



ORIGINAL ARTICLE

Lead Contamination in Playgrounds in Tuzla (Bosnia and Herzegovina)-environmental and Children Health Risk Assessment

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KEYWORDS

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ABSTRACT: Soil contamination with heavy metals has pervaded many parts of the world which can influence on human health. Environmental and health risk assessment processes are key steps in obtaining information on potential adverse effects on human health which may arise as a result of exposure to heavy metals from the environment. Thirty samples of soil were collected in summer 2018, from 10 municipal playgrounds in Tuzla (Bosnia and Herzegovina) to perform health risk assessment for children exposed to lead (Pb) from soil during their outdoor play activities. The Pb concentrations were determined by ICP-AES. Soil contamination was assessed using contamination factor and geoaccumulation index. Non-cancer risk due to exposure to Pb from soil thorough ingestion, dermal contact and inhalation pathway was estimated using a deterministic methodology. The results of the risk assessment indicated that the highest risk is associated with the ingestion of soil particles compared to two other exposure pathways. The mean total Hazard Index value calculated was 5.34E-02, below the threshold value of 1, indicating there was no increased health risk due to children's exposure to Pb. Although, the study showed there was no increased risk for children's health due to exposure to Pb from soil, there is still a rising concern regarding possible adverse effects of Pb on children's health. It is of great importance to monitor environmental pollution and continuously assess the risk to human health, not only thorough soil, but also through other important pathways of exposure such as food, water, air, toys, etc.

INTRODUCTION

Soil is one of the most important components of the living environment. Attention to the contamination of soils with heavy metals all over the world is increasing because environmental safety and human health have become important issues. Indeed, the toxic effects of heavy metals present in soil can be serious and even reach disastrous level [1]. Heavy metals have great potential to impact on the ecosystem, primarily destroying it [2]. Soil pollution by heavy metals can be a trigger a large number of adverse effects, both on the

human health and all spheres of environment. This leads to the disturbance of food chains, health related problems, as well as increased mortality rates [3, 4].

Soil in urbanized areas is particularly affected due to the constant pollution from traffic emissions, industrial wastes, thermal power plants and fuel combustion, household waste, increased urbanization and weathered particles of sidewalk and precipitation in the atmosphere, etc. Eventually, urban soil contaminated with heavy

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metals can become a serious threat to human health [5, 6].

Although, all heavy metals are characterized with their long persistence, non-biodegradable nature, and toxic properties, some of them dominate in the terms of high toxicity even in minor quantity. Among all heavy metals, lead (Pb) has received a paramount attention in recent studies [7-10]. Due to its ubiquity, lead is considered to be one of the most important heavy metals in terms of exposure of the general human population. Despite increasing knowledge about lead toxicity, its non-biodegradable nature, and bioaccumulative potential, its use has not decreased significantly over the years. As a result, there is an accumulation of lead originating from anthropogenic sources in the environment and an increase in hazard for environment and human health. Human exposure to Pb from soil can occur via three main routes: gastrointestinal ingestion of soil, inhalation of soil particles through respiratory system, and absorption of Pb from soil through the skin [11]. The high toxicity of lead is partly related to its ability to produce toxic effects on almost every organ and organ system. Of the all organic systems, the nervous system seems to be the target in Pb toxicity in people of all ages [12]. It is particularly characterized with great potency to cause toxic effects during the period of growth and development. Due to children's age is characterized with accelerated growth and development of their bodies, children have been repeatedly reported in several studies to be at higher risk for Pb poisoning than adults [13, 14]. The younger the children are, the higher the risk of the adverse effects of Pb exposure can be found. Signs of Pb intoxication in children and sever damage of nervous system can be mainly reflected in behavioral problems, decreased metal capacity, learning difficulties, a significant lower intelligence quotient [15]. Exposure to Pb can also be linked with severe damage of children's kidneys [13]. Thus, the effects of Pb intoxication in children are significantly more serious than in adults. Moreover, absorption of Pb through softer internal and external children's tissues occurs much faster than absorption through thicker and firmer adult tissues, because of, physiologically speaking, there is an evident difference in the absorption capacity of children and adult tissues [13].

Health risk assessment is an effective tool for human health risk quantification posed by heavy metals through various exposure pathways. Exposure assessment for children under the age of six requires consideration of child-specific behaviors and activities when defining exposure scenarios. The child specific behavior like hand-to-mouth activity can lead to a relevant exposure for children. The weight of evidence in the scientific literature generally suggests that the primary route of Pb exposure in child population is via ingestion, rather than the inhalation or dermal routes [16]. Young children, due to their childish carelessness and playfulness, have a tendency to ingest contaminated soil during their outdoor and play activities in the municipal playgrounds [17-19]. The aim of this study was to estimate the level of Pb contamination of municipal playground soils in Tuzla, Bosnia and Herzegovina, and to assess the potential non-carcinogenic health risk for children through multi-pathway exposures to the urban soil during outdoor activities at municipal playgrounds.

MATERIALS AND METHODS

Chemicals, Reagents and Glassware

Nitric acid and hydrochloric acid used for the digestion of soil samples were ultrapure grade from Lach-ner (Neratovice Praha, Czech). Double-distilled deionized water was used for all dilutions. Lead pure standard from Perkin Elmer (Walham, MA, USA) was used for the analysis. All glassware and plastics were cleaned by soaking in diluted HNO₃ (10% v/v) for 24 hours and rinsed with distilled water three times prior to use.

Study area

The Tuzla city is one of the largest cities in Bosnia and Herzegovina. It belongs to the Tuzla Canton, which is recognized as the most populous Canton and industrial center of Bosnia and Herzegovina. There are numerous children's playgrounds in the area of Tuzla which are parts of recreational parks constructed mainly in residential areas. Traditional social habits support and suggest everyday outdoor activities for children, especially younger children up to six years old.

The soil samples for this study were collected from the small municipal playgrounds at ten locations from the different parts of the city. From each location three

samples were taken. The locations of sampling sites are indicated in Figure 1. All samples were taken in summer 2018.



Figure 1. Locations of sampling sites in Tuzla city.

Sample collection and chemical analysis

Soil sampling and its preparation for the analysis was carried out according to the methodology described by standard ISO procedures [20-25]. The samples of approximately 1 kg of top-soils (0-15 cm) were sampled using non-metal (plastic) steel into the plastic bags and transferred to the laboratory. Samples were air-dried at the room temperature for seven days. A wooden rolling pin was used to mix and roll the soil samples. After obtaining soil samples grounded into fine powder, they were sieved and samples below 150 μm were repacked in labeled plastic pouches until. The fraction of each sample was reduced by repeated quartering until a 3.0 g of sample required for the analysis was obtained. Digestion procedure was performed according to the ISO 11466 standard procedure for extraction of trace elements from soil soluble in aqua regia. Mix of concentrated acids HCl and HNO₃ (3:1) was used for extraction of Pb from each sample. Reaction vessel, reflux condenser and absorption vessel were used for the preparation of soil samples. After cooling, sample solutions were filtered through filter paper and deionized water was added to the 250 mL marks on the volumetric flasks. A reagent blank was also prepared and passed through the same process. The total Pb content was determined by Inductive Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES Optima 2100 DV, Perkin Elmer, USA).

Calibration curves were prepared using analytical grade metal. A reagent blank was used to zero the instrument. The instrument was calibrated using calibration blank and five series of working standard solutions. Calibration range was from 30 $\mu\text{g}\cdot\text{L}^{-1}$ to 60 $\text{mg}\cdot\text{L}^{-1}$. Calibration curve showed good linearity ($r^2=0.999$). The instrument detection limit was 4 $\mu\text{g}\cdot\text{L}^{-1}$. The precision of method was 3.1 %. Each sample was analyzed in triplicates. The results were expressed in $\text{mg}\cdot\text{L}^{-1}$ of filtrate, and converted to $\text{mg}\cdot\text{kg}^{-1}$ of soil.

Assessment of heavy metal pollution

Soil pollution was assessed using equations for calculations of contamination factor (CF) and geoaccumulation index (I_{geo}).

Contamination factor is calculated as shown:

$$CF = \frac{C_n}{B_n} \quad (1)$$

where C_n is the concentration of Pb in the soil sample and B_n is a median concentration of Pb in the background soil sample previously established by Šljivić Husejnović et al. [26] for Tuzla Canton area.

Geoaccumulation index is calculated as shown:

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5 B_n} \right) \quad (2)$$

where, C_n and B_n are same values used for the calculation of CF, and 1.5 is the background matrix correction factor due to lithogenic effects.

Exposure and health risk assessment

Children playing in municipal playgrounds can be exposed to Pb via three main pathways. It involves direct oral ingestion of soil particles and absorption from gastrointestinal tract, inhalation of resuspended soil particles through the mouth and nose and absorption through respiratory system, and dermal exposure from particles adhered to exposed skin and absorption through skin [27-29]. To quantify children exposure to Pb the average daily dose (ADD) was calculated as follows:

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times CF \quad (3)$$

where, ADD_{ing} is average daily dose of Pb ingestion ($mg \cdot kg^{-1} \cdot day^{-1}$), C is concentration of Pb in soil ($mg \cdot kg^{-1}$), $IngR$ is ingestion rate of soil ($mg \cdot day^{-1}$), EF is exposure frequency ($days \cdot year^{-1}$), ED is exposure duration (years), BW is average body weight (kg), AT is average time (days) and CF is conversion factor ($1 \times 10^{-6} mg \cdot kg^{-1}$).

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad (4)$$

where, ADD_{inh} is average daily dose of Pb inhalation ($mg \cdot kg^{-1} \cdot day^{-1}$), $InhR$ is inhalation rate ($m^3 \cdot day^{-1}$) and PEF is particle emission factor ($1.36 \times 10^9 m^3 \cdot kg^{-1}$).

$$ADD_{derm} = \frac{C \times SA \times SAF \times ABS_{derm} \times EF \times ED}{BW \times AT} \times CF \quad (5)$$

where, ADD_{derm} is average daily dose of Pb intake through dermal contact ($mg \cdot kg^{-1} \cdot day^{-1}$), SA is surface area of the skin that contacts the soil (cm^2), SAF is skin adherence factor for soil ($mg \cdot cm^{-2}$) and ABS is dermal absorption factor for Pb.

The average daily doses calculated with previously described Eqs. (3)–(5) for each exposure pathway were used for calculating the Hazard Quotients (HQ) (6):

$$HQ = \frac{ADD}{RfD} \quad (6)$$

where, RfD is the chronic reference dose for Pb ($mg \cdot kg^{-1} \cdot day^{-1}$).

Considering that RfD_{ABS} for Pb exposure is not developed, the USEPA has proposed a method for extrapolation RfD_o values for the use in dermal risk assessment Eq. (7) [29].

$$RfD_{ABS} = RfD_o \times ABS_{GI} \quad (7)$$

where ABS_{GI} is the gastrointestinal absorption factor (unitless).

Although, there are three RfD s for three exposure pathways: RfD_o ($mg \cdot kg^{-1} \cdot day^{-1}$) for ingestion, RfD_{ABS} ($mg \cdot kg^{-1} \cdot day^{-1}$) for dermal contact and RfD_i ($mg \cdot m^{-3}$) for inhalation [30, 31], RfD_i for Pb and its compounds has not been developed [32]. Children exposure to Pb thorough soil may be considered safe if $HQ \leq 1$, while the values of $HQ > 1$, suggest that it is likely there will manifest some of the adverse health effects [33]. In order to assess the total potential for the occurrence of non-carcinogenic toxic effects posed by Pb thorough multiple exposure pathways, a hazard index (HI) has been calculated.

$$HI = \sum HQ_i = \sum \frac{ADD_i}{RfD_i} \quad (8)$$

The same criteria for the risk characterization were applied to HI, as for HQ. When $HI \leq 1$, it means that it is very likely that adverse effects on children's health due to Pb exposure from the soil will not be evidenced. When the $HI > 1$, the adverse health effects of children exposure to Pb may occur [31].

Health risk assessments using precisely defined exposure scenarios are essential to reflect the current population exposure to the chemicals. In this study, the exposure scenario included 236 young child residents (1-6 years old) of both gender from Tuzla area who were born between 2011 and 2016. Deterministic approach using mean values of observed parameter according to US EPA recommendation was applied to calculate the average daily exposure to Pb for children. All input parameters are presented in Table 1.

Table 1 Input parameters for exposure assessment and risk characterization

Exposure parameters	Description	Values	References
BW	Body weight (kg)*	16.20	Unpublished data**
EF	Exposure frequency (day·year ⁻¹)	350	[29]
ED	Exposure duration (years)	6	[29]
SA	Skin surface area available for exposure (cm ²)	2800	[29]
AT	Average time	EDx365	[34]
SAF	Soil to skin adherence factor (mg·cm ⁻²)	0.2	[29]
IngR	Ingestion rate (mg·day ⁻¹)	200	[29]
InhR	Inhalation rate (m ³ ·day ⁻¹)	20	[29]
PEF	Soil to air particulate emission factor (m ³ ·kg ⁻¹)	1.36 x 10 ⁹	[29]
ABS_{derm}	Dermal absorption factor (unitless)	0.001	[34]
RfD_{ing}	Pb (mg·kg ⁻¹ ·day ⁻¹)	3.50E-03	[29]
RfD_{derm}	Pb (mg·kg ⁻¹ ·day ⁻¹)	5.25E-04	[29]

* The average body weight for child population group.

** Unpublished data for children obtained from Public Health Institutions – Pediatrics departments from Tuzla Canton area. Data are presented as mean value of 236 body weights of randomly chosen children from 1-6 years old.

Statistical analysis

All experiments were set up in triplicates and the results were expressed as mean values. The data were analyzed using a statistical package IBM SPSS Statistics (Version 21).

RESULTS AND DISCUSSION

Contamination of soil with Pb

The occurrence of Pb in soil is not only dependent on the natural mineral soil composition, but also the anthropogenic activities being performed in the environment. It is well known that increased anthropogenic activities, industry activities and traffic lead to increasing in soil pollution with Pb. Previous study conducted in Tuzla Canton area [26] showed that 81.82% of analyzed soil samples were contaminated by Pb. It seems that Pb contamination of soil in this region very likely originates from anthropogenic sources. Therefore, this study is focused on the level of urban soil contamination by Pb primarily in areas where children often outdoor spend their time. The data of Pb concentrations in municipal playground soil samples are

presented in Figure 2. Concentration of Pb ranged from 8.02 to 26.01 mg·kg⁻¹. The decreasing order of arithmetic mean of the concentration of Pb at ten playground soil samples was 1 > 2 > 3 > 4 > 6 > 5 > 8 > 9 > 7 > 10. Higher Pb concentration was found in soil samples from the western part of the city which is located closer to the industrial areas of Tuzla Canton. Obtained results were compared to the maximum permitted limit value (PLV) of Pb in soil according to Federal Bosnian and Herzegovinian (F BiH) legislation and ordinance [35, 36] and to the background value of Pb obtained from the study previously conducted in this area [26]. Measured levels of Pb in soil samples revealed that the concentration of Pb was below the safe limits of F BiH legislation and ordinance. This means that, the PLV for Pb which is set to 50.00 mg·kg⁻¹ was not exceeded at none of ten sampling locations. Moreover, the results of ecotoxicological risk assessment of environmental pollution in municipal playgrounds in Tuzla city suggest that the majority of the sampling sites were not contaminated with Pb.

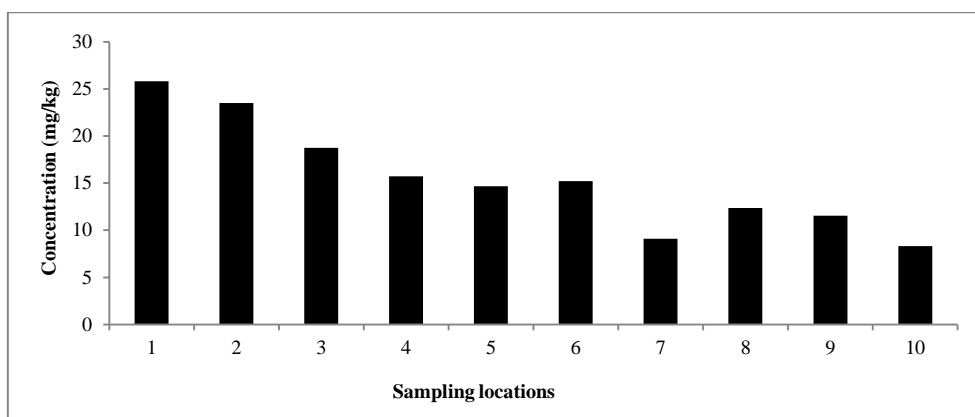


Figure 2. Clustered column of mean Pb concentrations in the playground soil samples

The results of CF and Igeo are shown in Figure 3. CF values indicated moderate soil contamination with Pb at two sampling locations (CF > 1), while at all other sampling locations CF value indicated low playground soil contamination with Pb (0.37 < CF < 0.84). The results

of Igeo values indicated that analyzed soil samples are practically uncontaminated with Pb with $I_{geo} < 0$ ($-2.01 < I_{geo} < -0.38$). From Figure 3 it could be inferred that the playgrounds in Tuzla city were not affected by anthropogenic soil pollution by Pb.

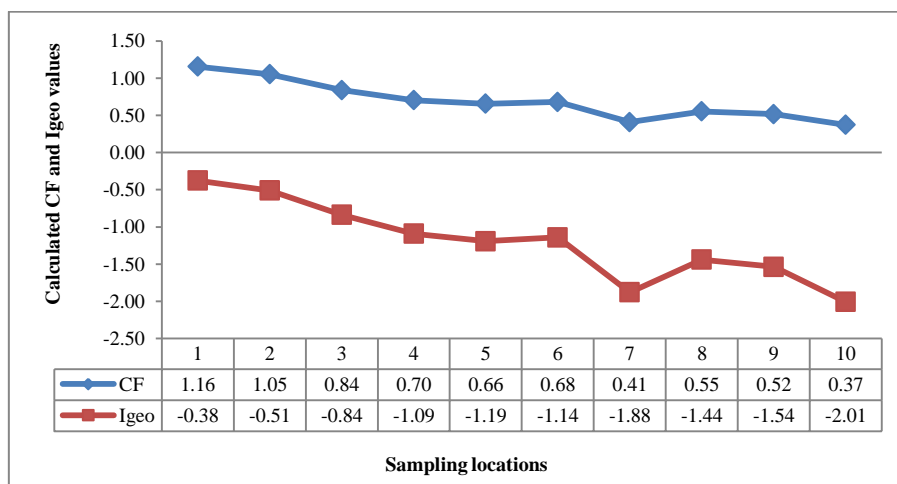


Figure 3. Contamination factor (CF) and geoaccumulation index (I_{geo}) in soil samples of municipal playgrounds in Tuzla

Health risk assessment

Table 2 summarizes the outcomes of the health risk assessment in context with non-carcinogenic health risk of Pb exposure. Estimation of average daily intake of Pb via three different exposure routes revealed that soil ingestion is the primary exposure route for children in case of exposure to Pb through soil. Moya and Phillips [37] reported that hand-to-mouth activities of young children, known as pizza behavior, is the main reason why children ingest more soil, compared to older children and adults. It is a worrying fact that, although it is estimated that 95% of children ingest an average of 200

mg soil per day, some children tend to ingest up to 300 times higher amounts of soil on a daily basis [38]. In light of the potential for children to ingest such large amounts of soil during a single day, an assessment was made of the possibility for soil pica episodes to result in acute intoxication from Pb concentrations the US EPA regards as acceptable values. Bearing in mind the realistic possibility that some children may ingest large amounts of soil in a single day, which could result in acute intoxication of Pb from the soil, even when Pb concentration in the soil are within the acceptable values,

it is necessary that the USEPA guidelines be applied with caution taking into account extreme cases of children exposure to heavy metals through soil.

In comparison to other similar studies conducted in urban regions, child residents from Tuzla, Bosnia and Herzegovina are at lower risk of adverse effects due to the exposure to Pb in soil than children living in Vilnius, Lithuania [19], Madrid, Spain [18], Port Pirie, Australia [17], Kabwe, Zambia [39], Hunan Province and Beijing, China [1, 40]. Moreover, the HI's for any applied concentration's distribution were lower than 1, therefore children's exposure to Pb in soil collected from municipal playgrounds has no potential to pose significant non-carcinogenic health impacts on 1-6 years old children residents of the Tuzla city.

Although, soil contaminated with Pb can endanger human health [5, 6], human exposure to Pb occurs mainly via ingestion of Pb-contaminated food, water, and

household paints, or via inhalation of Pb-contaminated dust particles or aerosols [41]. Further, there is strong scientific based evidence that Pb-based paint in children's toys poses an explicit health risk to vulnerable child population [42]. Since Pb exposure through soil is not the priority source of children exposure, and that this study did not evaluate exposure to this toxic metal via other media (food, air, water, toys, etc.), the estimated health risk is not definite indicator of overall children health risk due to Pb exposure in Tuzla city. Considering that Pb poisoning is one of the most common pediatric health problems in the United States and one of the major issue worldwide [41], it is necessary to continuously perform risk assessments due to children's exposure to Pb and Pb-compounds via different media and to apply aggregative and cumulative approach in extensive health risk assessments.

Table 2 Summary of average daily intakes and health risks of children exposed to Pb from municipal playgrounds in Tuzla city. HI<1 represent a very low probability of Pb toxic effect upon the specific exposure scenario.

Concentration's distribution	Exposure assessment				Health risk assessment			Total health risk
	ADD ing	ADD inh	ADD derm	ADD tot	HQ ing	HQ inh	HQ derm	HI
minimum	9.86E-05	7.25E-09	2.76E-07	9.89E-05	2.82E-02	2.07E-06	7.89E-05	2.83E-02
5 th percentile	1.03E-04	7.56E-09	2.88E-07	1.03E-04	2.94E-02	2.16E-06	8.22E-05	2.95E-02
25 th percentile	1.39E-04	1.02E-08	2.88E-07	1.39E-04	3.97E-02	2.92E-06	8.22E-05	3.98E-02
50 th percentile	1.77E-04	1.30E-08	4.95E-07	1.77E-04	5.05E-02	3.71E-06	9.43E-04	5.15E-02
75 th percentile	2.13E-04	1.57E-08	5.96E-07	2.14E-04	6.08E-02	4.47E-06	1.14E-03	6.20E-02
95 th percentile	2.93E-04	2.16E-08	8.21E-07	2.94E-04	8.38E-02	6.16E-06	1.56E-03	8.54E-02
maximum	3.06E-04	2.25E-08	8.56E-07	3.07E-04	8.73E-02	6.42E-06	1.63E-03	8.90E-02
median	1.77E-04	1.30E-08	4.95E-07	1.77E-04	5.05E-02	3.71E-06	1.41E-04	5.07E-02
mean	1.83E-04	1.35E-08	5.14E-07	1.84E-04	5.24E-02	3.86E-06	9.79E-04	5.34E-02

CONCLUSIONS

This study revealed that playground soil of Tuzla city contains Pb, one of the most important toxic heavy metals. However, its levels were within the permissible limits. The health risk assessment showed that the content of Pb in the soil from municipal playgrounds in Tuzla was not present in concentrations in which it could endanger the health of children who play there. However, the estimated health risk is not a definite indicator of overall children's health risk due to Pb exposure in Tuzla because it takes into account only the risk of Pb exposure during the time spent of the playground, and not other

more significant routes of exposure to this toxic metal. Although, exposure assessment has resulted in an acceptable level of children's health risk, in order to obtain more accurate overall health risk from Pb exposure in the child population, an aggregative and cumulative approach in risk assessment process should be applied.

CONFLICT OF INTERESTS

The authors declare that they have no known financial and non-financial competing interests or personal

relationships that could have appeared to influence the work reported in this manuscript.

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