if found in higher concentrations. Some of these elements present in the soil (such as Co, Zn, Cu, B, etc.) simultaneously serve as nutrients to higher plants if present in optimum concentrations. PTEs in the soil can be of natural, geochemical origin, from parent material or from anthropogenous sources. PTEs reach the soil and household dust mostly by atmospheric deposition from various sources. In an indoor atmosphere these substances become deposited on different surfaces along with other components such as mold spores, mites and their excrement or discharge (Wisniewska et al., 2017). Multi-element analyses of household dust differ significantly from multi-element analyses of soil and street dust. The data show that higher concentrations are found in household dust samples from older homes (Rasmussen et al., 2001).

The aim of this study was to determine Hg and PTE concentrations in household dust in individual housing facilities in the town of Šid and their correlation to the content of the same elements found in the surrounding garden soil.

MATERIAL AND METHODS

Sample collection and processing

Samples of soil and household dust were collected simultaneously from 16 locations of individual housing facilities in the town of Šid, Serbia (45.1224° N, 19.2209° E) on four occasions: 21 September 2016, 3 December 2016, 5 March 2017 and 14 May 2017. Total of 64 samples of soil and household dust were col-

lected in this way. Soil samples were taken from yards and gardens from 0–20 cm depth, so that one composite sample is a collection of several individual samples. Household dust was collected indoors by vacuuming recent surface dust into clean sample bags. Larger solid particulates were removed from these dust samples (circa 100 g); the samples were then sieved and dried at room temperature.

Laboratory analyses

Laboratory analyses were performed at the Laboratory of the Faculty of Environment Protection, University Educons in Sremska Kamenica, Serbia and Laboratory for Soil and Agroecology of the Institute of Field and Vegetable Crops in Novi Sad, Serbia.

The soil samples were air-dried at the room temperature, milled and sieved to <2 mm particle size, in accordance with ISO 11464 (2006).

The samples were analysed for total Hg content using Direct Mercury Analyzer DMA 80 Milestone, which combines techniques of thermal decomposition, catalytic conversion, amalgamation, and atomic absorption spectrophotometry (λ=253.65 nm) in solid samples. Quality assurance and quality control (QA/QC) were conducted by certified reference material BCR 142R. The accuracy was within interval 92.84–109.70% and recovery was 101.11%.

The samples were analysed for pseudo-total contents of PETs (As, B, Co, Cr, Cu, Ni, Pb, Zn) after wet digesting the soil and household dust in concentrated HNO_3 and H_2O_2 ($5HNO_3$: $1H_2O_2$, and 1:12 solid-to-solution ratio). The concentration of elements was determined by ICP-OES (Vista Pro-Axial, Varian) in accordance with US EPA method 200.7:2001. Quality control was periodically carried out with reference materials ERM CC 141 (contains all examined elements, except B) and deviations were within ±10% of the certified values.

Statistical analysis

Data were statistically processed by analysis of the main descriptive parameters for each element. Statistical parameters were shown in tables and box-plots graphs. The significance of differences in measured parameters between the elements was determined using Fisher's LSD test ($p \leq 0.05$). All statistical analyses were performed using STATISTICA for Windows version 12 (Dell Inc. 2016).

RESULTS AND DISCUSSION

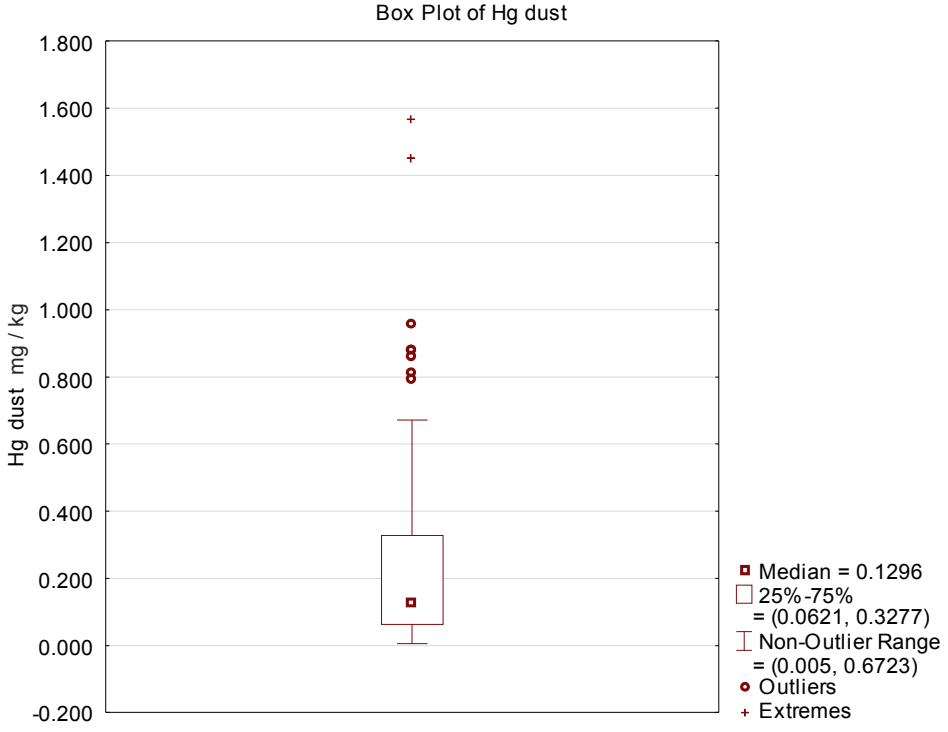
Concentration of Hg and PTEs in soil and dust

Significant correlation was not found between 4 occasions of sampling Hg and PTEs in household dust and surrounding soil on the analysed 16 locations. Regarding soil samples, this may be explained by too short a period between

sample collection occasions (12 days, 90 days and 70 days) to expect any increase in contents of the analysed elements in the soil.

Regarding Hg content in soil, none of the 64 analysed samples exceeded the threshold limit (TL) for soil, which is 0.3 mg kg^{-1} according to the Decree on Limit Values for Polluting, Harmful and Hazardous Substances in the Soil (*Official Gazette RS*, 30/2018) for non-agricultural soil. Hg content in household dust was much higher than in the surrounding soil and ranged from 0.005 to 1.566 with a mean value of 0.126 mg kg^{-1} (Table 1). The average Hg value of present research is lower compared to the obtained average value of the city of Novi Sad 0.32 (Kastori et al., 2009), and of the city of Budapest 1.03 mg kg^{-1} (Kastori et al., 2011) implying urban population size factors. Hg concentration in percentile interval 25–75% ranged from 0.061 to 0.328 mg kg^{-1} and was higher than the same interval for the soil (Figure 1). The concentration of toxic metals in household dust may be from 2 to 32 times higher than levels found in garden soil around the house (Rasmussen et al., 2001). It is hard to explain the origin of Hg in indoor dust samples. One of the possibilities of Hg source might be the damaged formerly used Hg thermometers (Kastori et al., 2009).

According to the defined ranges of concentrations for urban dust in Shanghai city (Wang et al., 2009), this study found 70% of household dust to be of clean level concentration (<0.259 mg kg^{-1}), 9% of the samples were slightly polluted (0.259–0.518 mg kg^{-1}), as much as 17% of samples were moderately polluted (0.518–1.295 mg kg^{-1}) and 3% of samples were heavily polluted (>1.295 mg kg^{-1}).



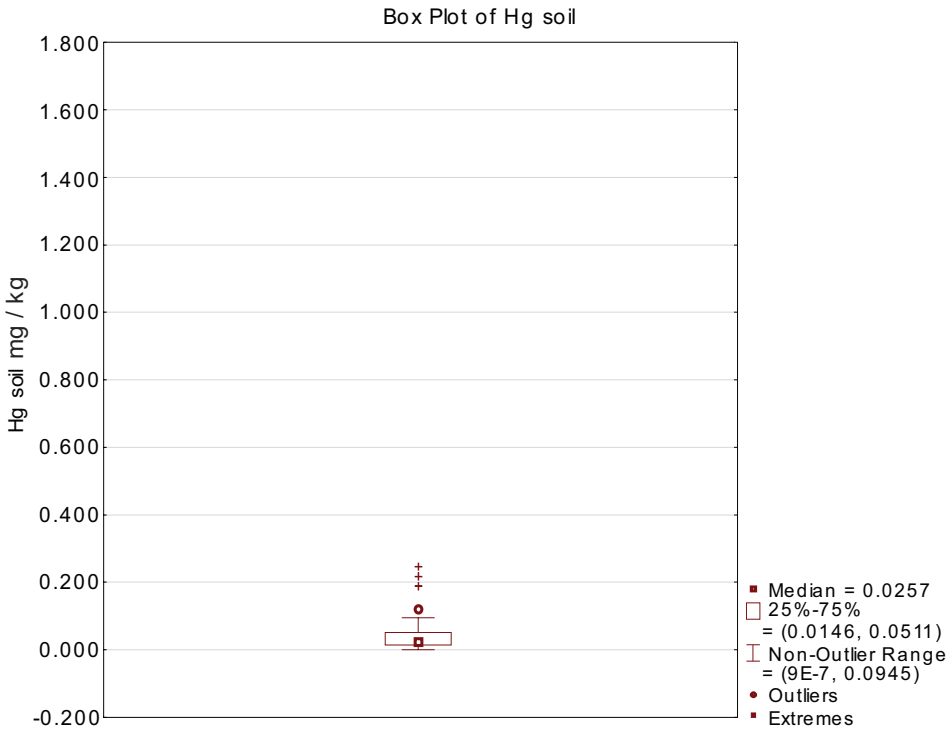


Figure 1. Box-plots graphic display of Hg in household dust and soil (mg kg^{-1} DM)

Some studies on the presence of mercury in household dust (Rasmussen et al. 2001) found total Hg concentration to range from 1.6 to 12.6 mg kg^{-1} in 40 dust samples. The highest Hg concentrations were reported in former Hg mining districts. In the Idrijan urban area (Slovenia), aqua regia soluble Hg (ARS Hg) was reported in topsoil in concentration of 8 to 1.210 mg kg^{-1} and in household dust, with 6 to 120 mg kg^{-1} (Bavec et al., 2018). Total Hg concentrations ranged from 3.06 to 926 mg kg^{-1} in dirt floors in Huancavelica, Peru (Hagan et al., 2013).

According to the box plots for other PTEs (figures not shown), as well as for Hg content (Figure 1), the obtained PTE values in household dust had statistically significant higher range with a larger number of extremes and outliers, which was expected when compared to the soil.

Contents of Hg, B, Cu, and Zn were much higher in household dust than in the surrounding soil. Pb content was higher in the dust, while Ni and Co contents were relatively higher in the soil. Contents of As and Cr were relatively even in the household dust and in the surrounding soil (Table 1). Relatively high concentrations of Cu and Zn found in this study are in accordance with other studies on office and household dust (Kefeni and Okonkwo, 2013) and street dust (Kastori et al., 2010; Žibret et al., 2013). Results of many researches suggest that there are a huge number of indoor pollutants sources such as: indoor activities, emission from building materials and furnishings, indoors tobacco smoking,

heating, pets, cooking activities etc. Accordingly, pollutants can originate from indoor environment itself as well as from outdoor source (Kastori et al., 2011).

Table 1. Minimum, maximum and mean value of PTEs in household dust and soil (mg kg^{-1} DM)

mg kg^{-1}	Home dust			Soil		
	min	max	mean	min	max	mean
Hg	0.005	1.566	0.126	0.00005	0.244	0.0257
As	2.4	12.9	5.0	3.3	10.5	6.8
B	6.9	221.1	28.3	2.3	10.4	5.5
Co	2.1	10.5	3.4	4.7	10.2	7.8
Cr	4.5	83.2	24.7	20.1	41.6	27.6
Cu	5.6	883.0	55.0	16.8	66.6	30.4
Ni	5.0	80.4	20.8	17.2	40.5	24.1
Pb	11.3	112.6	24.7	9.6	36.6	17.2
Zn	62.7	893.5	365.3	57.3	262.0	110.2

Correlations of Hg and PTEs between soil and dust

Hg in household dust was significantly positively correlated with B and Pb, and negatively correlated with Zn (Table 2). Soil Hg was not significantly correlated with either of the PTEs (Table 3). The obtained results show high Hg mobility throughout the environment, so this element does not correlate with other PTEs deposited into soil by atmospheric deposition.

There was a larger number of significant correlations between PTEs in the soil than in the household dust (Tables 2 and 3). In household dust, there were negative correlations between Cr and As, and between Zn and As (Table 2), while the same elements were positively correlated in the soil (Table 2). On the other hand, the following elements were positively correlated both in the dust and in the soil: Ni and Cr; Pb and Co; Zn and Cr, Cu and Ni. The results show heterogeneity of the potential PTE sources in household dust.

Table 2. Correlations between PTEs in household dust for all locations

	As	B	Co	Cr	Cu	Ni	Pb	Zn
Hg	0.1803	0.3351*	0.1771	-0.1343	-0.1782	-0.0973	0.2606*	-0.2526*
As		-0.1416	0.2255	-0.2723*	-0.1337	-0.2373	0.1048	-0.3580*
B			-0.0586	0.0785	0.1200	0.1914	0.2165	0.2000
Co				-0.2263	-0.0650	-0.2160	0.2705	-0.2354
Cr					0.2301	0.5746*	0.0496	0.3504*
Cu						0.1879	0.2113	0.6090*
Ni							0.0484	0.4218*
Pb								0.2021

* $p \leq 0.05$, significantly correlated

Table 3. Correlations between PTEs in soil for all locations

	As	B	Co	Cr	Cu	Ni	Pb	Zn
Hg	-0.0996	0.2078	-0.1526	-0.0697	0.1342	0.0081	-0.0041	0.1059
As		0.4310*	0.3672*	0.6464*	0.2787*	0.5795*	0.4464*	0.4974*
B			0.2388	0.4175*	0.4175*	0.2374	0.3046*	0.2976*
Co				0.6176*	0.3073*	0.3955*	0.5158*	0.2978*
Cr					0.2993*	0.8711*	0.6619*	0.5928*
Cu						0.2711*	0.5019*	0.4539*
Ni							0.6229*	0.6566*
Pb								0.7166*

* $p \leq 0.05$, significantly correlated

In order to determine the effects of PTE concentration in household dust, the correlations between Hg and PTE concentration were separately statistically processed for each of the analysed locations (data not shown). In 1,024 pairs (16 locations and 8 elements), significant correlations were found in 85 cases. Out of those, 12 cases had the same element positively correlated between dust and soil. According to the elements, Hg was positively correlated between dust and soil on two locations. Pb, Ni and Cu were positively correlated between dust and soil on two locations, and Zn and Cr on one location. The obtained results show that Hg and PTEs from the surrounding soil of yard and garden can affect their concentration in the indoor household dust; however, this has not been widely confirmed in this study. The results are in accordance with Rasmussen et al. (2001) who concluded that metal concentration in indoor dust cannot be predicted from outdoor soil levels.

CONCLUSION

Statistically significant correlation between the four sampling occasions on the analysed 16 locations was not found.

None of the 64 analysed samples exceeded the threshold limit (TL) for soil, which is 0.3 mg kg^{-1}. Content of Hg in household dust was much higher than in the surrounding soil and ranged from 0.005 to 1.566, with a mean value of 0.126 mg kg^{-1}.

Measured values of PTEs (As, B, Co, Cr, Cu, Ni, Pb, and Zn) in household dust had significant higher range than in the surrounding soil. Contents of Hg, B, Cu and Zn were much higher in household dust, content of Pb was higher in dust and contents of Ni and Co were relatively higher in soil. The contents of As and Cr were relatively even in dust and in soil. Relatively high concentrations of Cu and Zn found in this study are in accordance with other studies.

According to the results of correlations of the analysed elements in household dust and surrounding soil, and according to the analysed locations, direct effect of Hg and PTEs contents on the content of the same elements in household dust was not confirmed.

REFERENCES

Adamov Z (1984): *The effects of the long term mercury exposition of workers*. Thesis. Belgrade: School of Medicine.

Bavec Š, Biester H, Gosar M (2018): A risk assessment of human exposure to mercury-contaminated soil and household dust in the town of Idrija (Slovenia). *J. Geochem. Explor.* 187: 131–140.

Casley LS, Godec T, Logan JG, Pearce JC, Smith HMP, Stewart SA (2018): How clean is your house? A study of house dust mites, allergens and other contents of dust samples collected from households. *Int. J. Environ. Heal. R.* 28: 341–357.

Cukrowska E (2018): Risk assessment and bioavailability of mercury from dust in gold. *J. Environ. Anal. Toxicol.* 8.

Hagan N, Robins N, Hsu-Kim H, Halabi S, Espinoza Gonzales RD, Richter D, Vandenberg J (2013): Residential Mercury Contamination in Adobe Brick Homes in Huancavelica, Peru. *Plos One*. 16: e75179.

ISO 11464 (2006): Soil Quality – Pretreatment of Samples for Physico-chemical Analysis. *International Organization for Standardization*. Genève, Switzerland.

Kastori R, Maksimović I, Kadar I, Koncz J, Putnik-Delić M (2009): Elemental composition of indoor dust. *Proceedings of the 16th International Symposium on Analytical and Environmental Problems.* 28. 9. 2009, Szeged, Hungary, 198–202.

Kastori R, Maksimović I, Kadar I, Koncz J, Putnik-Delić M (2010): Monitoring of indoor pollution in Novi Sad and Budapest. *Proceedings of the 3rd International scientific conference "Remediation 2010*". 11–12. 5. 2010, Beograd, R. Serbia, 29–37.

Kastori R, Maksimović I, Kadar I, Koncz J, Putnik-Delić M (2011): Dust as environmental pollutant with special emphasis on indoor pollution. *Proceedings of the XII International Eco-conference, Environmental protection of urban and suburban settlements*. 21–24. 9. 2011. Novi Sad, R. Serbia, 37–49.

Kefeni K, Okonkwo J (2013): Trace metals, anions and polybromo diphenyl ethers in settled indoor dust and their association. *Environ. Sci. Pollut. R.* 20: 4895–4905.

MacLean LCW, Beauchemin S, Rasmussen PE (2011): Lead speciation in house dust from Canadian urban homes using EXAFS, micro–XRF and micro-XRD. *Environ. Sci.Technol.* 45: 5491–5497.

Rasmussen PE, Subramanian KS, Jessiman BJ (2001): A multi-element profile of housedust in relation to exterior dust and soils in the city of Ottawa, Canada. *Sci. Total. Environ.* 267: 125–140.

Rasmussen PE, Beauchemin S, Nugent M, Dugandzić R, Lanouette M, Chĕnier M (2008): Influence of matrix composition on bioaccessible copper, zinc and nickel in urban residential dust and soil. *Hum. Ecol. Risk. Assess.* 14: 351–371.

Rasmussen PE, Beauchemin S, Chĕnier M, Levesque C, MacLean LCW, Marro L, Jones-Otazo H, Petrovic S, McDonald LT, Gardner HD (2011): Canadian house dust study: Lead bioaccessibility and Speciation. *Environ. Sci. Technol.* 45: 4959–4965.

Rasmussen PE, Levesque C, Chĕnier M, Gardner HD, Jones-Otazo H, Petrovic S (2013): Canadian house dust study: Population–based concentrations, loads and loading rates of arsenic, cadmium, chromium, copper, nickel, lead, and zinc inside urban homes. *Sci. Total. Environ*. 443: 520–529.

Wang J, Chen Z, Sun X, Shi G, Xu S, Wang D, Wang L (2009): Quantitative spatial characteristics and environmental risk of toxic heavy metals in urban dusts of Shanghai, China. *Environ. Earth Sci.* 59: 645–654.

Wisniewska K, Lewandowska AU, Witkowska A (2017): Factors determining dry deposition of total mercury and organic carbon in house dust of residents of the Tri-city and the surrounding area (Baltic Sea coast). *Air Qual. Atmos. Health*. 10: 821–832.

Žibret G, Van Tonder D, Žibret L (2013): Metal content in street dust as a reflection of atmospheric dust emissions from coal power plants, metal smelters, and traffic. *Environ. Sci. Pollut. R.* 20: 4455–4468.

САДРЖАЈ И ДИСТРИБУЦИЈА ЖИВЕ (Hg) У КУЋНОЈ ПРАШИНИ И ЗЕМЉИШТУ ГРАДА ШИДА

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РЕЗИМЕ: Жива и потенцијално токсични елементи (ПТЕ) као саставни део кућне прашине представљају ризик за људско здравље. Циљ овог рада је утврђивање концентрација Hg и ПТЕ у кућној прашини, у објектима индивидуалног становања, града Шида и њихова веза са садржајем елемената у околном земљишту баштâ и окућница. Узето је 64 узорка кућне прашине на 16 локација индивидуалног становања у четири временска периода. Истовремено су узети узорци околног земљишта окућницâ и баштâ. Ниједан од 64 испитивана узорка земљишта не прелази законски прописану граничну вредност (ГВ) за непољопривредно земљиште од 0,3 mg kg^{-1} СМ. Садржај Hg у кућној прашини је много виши од околног земљишта и кретао се у интервалу од 0,005 до 1,566 mg kg^{-1} СМ. Добијене вредности ПТЕ (As, B, Co, Cr, Cu, Ni, Pb, Zn) у кућној прашини имају статистички значајан већи распон у односу на земљиште. Садржај Hg, B, Cu, Zn је много већи у кућној прашини у односу на околно земљиште. Према добијеним корелацијама посматраних елемената у прашини и земљишту, као и према посматраним локацијама, генерално није утврђен директни утицај садржаја Hg и ПТЕ у земљишту на садржај ових елемената у кућној прашини.

КЉУЧНЕ РЕЧИ: жива (Hg), потенцијално токсични елементи ПТЕ, кућна прашина, земљиште, град Шид