Global Sustainability Challenges
2023; Vol. 1(1): 33-42
https://gsc.unionnikolatesla.edu.rs/index.php/gsc/index
UDC: 504.5:669.018.674(497.11)
ISSN: 2956-1051 (Online)

Contamination and health risk assessment of heavy metals in PM10 in central Serbia

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To cite this article:
Mrazovac-Kurilić, S., Presburger-Ulniković, V. and Ćirišan, A. (2023). Contamination and health risk assessment of heavy metals in PM10 in central Serbia. *Global sustainability challenges, 1*(1), pp. 33-42

Received: Nov 7, 2022;: **Revised:** Nov 20, 2022; **Accepted:** Dec 16, 2022

Abstract: The objective of study was to investigate concentration and spatial distribution of heavy metals (As, Cd, Pb and Ni) in PM10 in central Serbia. Human health risks for heavy metals were assessed. Results showed that air in central Serbia does not contain significant heavy metal elements concentrations except in mining area (Bor). The contamination evaluation indicated that As, Cd, Ni and Pb in the air originated from anthropogenic sources. The non-cancer health risk assessment showed that ingestion was the primary exposure route for all metals in the air and that Pb, and As were the main contributors to non-cancer risks. HI values were calculated for children (HI=6.3E-07), indicating that children will likely experience higher health risks compared with adults (HI=7.1E-08). The non-cancer risks posed by all studied heavy metal elements and the cancer risks posed by As, Cd, and Ni to children and adults fell within the acceptable range.

Keywords: air; heavy metal elements; contamination assessment; health risk assessment; particulate matters PM10

1. Introduction

The particulate matters (PM) not only influence the natural conditions and make climate change (L. Wang et al., 2014). They have critical effects on human health under the high levels due to their deposition in the respiratory system (Makkonen et al., 2010; Moreno et al., 2011). The trace elements within particulates are important components in the atmosphere. They are present in our surrounding and have a significant contribution to potential health on the lung under long-term environmental exposure owing to their high bioreactivity and toxicity (Betha et al., 2014; Han et al., 2015; Moreno et al., 2011; Pan et al., 2015) According to the International Agency for Research on Cancer (IARC) of WHO (World Health Organization), the carcinogenic compounds are classified into five groups: Group 1 (carcinogenic to humans), Group 2A (probably carcinogenic to humans), Group 2B (possibly carcinogenic to humans), Group 3 (not classifiable as to its carcinogenicity to humans), and Group 4 (probably not carcinogenic to humans). In that study, As, Cd and Ni belong to Group 1, and Pb is included in Group 2A (Han et al., 2015; Tian et al., 2015; Wei et al., 2018).These toxic elements associated with particulate matter have potential threat to human health through air pollutants inhalation, dermal contact and ingestion in the human bodies (Zhang et al., 2015) Especially for children heavy metal adsorptions from the digestion system and hemoglobin sensitivity to these metals are both much higher than those for adults due to children's body at early ages and lower body weight Click or tap here to enter text. Therefore, trace elements in particulates are a serious concern for human health.

Over the past decades, there has been increasing concern about exposure of people, adults and children, to air contaminants in order to assess the health impact and to reduce human health risks. Therefore, air quality is an increasing public health concern due to presence of chemical and biological contaminants that might pose health effects. Heavy metals exist in the environment naturally as trace elements in rocks and soils, however they also are released to the environment as a result of human activities. Heavy metals may originate from various sources in urbanized areas, including emissions from vehicles, discharges from industrial facilities and other activities (Thornton, 1991).

As pollutant metals are generally non-biodegradable and no known homeostasis mechanisms of them, it is very likely that any high levels of heavy metals will pose serious effects on biological life. Accumulation in fatty tissues and circulatory system, negative effects on central nervous system and functioning of internal organs as well as acting as cofactors in other diseases and cancer can be listed as their negative effects on human (Dockery & Pope, 1996; Nriagu, 1988). Due to the maximal brain growth and differentiation of children at early ages, infants and toddlers are particularly vulnerable to heavy metal exposure and poisoning. Moreover, heavy metal adsorptions from the digestion system and hemoglobin sensitivity to these metals are much higher in children compared to adults (Bellinger, 1995).

Young children might be exposed to dust bounded contaminants, including heavy metals, at elevated levels due to their behaviors increasing indirect ingestion by way of hand-to-mouth activities, touching and mouthing of various dust-contaminated objects. Moreover, lower body-weight of children would result ingestion of greater amounts of dust compared to adults (Beamer et al., 2008). Health risk is especially high for children since their tolerance to toxins is lower (Acosta et al., 2009). A number of studies reported exposure of children to contaminated soils, dust and air particulates by ingestion a significant amount of toxic elements through the hand-mouth pathway as well as other routes of exposure (Chirenje et al., 2006; Inyang & Bae, 2006; Mielke et al., 1999; Raghunath et al., 1999).

Heavy metal elements in air (in PM10) are known to easily enter the human body through ingestion, inhalation, and dermal contact (Cook et al., 2005). The adverse effects on human health from exposure to heavy metal elements have been well-documented (Sun et al., 2010; Valko et al., 2006; Zheng et al., 2006).

Epidemiological studies have consistently shown an association between PM pollution and the number of deaths from cancer and cardiovascular and respiratory diseases (Pope et al., 2002). There is also evidence linking particulate air pollution and increases in hospital admissions for respiratory (Burnett et al., 1995; Pope, 1991; Roemer et al., 1993) and cardiovascular diseases (Burnett et al., 1995; Schwartz & Morris, 1995). In response to these adverse effects of air pollution, the EU Commission defined limit values for PM10 concentrations in ambient air (EU Directive 1999/30/EC).

Mining activities are notorious for their adverse impacts on the environment (Wang et al., 2008). Large quantities of dust laden with high levels of heavy metals can be released into the air as a result of mining operations including crushing, grinding, excavating, smelting, and refining (Csavina et al., 2012) Despite that, small regions affected by mining activities have received relatively limited attention. Thus, in comparison with mega-cities or capital cities, the environmental and human health risks in mining regions requires further investigation.

In this study, the attemption was made to evaluate the above mentioned issues by analyzing available PM10 data from 15 places in Serbia during 2017.

2. Materials and Methods

2.1. Site description

Central Serbia also referred to as Serbia proper, is the part of Serbia lying outside the provinces of Vojvodina to the north and the territory of Kosovo to the south. Central Serbia takes up, roughly, the territory of Serbia between the natural borders consisting of the Danube and Sava (in the north), the Drina (in the west), and the "unnatural" border to the southwest with Montenegro, south with Kosovo and FYR Macedonia, and to the east with Bulgaria, with a small strip of the Danube with Romania in the northeast. The Danube and Sava divides central Serbia from the Serbian province of Vojvodina, while the Drina divides Serbia from Bosnia and Herzegovina. The Great Morava, a major river, goes through central Serbia. Extensions of three major mountain chains are located within Serbia proper: Dinaric Alps in the west and south, and the Carpathians and Balkan Mountains in the east.

Table 1 lists the 15 towns (sites) in central Serbia where heavy metals in PM10 measurements have been performed. Figure 1 shows the geographical position of towns in central Serbia where the sites are placed.

Table 1. GPS coordinates of sites

Location	Sign	N (Latitude)	E (Longitude)
Belgrade 1	1	44.817237	20.477730
Belgrade 2	2	44.809956	20.380088
Belgrade 3	3	44.799531	20.475025
Bor 1	4	44.075725	22.108439
Bor 2	5	44.069892	22.098509
Valjevo	6	44.268273	19.890655
Veliko Gradište	7	44.749536	21.505809
Kragujevac	8	44.012793	20.911423
Kraljevo	9	43.723848	20.687254
Niš	10	43.320902	21.895759
Pančevo	11	44.874000	20.647567
Čačak	12	43.891414	20.350165
Ćuprija	13	43.925249	21.373496
Užice 1	14	43.855402	19.842094
Užice 2	15	43.679864	19.884480

The measurements of concentrations of pollutants were carried out by monitoring the air quality in the national and local networks.

2.2. Sample collection and processing

The urban stations monitor the influence of urban and suburban settlements over air quality. Radius of monitoring area varies between 1 and 5 km. Measurements of PM10 were performed by gravimetric analysis and those of heavy metals (Pb, Cd, Ni), in PM10, by atomic absorption spectrometry. Obtained data represent the average values of PM particles for a given region.

Sampling of suspended PM10 particles is carried out by an ambient air sampler Model LVS 3 Sven Leckel and Model MVS 6 Sven Leckel, Germany.

Test methods for suspended particles are:

-SRPS EN 12341:2008 Air quality - Determination of PM10 fraction of suspended particle-reference method and field test procedure to demonstrate the equivalency of measurement methods (gravimetric method);

-SRPS EN 12341:2015 Ambient air - Standard gravimetric measurement method for determining the mass concentration of PM10 suspended particles;

-SRPS EN 14902:2008 Ambient air quality - Standard method for the determination of Pb, Cd, As and Ni in the PM10 fraction of suspended particles (ICP-MS).

2.3. Health risk assessment model

2.3.1. Exposed doses

Figure 1. Map of the study area and sampling sites in Serbia

In this study, the risk assessment model developed by the Environmental Protection Agency of the United States (US EPA) was used to evaluate the health risks posed by heavy metals in road dust. Local residents were divided into adults and children and the following exposure categories were used: (1) adults and children through mouth and nose; 2) ingestion of dust particles through mouth; and 3) dermal contact with dust through exposed skin. According to the human health evaluation manual (Part A) and supplemental guidance for dermal risk assessment (Part E) (US EPA, 1989, 2004), the daily intake dose (D) of a pollutant through each pathway can be evaluated:

$$Ding = (C \times IngR \times EF \times ED \times CF) / (BW \times AT)$$

$$Dinh = (C \times InhR \times EF \times ED) / (BW \times AT \times PEF) \quad (1)$$

$$Ddermal = (C \times SA \times SL \times ABS \times EF \times ED \times CF) / (BW \times AT)$$

According to the classification list developed by the International Agency for Research on Cancer (IARC), three carcinogenic metals (As, Cd and Ni) were investigated for their carcinogenic risks (IARC, 2014). The life time average daily dose for these three metals was calculated by:

$$LADD = ((C \times EF) / (AT \times PEF)) \times ((InhRchild \times EDchild) / (BWchild) + (InhRadult \times EDadult) / (BWadult)) \quad (2)$$

where C is the upper limit of the 95% confidence interval for the mean (95% UCL), which is considered as a conservative estimate of the “reasonable maximum exposure” (US EPA, 1992). The 95% UCL in this study was calculated using previously described methods (Zheng et al., 2010a, 2010b) The other exposure factors for these models are shown in Table 2.

2.3.2 Risk characterization

For non-carcinogenic risks, Hazard quotient (HQ) was used to assess the non-carcinogenic risks posed by metals in road dust.

$$HQ = D / RfD \quad (3)$$

where RfD is the corresponding reference dose. An HQ<1 indicates no adverse health effects, while HQ>1 indicates that adverse health effects are likely to occur.

Table 2. Exposure factors

Factor	Definition	Adults value	Children value	Unit	Reference
BW	Average body weight	70	15	kg	EPA, 1989
IngR	Ingestion rate	100	200	mg/day	EPA, 1989
InhR	Inhalation rate	20	7.6	m^3/day	Zheng et al., 2010a
PEF	Particle emission factor	1.36×10^9	1.36×10^9	m^3/kg	EPA, 2001
SA	Sourface areas of the skin that contacts the airborne particulates	5700	2800	cm^{-2}	EPA, 2004
SL	Skin adherence factor	0.07	0.2	mg/m^3	EPA, 2004
EF	Exposure frequency	180	180	days/year	Zheng et al., 2010a
ED	Exposure duration	24	6	years	EPA, 2004
ET	Exposure time	24	24	hour/day	EPA, 2001
AT (non-carcinogenic risk)	Averaging time	ED×365	ED×365	days	EPA, 2001
AT (carcinogenic risk)	Averaging time	70×365	70×365	days	EPA, 2001
ABS	Dermal absorption factor	0.03(As); 0.001(others)	0.03(As); 0.001(others)	-	EPA, 2004
CF	Conversion factor	1×10^{-6}	1×10^{-6}	kg/mg	EPA, 2004

The hazard index (HI) is equal to the sum of HQs and is used to represent the total potential non-carcinogenic risks of different pollutants via three exposure routes described previously. An HI<1 indicates that there is no significant risk of non-carcinogenic effects. If HI>1, then a noncarcinogenic effect is likely to exist (US EPA, 1989).

In the case of carcinogenic risks, the life time cancer risk can be estimated by:

$$R = LADD / SF \quad (4)$$

where SF is the corresponding slope factor. Any cancer risk in the range of 10–6–10–4 is considered acceptable by the US EPA (US 1989). The RfD and SF values of all investigated metals (Ferreira-Baptista & de Miguel, 2005; Zheng et al., 2010b) are presented in Table 3.

2.4. Statistical analysis

CUCL (exposure-point upper confident limit content, mg kg−1) is considered to yield an estimate of the "reasonable maximum exposure" (Hu et al., 2011; US EPA, 1989; Zheng et al., 2010a, 2010b), which is the upper limit of the 95% confidence interval for the mean. The 95% upper confidence limit (UCL) was calculated by employing an approach called "adjusted Central Limit Theorem (CLT)" (Singh et al., 1997; US EPA, 2002). Although the approach was developed for normally distributed large data sets, the theorem does not say how many samples are sufficient for normality to hold. However, when sample size is moderate or small, the means will not generally be normally distributed yet the non-normality is intensified by the skewness of the distribution. Therefore, it is suggested that it can be employed for non-normal distributed moderate or small size data (US EPA, 2002). The 95% UCL concentration (CUCL) has been calculated as

Table 3. RfD and SF values

	Oral RfD	Inhal RfD	Dermal RfD	Inhal SF
Pb	3.50E-03	3.52E-03	5.25E-04	-
As	3.00E-04	3.01E-04	1.23E-04	1.51E+01
Cd	1.00E-03	1.00E-03	5.00E-05	6.30E+00
Ni	2.00E-02	2.06E-02	5.40E-03	8.40E-01

$$C_{UCL} = \bar{X} + \left[z_\alpha + \frac{\beta}{6\sqrt{n}}(1 + 2 \cdot z_\alpha^2)\right] \cdot \frac{S.D.}{\sqrt{n}} \quad (5)$$

X¯ = arithmetic mean; S.D.= standard deviation; β= skewness; α is the probability of making Type I error (false positive) and its value is 0.05; Zα= (1 −α)th quantile of the standard normal distribution. For the 95% confidence level, Zα= 1.645; n= number of samples and its value is 15 for all elements.

All statistical analysis were done in Statistica software package.

3. Results

3.1. Heavy metals contents in air

The content of heavy metals Pb, As, Cd and Ni in suspended PM10 particles was determined during 2017. Mean annual value measurement of heavy metals were used for analysis in 2017. The descriptive statistics related to heavy metal content in air in Serbia are listed in Table 4.

Table 4. Statistical parameters for 15 sites and 4 HM

Concentration	Max ($\times 10^{-6}$ mg/kg)	Min ($\times 10^{-6}$ mg/kg)	Mean ($\times 10^{-6}$ mg/kg)	Geom. Mean ($\times 10^{-6}$ mg/kg)	Median ($\times 10^{-6}$ mg/kg)	S.D.	Skewnes (β)	$C_{95\%\ UCL}$ ($\times 10^{-6}$ mg/kg)
Pb	122.58	1.61	22.93	9.94	6.86	35.51	2.28	43.78
As	58.06	0.24	8.84	2.10	1.61	19.05	2.40	20.19
Cd	3.55	0.08	0.98	0.46	0.32	1.10	1.13	1.54
Ni	9.68	1.61	4.73	3.97	4.03	2.79	0.64	6.04

The concentrations of 4 metals varied widely in this region and followed the order Pb>Ni>As>Cd.

In 2017, the mean annual value of arsenic in PM10 in Bor exceeded the limit values at stations Bor1 and Bor2. These are the only stations where the values for the arsenic exceeded limit values, while the average annual value of indicative measurements did not exceed the limit value at other stations. The value of cadmium is not exceeded limit value at any station. The highest mean annual concentrations of cadmium were measured in Bor: at the stations Bor1 and Bor2. The content of nickel in PM10 during 2017 was not exceeded limit values. The maximum daily value was in Bor on station Bor1.

In comparison with measurements in other countries, a significantly lower concentration of heavy metals in the air is observed in our observed territory (Botsou et al., 2020; Chen et al., 2022; Hou et al., 2019; Men et al., 2020; X. Wang et al., 2020; Yang et al., 2020).

Within the statistical analysis of data, the correlation between the elements was made, the analysis of the main components, considering the values of the correlation coefficients, and the cluster analysis.

The correlation coefficients with values that mostly exceed 0.5 show relatively high linear interdependence of elements. A high degree of correlation between the parameters indicates the common origin of the analyzed metals. The same was confirmed using factor analysis.

Table 5. Correlations

	Pb	As	Cd	Ni
Pb	1.00	0.93	0.82	0.65
As	0.93	1.00	0.75	0.48
Cd	0.82	0.75	1.00	0.68
Ni	0.65	0.48	0.68	1.00

Based on Figure 2, it can be concluded that Pb and Cd, as well as Ni and As make the primary clusters.

Table 6. PCA

	Factor1	Factor2	Factor3	Factor4
Pb	-0.963804	0.178534	0.121523	-0.156335
As	-0.898914	0.397479	0.133536	0.127010
Cd	-0.916590	-0.061108	-0.395017	0.009531
Ni	-0.773149	-0.612250	0.161556	0.035917

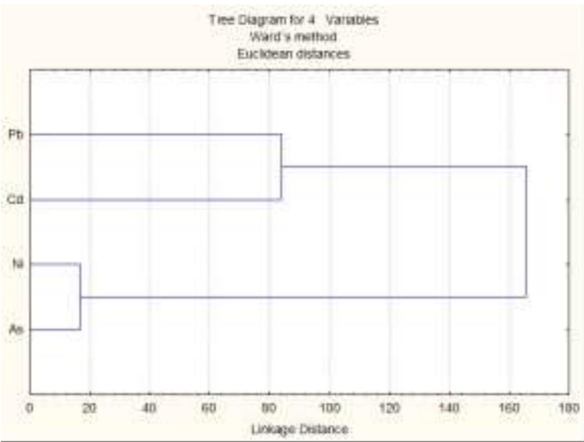


Figure 2. Clasters

Factor analysis showed the dominant influence of only one factor on the presence of analyzed 4 heavy metals in the air, and that is an anthropogenic factor-industry.

3.2. Spatial distributions of heavy metals in road dust

The spatial distribution pattern of 4 potentially toxic metals (As, Cd, Pb, and Ni) in PM10 in Serbia is presented in Figure 3. As, Cd, Pb, and Ni show spatial distribution patterns which coincide with the locations of industrial areas. The concentrations of those metals were higher near tailing pond and ore bodies (Bor). These results indicated that heavy metals likely originate from industrial sources in the study region.

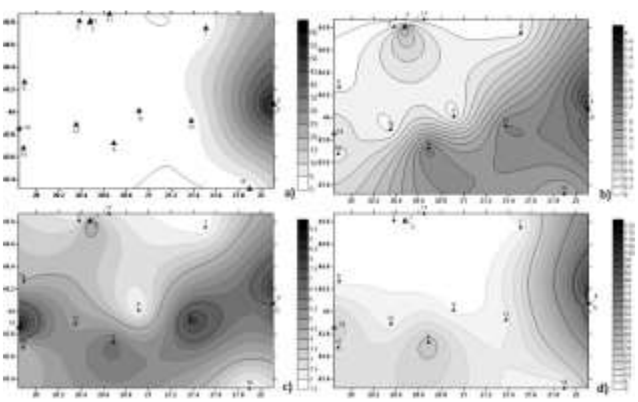


Figure 3. The spatial distribution of heavy metal elements (As, Cd, Ni and Pb) in air in Serbia (2017)

3.3. Health risk assessment results

3.3.1 Non-carcinogenic risk assessment

The HQ and HI for Ni, Pb, As and Cd in PM10 samples in Serbia were calculated (Table 7). The integrated HI values were 6.3E-07 for children and 7.1E-08 for adults living in Serbia, indicating children are likely to experience significantly higher non-cancer risks.

Among three different exposure pathways, the HQing values were the highest and contributed the most to HIs for both children and adults, indicating that ingestion of air appears to be the most threatening exposure way to human health in Serbia (Figure 4). The inhalation had the lowest contribution to health risks for children and the HQinh values were 3–4 orders of magnitude lower compared with the other two pathways for children, indicating that the non-cancer risks posed by the inhalation might be negligible compared with ingestion and dermal contact. Similar results were obtained by previous studies (Ferreira-Baptista & de Miguel, 2005; Zheng et al., 2010a).

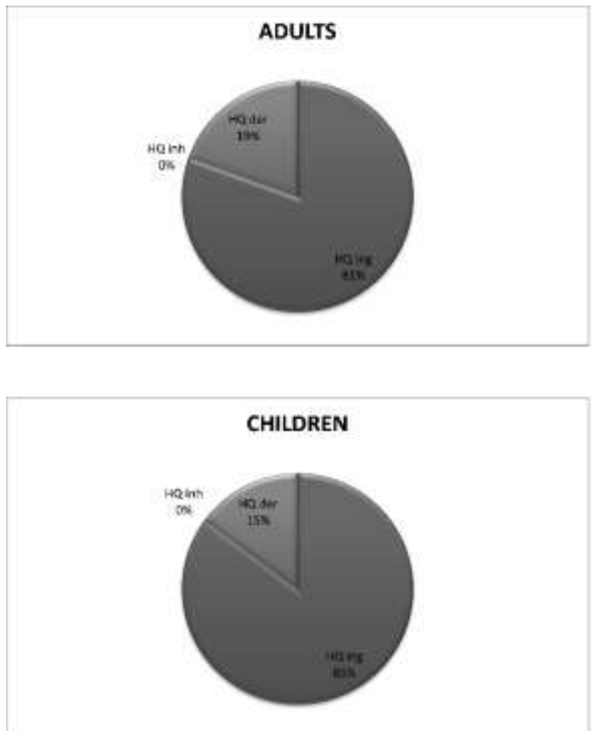


Figure 4. Non-carcinogenic risk distribution of different exposure ways for children and adults in Serbia (2017)

Additionally, children were found to experience higher health risks through ingestion compared with adults. The values of HQing for children were 9 times higher than those for adults and accounted for larger proportions (85% for children, 81% for adults) in integrated HI values. This result may be partially attributed to the special behavior patterns of children, particularly frequent hand-to-mouth contact (Figure 4).

The HIs for all studied metals were ranked in the order: As>Pb> Cd> Ni for adults, and for children (Table 7 and Figure 5). Pb and As were the main contributors to health risks posed by PM10 exposure for both children and adults, and Ni had the smaller contribution.

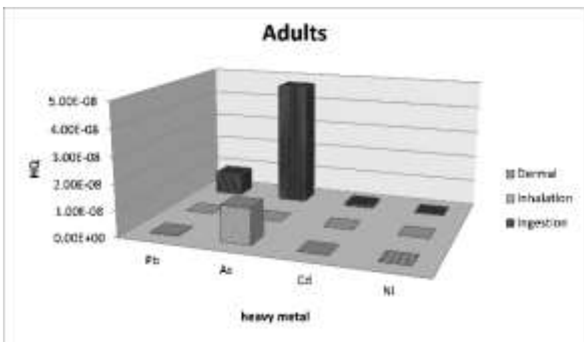
The HI values for all metals tested in this study and the integrated HI values for children and adults, were within the safe level (<1), suggesting minimal non-carcinogenic risk to children and adults from exposure to PM10 metals.

3.3.2 Carcinogenic risk assessment

The cancer risks according to inhalation exposure to Cd, Ni and As are presented in Table 7. Results showed that the overall risk of cancer decreased in the order Ni>As> Cd. The leading heavy metal was Ni for which cancer risks were 1 order of magnitude higher than those for other metals. Overall, cancer risk values for all heavy metals in this study were within the acceptable range, implying negligible carcinogenic risk.

Table 7. HQ, HI and R values for each non-carcinogenic and carcinogenic heavy metal measured in air in Serbia (2017)

HM	HQ ingestion		HQ inhalation		HQ dermal		HI		R
	A	Ch	A	Ch	A	Ch	A	Ch	
Pb	8.81E-09	82.25E-09	1.29E-12	2.28E-12	0.023E-08	0.15E-08	9E-09	8.4E-08	-
As	4.74E-08	44.25E-08	0.69E-11	1.23E-11	1.38E-08	9.07E-08	6.1E-08	5.3E-07	0.69E-16
Cd	1.08E-09	10.13E-09	0.16E-12	0.28E-12	0.0008E-07	0.0056E-07	1.1E-09	1.1E-08	0.01E-15
Ni	2.13E-10	19.86E-10	0.31E-13	0.54E-13	0.003E-09	0.02E-09	2.2E-10	2E-09	0.04E-14
SUM	5.8E-08	5.4E-07	8.4E-12	1.5E-11	1.4E-08	9.3E-08	**7.1E-08**	**6.3E-07**	**4E-16**



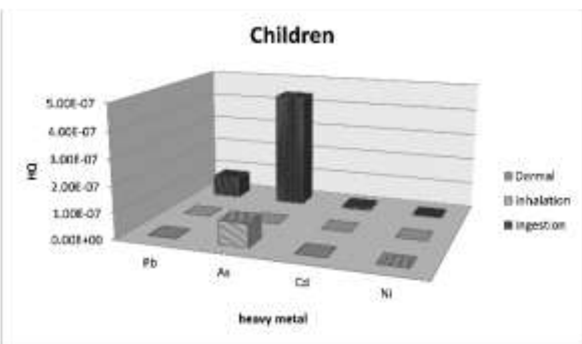


Figure 5 The HQs of each heavy metal in air in Serbia for adults (a) and children (b) (2017)

4. Conclusion

A total of 15 PM10 samples were collected from Serbia in 2017. The concentration and spatial distribution patterns of 4 potentially toxic heavy metal elements (As, Cd, Pb and Ni) in PM10 were analyzed. Human health risks for each heavy metal element were assessed using a human exposure model.

Results showed that concentrations of Cd, Pb, Ni and As, were not higher compared with limit values. The spatial distribution of Cd, Pb, Ni and As were all in accordance with the locations of industrial areas, indicating that these four heavy metals likely originated from industrial sources.

The health risks analysis showed that ingestion was the dominant exposure pathway for both children and adults. The HI value for As accounted for nearly 86% of the integrated HI value for adults and 84% for children, indicating that this heavy metal was the greatest contributors to non-cancer risks. Among the 3 carcinogenic metals, Ni was the leading contributor to cancer risks, followed by As and Cd. Although both non-carcinogenic and carcinogenic risk for each metal fell within acceptable values, children were more susceptible than adults and experienced higher non-carcinogenic risk from exposure to metals in air. The risks to children living in the central Serbia from exposure to heavy metal in air should receive greater attention.

Acknowledgments

Authors wish to thank University Union-Nikola Tesla, Belgrade, Serbia, for the support.

Conflicts of Interest

Authors declare that there is no conflict of interest.

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Procena zagađenja i zdravstvenog rizika od teških metala u PM10 u centralnoj Srbiji

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Abstrakt: Cilj rada bio je da se ispita koncentracija i prostorna distribucija teških metala (As, Cd, Pb i Ni) u PM10 u centralnoj Srbiji. Procenjeni su rizici po ljudsko zdravlje od teških metala. Rezultati su pokazali da vazduh u centralnoj Srbiji ne sadrži značajne koncentracije teških metala osim u rudarskom području (Bor). Procena kontaminacije je pokazala da As, Cd, Pb i Ni u vazduhu potiču iz antropogenih izvora. Procena nekancerogenog zdravstvenog rizika je pokazala da je gutanje bio primarni put izlaganja svim metalima u vazduhu i da su Pb i As dali glavni doprinos nekarcinogenom riziku. HI vrednosti su izračunate za decu (HI=6,3E-07), što ukazuje da će deca verovatno imati veće zdravstvene rizike u poređenju sa odraslima (HI =7,1E-08). Nekancerogeni rizik koji predstavlja uticaj sva četiri proučavana teška metala i kancerogeni rizik koje predstavljaju As, Cd i Ni, u slučajevima za decu i odrasle bili su u prihvatljivom opsegu.

Ključne reči: vazduh; teški metalni elementi; procena kontaminacije; procena rizika po zdravlje; čestice PM10
