

Human health risk assessment of lead, cadmium, and mercury co-exposure from agricultural soils in the Tuzla Canton (Bosnia and Herzegovina)

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The aim of this study was to assess the risk of human exposure to lead (Pb), cadmium (Cd), and mercury (Hg) through agricultural soil by considering both uncertainty and variability in key exposure parameters. For this reason we collected soil samples from 29 locations in the Tuzla Canton (Bosnia and Herzegovina) and measured their metal levels with inductively coupled plasma atomic emission or absorption spectrometry (ICP-AES and ICP-AAS, respectively). The levels of Pb ranged from 13.33 to 1692.33 mg/kg, of Cd from 0.05 to 3.67 mg/kg, and of Hg from 0.02 to 2.73 mg/kg. To estimate cancer and non-cancer risks we used deterministic and semi-probabilistic methods. Lead was found to involve higher health risk than the other two heavy metals. Its hazard index (HI) decreased between population groups (children>women>men) and exposure routes (ingestion>skin contact>inhalation). Our Monte Carlo simulations indicated that Pb HIs for both adult populations had a 0.6 % probability to exceed the threshold value of 1, while in children this probability was 14.2 %. Cd and Hg showed no probability to exceed the threshold in any scenario. Our simulation results raise concern about possible adverse health effects of heavy metals from soil, especially in children. It is very important to continue monitoring environmental pollution and assess human health risk, not only with respect to soil, but also with other important environmental compartments, such as air and water.

KEY WORDS: cancer risk, deterministic methodology, heavy metals, Monte Carlo simulations, non-cancer risk, probabilistic methodology

While ecological risk assessment can provide useful information about heavy metal pollution levels from environmental monitoring in specific sampling sites (1–3), it often ignores their behaviour in the soil-human system and impact on human health. These are addressed by health risk assessments of exposure from contaminated soils (4–10). Heavy metals from soil can enter the human body through ingestion, inhalation, or skin (11). Soil ingestion is of special concern in terms of acute exposure in children (9, 12–16). Heavy metals from soil can cause oxidative stress and DNA damage (17–23) leading to multiple organ damage, cancer, developmental problems, and even death (24–27).

Another issue related to soil pollution is combined exposure to several metals, each having more or less specific targets of toxicity and adverse outcomes. Such exposure can result in qualitatively and quantitatively different effects, often greater (additive or even synergistic) from those of a single substance (28) and is very difficult to predict.

Recently, agricultural and industrial soils in the Tuzla Canton of Bosnia and Herzegovina have drawn some attention, as a number of studies reported high heavy metal levels (9, 29–35), which pose a significant health risk due to several factors: 1) the Tuzla Canton is the most densely populated area of Bosnia and Herzegovina; 2) it has a number of polluting industries, including several large coal and salt mines; and 3) most agricultural soils are in their vicinity. However, health risk assessment (HRA) studies due to soil contamination with heavy metals in this area are rare, and the aim of this study was to address this issue, primarily by assessing the health risks of human exposure to a mixture of lead (Pb), cadmium (Cd), and mercury (Hg) using a deterministic and (semi)probabilistic approach. Considering both uncertainty and variability in key exposure parameters, our secondary aim was to gain a better understanding of each exposure route and of possible health risk differences between adults and children.

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MATERIALS AND METHODS

Study area

The Tuzla Canton is a resource-rich area located in the north-eastern part of Bosnia and Herzegovina with mining, coal, salt, chemical, and metal processing industry and a thermal power plant, all of which pollute its environment (Figure 1). Most of the industry is located near the rivers Jala and Spreca, which have been receiving large amounts of wastewater. In the rainy seasons, these rivers repeatedly flood adjacent agricultural land, which covers about 49 % of the Canton and often surrounds industrial zones. Our study area has a temperate continental climate with cold winters and hot summers and dominant south-westerly and north-easterly winds.

Sample collection and chemical analysis

Soil samples were collected in triplicates from 29 sites (Figure 1) four times a year in all four seasons from 2016 to 2017, totalling 348 samples. In reference to our previous study (17), we significantly increased the number of sampling sites and extended the sampling period to one year. The sites are well known for intensive agricultural production near industrial or mining activities and lie from 500 m to 50 km apart.

The samples were taken and prepared following standard ISO 11464, 10381-5, and 10381-6 procedures (36–38). Topsoils (0–15 cm) were sampled using a plastic tool. Each sample was obtained by collecting 8–10 subsamples using a combination of two sampling designs: simple random and judgmental sampling.

Samples were air-dried in the laboratory at room temperature for seven days, ground into fine powder, sieved through a 0.15 mm polyethylene sieve, and packed until analysis. According to the ISO 11466 (39) standard procedure for extraction of trace elements from soil, 3 g of each sample was digested with concentrated *aqua regia* (in a 3:1 HCl to HNO₃ ratio) at room temperature for 16 h and then boiled under reflux for 2 h. The extract was then filtered through Whatman filter paper and diluted with deionised water in a volumetric flask. We also prepared a reagent blank for each metal following the same steps. Calibration curves were prepared with analytical standards, and the blank was used to zero the instrument. Each sample was analysed in triplicate, and results expressed in mg/L of filtrate.

Total Pb content was determined with inductively coupled plasma atomic emission spectroscopy (ICP-AES, Optima 2100 DV, Perkin Elmer, Waltham, MA, USA), while total Cd and Hg were determined with atomic absorption spectroscopy (AAS) using a Varian SpectraAA 220 with either GTA-110 or VGA-77, respectively (Varian, Belrose, Australia).

Soil texture was evaluated with particle-size analysis (PSA) as described earlier (17). Air-dried soil samples were manually passed through nine sieves of different mesh sizes in order to calculate the percentage of individual soil types in soil samples.

Exposure assessment

To assess human exposure to heavy metals we calculated the average daily dose (ADD) for each route of exposure using the following equations:

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times CF \quad [1]$$

where ADD_{ing} is the average daily intake of metal through ingestion (mg/kg/day), C is heavy metal mass fraction in soil (mg/kg), $IngR$ is the soil ingestion rate (mg/day), EF exposure frequency (days/year), ED exposure duration (years), BW average body weight (kg), AT average time (days), and CF the conversion factor (10^{-6} kg/mg);

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad [2]$$

where ADD_{inh} is the average daily intake of metal through inhalation (mg/kg/day), $InhR$ is inhalation rate (m^3 /day), and PEF is the particle emission factor (1.36×10^{-9} m^3 /kg), and

$$ADD_{derm} = \frac{C \times SA \times SAF \times ABS_{derm} \times EF \times ED}{BW \times AT} \times CF \quad [3]$$

where ADD_{derm} is the average daily intake of metal through skin (mg/kg/day), SA is the skin surface area in contact with soil (cm^2), SAF is the skin adherence factor for soil (mg/ cm^2), and ABS is the dermal absorption factor for metals (unitless).

Human health risk assessment

Non-cancer risk was calculated for all three heavy metals, while cancer risk was calculated only for Cd, because the International Agency for Research on Cancer (IARC) has classified only Cd and Cd compounds as carcinogens to humans (Group 1) (40).

Human non-cancer risk was calculated using the following equation:

$$HQ = \frac{ADD}{RfD} \quad [4]$$

where HQ is the hazard quotient and RfD chronic reference dose for the analysed metal (mg/kg/day). There are three $RfDs$, one for each exposure route: RfD_o (mg/kg/day) for ingestion, RfD_{ABS} (mg/kg/day) for skin, and RfD_i (mg/ m^3) for inhalation (41). The United States Environmental Protection Agency (US EPA) has not yet developed an inhalation reference concentration for Pb and Pb compounds (42). If ADD is lower than the RfD , HQ is ≤ 1 , and the risk is considered acceptable. If ADD is higher than the RfD , HQ is > 1 , adverse health effects are likely (43, 44). To assess

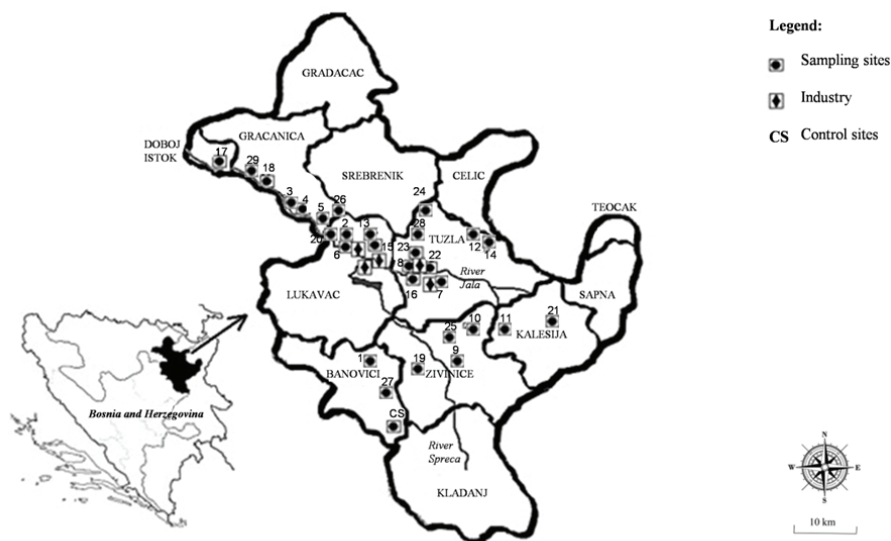


Figure 1 Locations of the sampling sites in the Tuzla Canton

the total non-cancer risk posed by more than one heavy metal, we calculated the hazard index (HI) as follows (45):

$$HI = \sum HQ_i = \sum \frac{ADD_i}{RfD_i} \quad [5]$$

If HI is ≤ 1 , the exposed population is unlikely to experience adverse health effects, and if it is >1 , then adverse health effects are likely (46).

The US EPA has developed a method to extrapolate oral toxicity values to assess dermal risk and calculate RfD_{ABS} (41) as follows:

$$RfD_{ABS} = RfD_o \times ABS_{GI} \quad [6]$$

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

Cancer risk for Cd was estimated as incremental probability for an individual to develop cancer over a lifetime as a result of exposure to a potential carcinogen. It was calculated as follows (43):

$$\text{Cancer Risk} = ADD \times CSF \quad [7]$$

where, ADD is the chronic daily intake of Cd averaged over 70 years (mg/kg/day) and CSF cancer slope factor. Risks in the range from 10^{-6} to 10^{-4} have typically been considered acceptable by the US EPA (47–49).

Semi-probabilistic exposure assessment

Non-cancer risk was computed using the Monte Carlo simulation, a statistical method which repeatedly calculates random “what-if” scenarios in a single operation to produce the full range of possible outcomes and their likelihoods (50). For uncertainty analysis of estimated risks, we used the @RISK 5.5 software (Palisade, Ithaca, NY, USA). @RISK works with data in Microsoft Office Excel spreadsheets and relies on Monte Carlo simulations to

estimate the probability of different outcomes that cannot easily be predicted with the deterministic approach.

Based on behavioural and physiological differences, the population in our study was divided into three groups: adult men (20–70 years old), adult women (20–70 years old), and children (1–6 years old). Health risks associated with exposure to heavy metals in soil for these population groups were calculated using the above equations [1–7]. The statistical distributions of parameters heavy metal concentrations and body weight were assumed to be lognormal. Instead of single-point values, we used different variable values for 348 concentrations of Pb, Cd, and Hg, as well as for body weights of 87 adult men (average age 37.17 ± 1.28 years), 80 adult women (average age 34.72 ± 1.05 years), and 236 children (average age 4.01 ± 0.87 years). Body weight data for the adults were obtained from an independent research project conducted by the students of the University of Tuzla, Faculty of Pharmacy. Body weight data for children were pooled from paediatrics departments of Public Health Institutes in the Tuzla Canton. Soil heavy metal levels and body weights of each population group were considered input parameters to evaluate the probability function for human exposure to heavy metals. The stability of the results was tested at 5,000, 10,000, 25,000, and 50,000 iterations by running randomly selected values of independent variables according to their distribution function. The stability test showed that 10,000 iterations sufficed for a reliable yield. The number of iterations for every equation was then set to 10,000 to derive the certainty level, mean, the 75th and 95th percentile, and maximum values of exposure.

Exposure scenarios

Exposure was evaluated through three scenarios with adult men, women, and children from the Tuzla Canton.

We assumed exposure to Pb, Cd, and Hg from soil through skin, ingestion, and inhalation. All input parameters are presented in Table 1.

Scenarios 1 and 2 involved adult men and women, respectively, and Scenario 3 children (1–6 years old) of both genders born between 2011 and 2016.

Statistical methods

All experiments were done in triplicate and the results expressed as means of 36 measurements. The data were analysed using the IBM SPSS Statistics (version 21) package (Armonk, NY, USA). The normality of variable distribution was tested with the Shapiro-Wilk test. Spearman's rho correlation analysis of heavy metal concentrations was used to identify the same source of soil contamination if any.

RESULTS AND DISCUSSION

Heavy metal soil concentrations

Particle-size analysis showed that all soil samples were sandy. Heavy metal levels were compared to respective national permitted limit values (PLVs) in sandy agricultural soils (51, 52).

Soil levels of Pb, Cd, and Hg are presented in Table 2. Lead levels ranged from 13.33 to 1692.33 mg/kg (median 92.83 mg/kg) and exceeded PLVs in 71.55 % of all soil samples. Cadmium ranged from 0.05 to 3.67 mg/kg (median 0.32 mg/kg) and exceeded PLVs in 35.61 % of all soil samples. Mercury ranged from 0.02 to 2.73 mg/kg (median 0.11 mg/kg) and exceeded PLVs in 8.62 % of samples.

The Shapiro-Wilk test showed that the levels of all three heavy metals were not distributed normally. Spearman's

Table 1 Input parameters for exposure assessment

Exposure parameters	Description	Values	References
BW	Average body weight* (kg)	82.78 for adult males	Unpublished data**
		62.66 for adult females	
		16.20 for children	
EF	Exposure frequency (day/year)	350	(41)
CF	Conversion factor (kg/mg)	10 ⁻⁶	(43)
ED	Exposure duration (years)	30 for adults	(41)
		6 for children	
SA	Skin surface area available for exposure (cm ²)	5700 for adults	(41)
		2800 for children	
AT	Average time	EDx365 for non-carcinogenic 25550 for carcinogenic risk	(43, 69, 70)
SAF	Soil to skin adherence factor (mg/cm ²)	0.07 for adults	(41)
		0.2 for children	
IngR	Ingestion rate (mg/day)	100 for adults	(41)
		200 for children	
InhR	Inhalation rate (m ³ /day)	20	(41)
PEF	Soil to air particulate emission factor (m ³ /kg)	1.36 x 10 ⁹	(41)
ABS _{derm}	Dermal absorption factor (unitless)	0.001 (for all metals)	(69, 70)
RfD _o	Pb (mg/kg/day)	3.50E-03	(41)
	Cd (mg/kg/day)	1.00E-03	
	Hg (mg/kg/day)	3.00E-04	
RfD _{ABS}	Pb (mg/kg/day)	5.25E-04	(41)
	Cd (mg/kg/day)	1.00E-05	
	Hg (mg/kg/day)	2.10E-05	
RfD _i	Hg (mg/kg/day)	8.57E-05	(49)
CSF	Cd (mg/kg/day)	6.30E-00	(71)

*The average body weight for each population group used for health risk assessment based on deterministic approach. **Unpublished data for adults and children presented as mean body weights of randomly chosen 87 male and 80 female adults and 236 1–6-year-old children from the Tuzla Canton

Table 2 Heavy metal levels (mg/kg) and corresponding permitted limit values (PLVs) for sandy agricultural soils

Sampling site	Mean ± SD (mg/kg)*		
	Pb	Cd	Hg
1	129.75±23.93	0.15±0.05	0.11±0.01
2	69.58±15.69	0.23±0.09	0.08±0.02
3	92.47±8.45	1.61±0.97	1.14±0.59
4	34.53±7.46	0.77±0.58	0.20±0.13
5	44.67±10.09	0.39±0.07	0.06±0.02
6	400.64±284.62	1.03±0.88	0.09±0.02
7	376.67±72.54	0.15±0.02	0.22±0.12
8	48.56±13.94	0.33±0.12	0.06±0.02
9	96.14±27.92	0.16±0.03	0.09±0.08
10	93.94±4.32	0.09±0.02	0.05±0.02
11	98.64±13.57	0.14±0.05	0.08±0.04
12	159.75±18.98	0.31±0.04	0.09±0.05
13	101.78±14.47	0.18±0.06	0.08±0.04
14	440.87±72.83	0.22±0.11	0.11±0.03
15	301.22±66.37	0.07±0.02	0.08±0.02
16	1724.45±854.87	0.29±0.05	0.29±0.11
17	61.89±15.09	0.53±0.03	0.25±0.03
18	51.14±4.19	1.67±0.11	0.57±0.02
19	154.00±7.99	0.21±0.04	0.05±0.01
20	45.69±9.05	0.18±0.01	0.10±0.03
21	118.86±3.36	0.38±0.02	0.08±0.04
22	44.69±7.41	0.09±0.01	0.05±0.02
23	28.81±3.28	1.12±0.10	0.14±0.01
24	16.86±3.63	2.45±0.18	0.18±0.05
25	143.55±6.90	0.68±0.05	0.08±0.01
26	75.39±7.12	2.99±0.31	1.55±1.12
27	143.92±28.74	0.51±0.07	0.13±0.01
28	33.5±4.41	1.36±0.15	0.16±0.01
29	35.81±5.45	1.35±0.08	0.20±0.01
Maximum permitted limit value (mg/kg)			
PLV	50.00	0.50	0.50

*Each result is presented as the average of 36 measurements. Three samples were taken from each site in each season of the year. Each of the 12 samples from the 29 sites was analysed in triplicate. Bolded figures indicate values greater than the respective permitted limit values (PLVs)

rho correlation analysis showed that Pb negatively correlated with Cd ($r=-0.38$; $p=0.00$), Cd positively correlated with Hg ($r=0.50$; $p=0.00$), and Pb negatively correlated with Hg levels ($r=-0.14$; $p=0.01$). Judging by these correlations, all three heavy metals could originate from the same source of pollution.

Table 3 shows that the lowest mean soil Pb level remained similar to those reported earlier for the same area, but the highest Pb level measured in agricultural soil near the chloralkali plant in Tuzla significantly exceeds the maxima reported earlier. Additional large sources of Pb soil contamination were located near the highway and the coal-

fired power plant, which is the largest power producer in Bosnia and Herzegovina. Its wastewaters contain Pb, Cd, Cr, and Ni in levels above the prescribed limits (29, 30).

The lowest mean Cd level is also within national limit, just like the level reported earlier for alkaline coal ash landfills in the County (29), whereas its levels in soil sampled near the salt mine Tetima and urban areas of Lukavac and Kalesija contained much higher Cd levels (31, 32). However, the highest mean Cd level in our study exceeds earlier maxima. Lukavac is an industrial zone with mining, coke, and cement industries, which release large quantities of waste materials and contaminate soil.

Table 3 Comparison of soil heavy metal levels in the Tuzla Canton across studies

Pb (mg/kg)		Cd (mg/kg)		Hg (mg/kg)		Year of soil sampling	References
Min	Max	Min	Max	Min	Max		
18.00	24.00	0.20	0.40	N/A	N/A	2005	29
N/A	N/A	N/A	N/A	0.23	65.5	2004	30
8.02	26.01	N/A	N/A	N/A	N/A	2018	9
22.60	41.04	1.29	2.43	N/A	N/A	2017	31
17.20	36.09	0.00	1.86	0.00	27.35	2013	32
N/A	N/A	N/A	N/A	0.00	3864.00	2013	33
14.14	190.82	N/A	N/A	N/A	N/A	N/A	34
18.00	92.00	N/A	N/A	N/A	N/A	N/A	35
14.33	2784.00	0.05	3.67	0.03	2.73	2016-2017	This study

N/A – data not available

Table 4 Deterministic non-cancer risk due to Pb, Cd, and Hg exposure through soil

Heavy metal	Oral intake (mg/kg/day)	Oral risk (unitless)	Dermal intake (mg/kg/day)	Dermal risk (unitless)	Inhalation intake (mg/kg/day)	Inhalation risk (unitless)
Pb	Adult men (Scenario 1)					
	2.06E-04	5.90E-02	8.24E-07	1.57E-03	3.04E-08	8.67E-06
	Adult women (Scenario 2)					
	2.73E-04	7.79E-02	1.09E-06	2.07E-03	4.01E-08	1.15E-02
	Children (Scenario 3)					
	6.03E-01	4.29E-02	5.91E-06	1.13E-02	1.55E-07	4.43E-05
Cd	Adult men (Scenario 1)					
	7.84E-07	7.84E-04	3.13E-09	3.13E-04	1.15E-10	1.15E-07
	Adult women (Scenario 2)					
	1.04E-06	1.04E-03	4.13E-09	4.13E-04	1.52E-10	1.52E-07
	Children (Scenario 3)					
	8.01E-06	8.01E-03	2.24E-08	2.24E-03	5.89E-10	5.89E-07
Hg	Adult men (Scenario 1)					
	2.55E-07	8.49E-04	1.02E-09	4.84E-05	3.74E-11	4.37E-07
	Adult women (Scenario 2)					
	3.36E-07	1.12E-03	1.34E-09	6.39E-05	4.95E-11	5.77E-07
	Children (Scenario 3)					
	2.60E-06	8.68E-03	7.29E-09	3.47E-04	1.91E-10	2.23E-06

Table 5 Hazard indices for Pb, Cd, and Hg and Cd cancer risk due to exposure of adult and children populations through soil in the Tuzla Canton

Scenario	HI				Cancer risk (Cd, unitless)
	Pb	Cd	Hg	Total	
Adult men (Scenario 1)	6.06E-02	1.10E-03	8.98E-04	6.26E-02	3.11E-10
Adult women (Scenario 2)	8.00E-02	1.45E-03	1.19E-03	8.26E-02	4.11E-10
Children (Scenario 3)	6.14E-01	1.03E-02	9.03E-03	7.26E-01	3.18E-10

Even though the highest Hg level was expected to be measured near the chloralkali plant as reported earlier (30, 33), it was many times lower, and in fact measured in agricultural soil downstream of the river Spreca. The Spreca and its tributary Jala have been industrial and household wastewater recipients for many years, spreading contaminants over agricultural land during floods. This drop in the maximal level may suggest that Hg leakage at the chloralkali plant has been remedied in the meantime.

Deterministic human health risk assessment

The non-cancer health risk to adults and children from exposure to Pb, Cd, and Hg in soil through ingestion, skin, and inhalation was calculated based on deterministic approach (Table 4). As expected, Pb contributed to the health risk more than the other two heavy metals. Exposure decreased in the following order: ingestion>skin contact>inhalation. Our results are consistent with similar studies pointing to ingestion as the primary route of exposure to heavy metals from soil (9, 15, 53).

Table 5 shows that hazard indices vary greatly between the exposure scenarios. Children are far more susceptible to heavy metal exposure from soil per body weight than adults due to their physiological characteristics and behaviour, particularly in terms of higher hand-to-mouth ingestion. Their overall health risk is about seven times higher than in adult women and 10 times higher than in adult men. In this respect, our findings are in line with those reported elsewhere, which largely depended on measurement sites (15, 41, 54–56). Some researchers (57, 58) found that high blood Pb levels in children were linked with seasonal trends of suspended soil dust, which, in contrast to hand-to-mouth behaviour, implies that inhalation might be the primary route of exposure to Pb from soil. This calls for closer examination of Pb exposure risk associated with fine airborne soil particles in the summer and near the roads (59).

Even though the hazard indices for all three exposure scenarios and all three heavy metals were lower than 1, indicating exposure below risk thresholds, this estimation does not take into account other sources of exposure, such as food, water, and air (22, 60–63).

In order to obtain a more in-depth view of human health risk, we also took an integrative approach to risk assessment under the assumption that the total effect of the three analysed heavy metals would be additive. In other words, even if the metals are present in soil at levels lower than their No Observed Adverse Effect Level (NOAEL), the sum of their levels might lead to increased health risk. In our study, however, the calculated health risk of exposure to a mixture of all three metals for all three scenarios remained below the risk threshold (HI<1; Table 5).

Still, this additive approach has its own limitations and disadvantages, as it completely neglects different mechanisms of action of the analysed heavy metals and

their possible interactions, which might produce weaker or even stronger (synergistic) effects or effects that each metal alone would not exhibit.

As for the cancer risk for Cd exposure through soil, none was found in any of the three scenarios (HI<1; Table 5).

Probabilistic human health risk assessment

Bearing in mind that deterministic risk assessment, which relies on point estimation, is mainly based on mean or median heavy metal levels and the most likely exposure parameters, it is possible that it could either under-or overestimate health risk (5, 64). To minimise uncertainties and errors, risk levels can also be evaluated with probability distribution functions. Table 6 summarises our Monte Carlo simulations, while Figures 2–4 show simulated probability distribution for the hazard index, calculated as the sum of three hazard quotients (HI=HQ_{oral}+HQ_{dermal}+HQ_{inhalation}). Considering individual differences and spatial variations, the hazard indices of Scenarios 1 and 2 for Pb showed 0.6 % probability to exceed the threshold value of 1 (Figure 2).

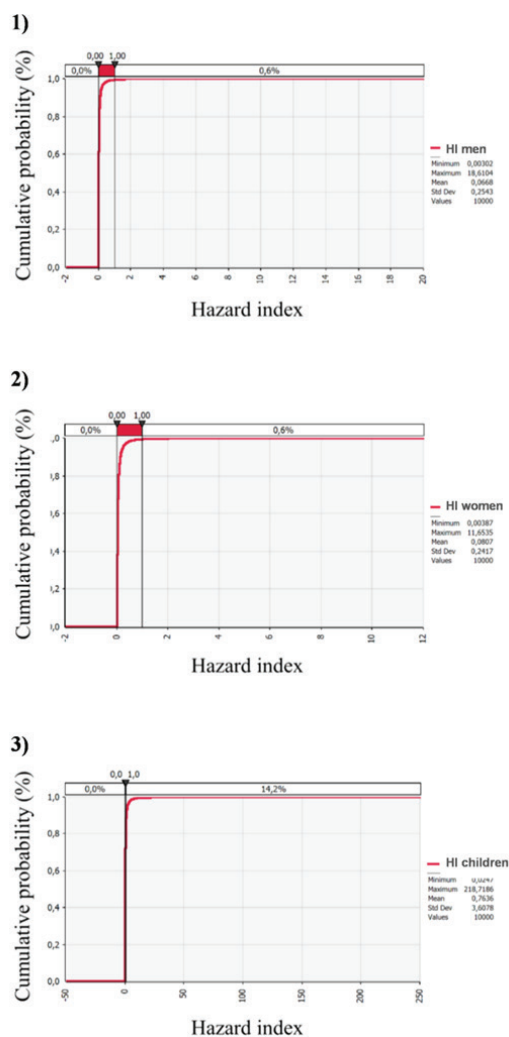


Figure 2 Hazard indices for Pb by Scenarios 1 (men), 2 (women), and 3 (children)

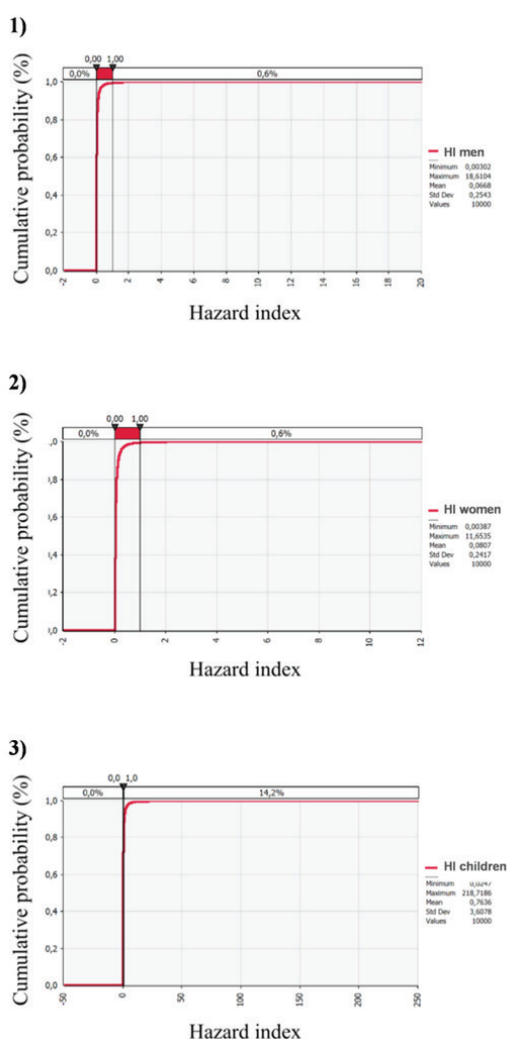


Figure 3 Hazard indices for Cd by Scenarios 1 (men), 2 (women), and 3 (children)

For Scenario 3 (with children), this probability rose to 14.2%. In contrast, the hazard indices for Cd and Hg for all three scenarios showed no probability to exceed the health risk threshold, save for the children scenario for Hg intake (Figures 3 and 4).

Hazard indices higher than 1 were obtained only when exposure assessment was based on the maximal and 95th percentile levels (Table 6) and all point to higher health risk for children. For Pb exposure, they were several times higher than those reported by studies conducted in Spain and China (HI 3.11E-02 and 4.19E-04, respectively) (16, 65). The child average daily exposure to Cd did not exceed the safe reference dose, and the HIs were within the acceptable limits. Unlike Pb, Cd showed several times lower hazard indices compared to the Spanish and Chinese studies (HI 1.09E-03 and 1.09E-05, respectively) (16, 65). Our findings are also in line with other studies conducted in China and reporting higher health risk in children due to soil heavy metals (54, 55).

As for cancer risk associated with exposure to Cd, our Monte Carlo simulations showed no increased risk. However, as soil is not the main source of human Cd exposure (66–68), our estimate should be taken with reserve, and future modelling should combine soil data with other major sources, such as food, air, cigarette smoking, and water.

CONCLUSION

As a strong industrial region, the Tuzla Canton significantly contributes to environmental pollution by heavy metals in Bosnia and Herzegovina, which is apparent from Pb, Cd, and Hg levels in agricultural soil exceeding

Table 6 Summary of health risk assessment for three exposure scenarios based on the Monte Carlo simulation.

Scenario	Concentration	HI (unitless)			Cancer risk (Cd, unitless)	
		Pb	Cd	Hg		
1	Mean	6.68E-02	1.14E-03	8.67E-04	6.88E-02	3.11E-10
	75 th percentile	5.66E-02	1.28E-03	8.46E-04	5.87E-02	5.15E-10
	95 th percentile	2.09E-01	4.08E-03	2.53E-03	2.16E-01	1.16E-09
	Max	1.86E+01	3.46E-02	6.43E-02	1.87E+01	1.69E-09
2	Mean	8.07E-02	1.48E-03	1.19E-03	8.34E-02	4.11E-10
	75 th percentile	7.24E-02	1.68E-03	1.13E-03	7.52E-02	6.80E-10
	95 th percentile	2.46E-01	5.27E-03	3.57E-03	2.55E-01	1.53E-09
	Max	1.17E+01	2.99E-02	2.99E-01	1.2E+01	2.23E-09
3	Mean	7.64E-01	1.18E-02	1.00E-02	7.86E-01	3.18E-10
	75 th percentile	6.07E-01	1.28E-02	9.50E-03	6.29E-01	5.27E-10
	95 th percentile	2.24E+00	4.21E-02	3.22E-02	2.31E+00	1.18E-09
	Max	2.19E+02	6.28E-01	1.24E+00	2.21E+02	1.73E-09

1 – adult men; 2 – adult women; 3 – children. Figures in bold indicate hazard indices above the no-risk threshold (HI>1)

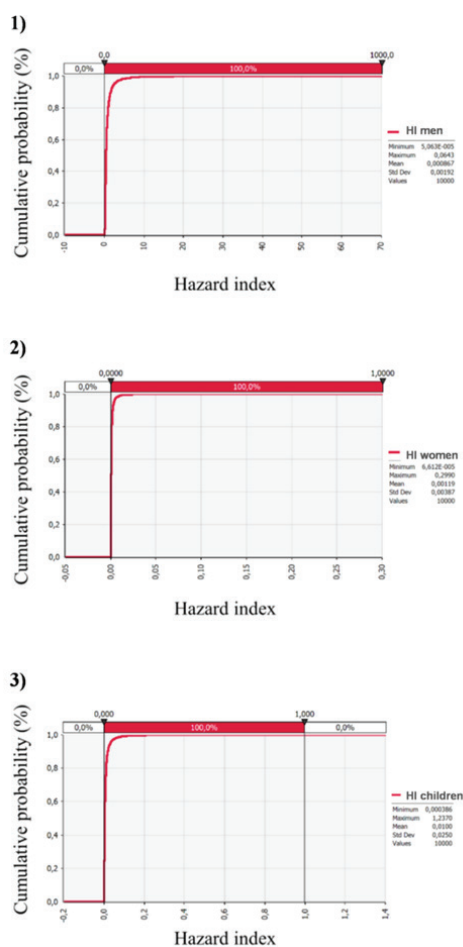


Figure 4 Hazard indices for Hg by Scenarios 1 (men), 2 (women), and 3 (children)

permitted limit values in 71.55 %, 35.61 %, and 8.62 % of soil samples, respectively.

We combined the deterministic and semi-probabilistic approach to assess both cancer and non-cancer health risks, which turned out to provide a better understanding of the issues at hand, as the deterministic assessment alone did not raise red flags, and the Monte Carlo simulation did give some reason for concern, especially with children. Cancer risk of Cd from soil was not established, but this finding should not put our minds at rest, as soil is not the major direct source of human exposure to Cd, especially bearing in mind its persistence in the environment and its bioavailability.

Our study lays the groundwork for further research of the impact of soil contamination on human health in the region through continuous monitoring and health risk assessment that should include other exposure sources, such as air, inhalable dust, water, and food.

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Procjena zdravstvenoga rizika nakon istovremenog izlaganja olovu, kadmiju i živi iz poljoprivrednoga tla s područja Tuzlanskoga kantona (Bosna i Hercegovina)

Cilj ovog istraživanja bio je procijeniti izloženost ljudi olovu (Pb), kadmiju (Cd) i živi (Hg) iz poljoprivrednog tla, uzimajući u obzir pridružene nesigurnosti i varijabilnosti u ključnim parametrima izloženosti. Primjenom metoda induktivno spregnute plazme i atomske emisijske ili apsorpcijske spektrometrije (ICP-AES i AAS) određena je koncentracija metala u uzorcima poljoprivrednoga tla prikupljenog s 29 lokacija u Tuzlanskom kantonu (Bosna i Hercegovina). Koncentracije Pb kretale su se u rasponu od 13,33 do 1692,33 mg/kg, Cd od 0,05 do 3,67 mg/kg i Hg od 0,02 do 2,73 mg/kg. Za procjenu kancerogenog i nekancerogenog rizika koristili smo se determinističkim i semiprobabilističkim pristupom u procjeni rizika. Utvrđeno je da Pb doprinosi povećanom zdravstvenom riziku više nego druga dva teška metala. Indeks opasnosti (eng. *hazard index* – HI) smanjivao se među populacijskim skupinama (djeca > žene > muškarci) i putevima izloženosti (ingestija > dermalni kontakt > inhalacija). Naše Monte Carlo simulacije pokazale su da HI za Pb uključujući obje populacije odraslih imaju 0,6 % vjerojatnosti da će prijeći vrijednost praga od 1, dok je u djece ta vjerojatnost bila 14,2 %. Vjerojatnost da će premašiti prag u bilo kom scenariju nisu pokazali Cd i Hg. Rezultati naših simulacija izazivaju zabrinutost zbog mogućih štetnih učinaka teških metala iz tla, posebice u djece. Vrlo je važno nastaviti pratiti onečišćenje okoliša i procijeniti rizik za zdravlje ljudi, ne samo putem tla, već i putem drugih značajnih dijelova okoliša, poput zraka i vode.

KLJUČNE RIJEČI: deterministička metodologija; kancerogeni rizik; Monte Carlo simulacije; nekancerogeni rizik; probabilistička metodologija; teški metali; uzorci tla