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HEALTH RISK ASSESSMENT OF HEAVY METALS IN THE LANDS AND PLANT CULTIVARS OF ZENICA MUNICIPALITY, BOSNIA AND HERZEGOVINA

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ABSTRACT

Estimating the risk to public health from heavy metals can help address basic questions about their potential dangers. This is the first study aimed to assess the health risk for the residents of the municipality of Zenica, on the land closest to the steel industry, by examining the heavy metals Zn, Ni, Pb, Cd, Cr, and Cu in the soil and plant cultivars corn and chard that could be used for the potential remediation of such soil. Using the calculated data for HQ and HI, the non-carcinogenic and carcinogenic risks for adults and children were estimated based on heavy metals in the selected topsoil and plants commonly grown in the area of interest, through different routes of intake. The results showed that there is an unacceptable risk for children and adults due to long-term consumption of investigated plant cultivars from soil contaminated with heavy metals. Constant monitoring and measures to reduce the heavy metal pollution, primarily Cd, Pb, and Cr, are necessary in the lands of the municipality of Zenica. Special caution is required for residents who intend to grow plant cultivars near the steel industry.

Keywords: Heavy metals, plant cultivars, soil, and health risk assessment.

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1. INTRODUCTION

Risk analysis, as a paradigm, is science-based, problem-oriented, performed with fresh data, draws on information from different sources, makes the best decision at the most appropriate time, and produces flexible, up-to-date decisions. Risk assessment is a complex process for determining the risks related to some types of hazards in food - biological, chemical, or physical [1,2]. Risk characterization aims to assess the nature and likelihood of harm as a result of human exposure to various pollutants from plants and the land where they are grown [1,2]. Furthermore, it provides structured information that helps decision makers identify interventions that have public health significance and provides a basis for the use of various options. These options include regulatory measures when necessary. Risk assessment can also be used to identify collected data and target research that should increase the value of the assessment. In the future, risk assessment can help in the planning of science-based remediation plans [3,4]. Namely, the basis that can be used for risk assessment helps in the identification of hazards whose occurrence is possible [2,5,6,7]. The real benefit is that the hazard is defined as the risk of negative health consequences, before it becomes contamination in the food chain [8]. With the introduction of risk analysis posed to heavy metals pollution worldwide, a new way of thinking about problems has been introduced. Many problems (e.g., food and environmental safety) still occurred and increased. The steel industry-associated activities, coal combustion, and intensive traffic in Zenica's municipality have become the main anthropogenic routes by which residents are exposed to elevated levels of toxic elements (such as Zn, Ni, Pb, Cd, Cr, and Cu) exceeding normal background levels [1,9,10]. Urban and agricultural lands are the major sink for these metals released into the environment, whereas they can negatively affect the chemical and biological soil quality [6,11,12,13]. Because of their potential toxicity, persistence, and bioaccumulation, elevated levels of these elements in the environment pose a serious threat to the public health of Zenica's population and cause serious concern, especially from malignant diseases [14,15,16]. Of particular concern is the increase in the number of patients and deaths from different kinds of cancer cause of heavy metal intoxications [17-20]. That is not surprising if it is known that in the winter months, a "cover" is formed over the small and narrow Zenica valley, under which all the toxic substances released from the chimneys and plants of the ironworks and other anthropogenic emissions remain as the toxic living atmosphere for the residents [11,15]. Many studies have confirmed similar health problems related to the toxic effects of heavy metals, especially in environments near various industrial plants, around the world [10,12,22,23,24]. The goal of this work was estimating the risk to adults and children health based on the content of heavy metals in the land of the potential agricultural area of the municipality of Zenica, where Zn, Ni Pb, Cd, Cr and Cu were identified as potential dangerous inorganic pollutants in the observed topsoil and plant cultivars, that are oftenly planted in this area and could serve for its potential phytoremediation.

2. EXPERIMENTAL PART

Topsoil and related plant cultivars (corn and chard) were collected and prepared from selected areas in the municipality of Zenica, near the steel company ArcelorMittal Zenica [2,25]. Specific locations (about 1000 m^2 each) for investigation were: Tetovo 44 13'52''N, 17 53'18''E, Podbriježje 44 21'17''N, 17 88'70''E, and Gradišće 44 24'04''N, 17 87'66''E, where the selected plants: corn and chard were cultivated. All samples were analyzed for the total content of the heavy metals by using an AAS method (Perkin Elmer spectrometer 3110), according to standard ISO 11466 and the Official Gazette of the Federation of Bosnia and Herzegovina [26]. In the accredited labs of the "Kemal Kapetanović" Institute, University of Zenica, all chemical analyses were performed.

a) Calculation of health risk assessment for soil:

The potential health risk assessment calculated for a lifetime of exposure (ingestion, inhalation, and dermal) based on USEPA model [27-32]., was determined as the cumulative carcinogenic and non - carcinogenic risk. The value of the total hazard index is calculated for the maximum content of heavy metals for adults and children. Non-carcinogenic and carcinogenic effects of heavy metals in soil were estimated by using equations:

$$ADDing = CM\ X\ \frac{IngR\ X\ EF\ X\ ED\ X\ CF}{BW\ X\ AT}$$

$$ADDinh = CM\ X\ \frac{InhR\ X\ EF\ X\ ED}{PEF\ X\ BW\ X\ AT}$$

$$ADDder = CM\ X\ \frac{SA\ X\ AF\ X\ ABS\ X\ EF\ X\ ED\ X\ CF}{BW\ X\ AT}$$

Where ADD_{ing} - Average daily doses (mg/kg day) via ingestion; ADD_{inh} - Average daily doses (mg/kg day) via inhalation, and ADD_{der} - Average daily doses (mg/kg day) via dermal contact.

Hazard identification basically aims to investigate pollutants that are present at any given location, their content, and spatial distribution. In the study area, Zn, Ni, Pb, Cd, Cr, and Cu were identified as possible hazards for the community. The non-carcinogenic effects of the elements were assessed by calculating the hazard coefficient and hazard index.

$$HQ = \frac{ADD}{RfD}$$

where HQ-hazard coefficient, RfD- Chronic reference dose (mg/kg day) of specific heavy metal, and HI-hazard index.

$$HI = \sum_{k=1}^{n} HQ_k = \sum_{k=1}^{n} \frac{ADD_k}{RfD_k}$$

The risk index (RI) predicts the potential cancer risk for children and adults in the study area by integrating all gathered information to produce quantitative estimates of cancer risk and hazard indices. Due to the lack of available values for slope factors (SF), we estimated carcinogenic risk only for exposure to Cd, Pb, and Cr, whose concentrations were the highest among the measured concentrations in the tested samples.

$$RI_{pathw} = \sum_{k=1}^{n} ADD_k\ CSF_k$$

where RI is unitless probability of an individual developing cancer over a lifetime; ADD_k is Average daily intake (mg/ kg day) for kth heavy metal, and CSF_k is Cancer slope factor $(mg/\ kg\ day)^{-1}$ for kth heavy metal.

$$RI_{total} = RI_{ing} + RI_{inh} + RI_{der}$$

where RI_{ing}, RI_{inh}, and RI_{der} are risk contributions through ingestion, inhalation, and dermal pathways.

The assessment is based on the following: If the elemental content of the metal is less than Rf, then HQ ≤ 1, which means that there are no adverse health effects. In the case that elemental content value is above Rf (HQ > 1), then adverse health effects are probable [28]. An HQ value higher than 10 indicates a high chronic risk for carcinogenic effects of the elements [22]. HI is the sum of all non-carcinogenic hazard quotients of the elements across all three exposure routes. If HI < 1, there is no risk of non-carcinogenic effects, while HI > 1 indicates a probability of adverse health effects. The RI is the probability that an individual will develop any type of cancer in their lifetime due to exposure to carcinogenic hazards. If the calculated RI < 10^{-6}, the carcinogenic health risk can be considered negligible. If the RI is in the range of 1×10^{-6} to 1×10^{-4}, then these values are within the acceptable risk to human health. If RI > 10^{-4}, then there is a high risk for developing cancer in humans.

b) Calculation of health risk assessment for plant cultivars consumption

To estimate the non-carcinogenic and carcinogenic health risk from consuming the plant cultivars (alfalfa, corn, and chard) contaminated with heavy metals, the target hazard quotient (THQ) was calculated as per the US EPA Region III Risk- Based Concentration Table. The THQ is calculated by the following equation:

$$THQ = \frac{EDI}{RfD}$$

where EDI is estimated daily intake; target hazard coefficient THQ is non-carcinogenic risk and is dimensionless; RfD is the reference dose of individual metal (mg/kg/ day).

Table 1. Parameters for health risk assessment of heavy metals in soil samples [27]

Factor	Definition	Unit	Value	
			Adults	**Children**
CM	Heavy metal content in soil or plants	mg/kg		
IngR	Ingestion rate of soil	mg/day	100	200
EF	Exposure frequency	days/year	350	350
ED	Exposure duration	years	30	6
BW	Body weight of the exposed individual	kg	70	15
AT	Average time	days	365ED	365ED
InhR	Inhalation rate of soil	m^3/day	20	10
PEF	Particle emission factor	m^3/day	$1.36x10^9$	$1.36x10^9$
SA	Exposed skin surface area	cm^2	5700	2800
AF	Skin adherence factor	mg/cm^2	0.07	0.2
CF	Calculation factor	kg/mg	10^{-6}	
ABF	Dermal absorption factor	none	0.1	0.1

Table 2. Reference doses and Cancer Slope factors for heavy metals [29]

Heavy metal	RfD_{ing} (mg/kg/dan)	RfD_{inh} (mg/kg/dan)	RfD_{der} (mg/kg/dan)	Oral CSF	Inhalation CSF
Zn	$3.00×10^{-1}$	$3.00× 10^{-1}$	$6.00×10^{-2}$		
Ni	$1.00×10^{-1}$	$2.00×10^{-3}$	$5.00×10^{-3}$		$8.40×10^{-1}$
Pb	$3.00×10^{-1}$	$3.00×10^{-3}$	$5.00×10^{-4}$	$8.50×10^{-3}$	$4.20×10^{-2}$
Cd	$1.00×10^{-3}$	$1.00×10^{-3}$	$1.00x10^{-5}$	15	6.30
Cr	$3.00×10^{-3}$	$2.00×10^{-4}$	$7.00×10^{-3}$	$5.00×10^{-1}$	$4.10×10^{1}$
Cu	$4.00×10^{-2}$	$4.00×10^{-1}$	$1.00×10^{-2}$		

The reference oral dose (mg/kg/day) of selected metals is given in Table 2. To assess the overall potential health risk posed by different plant cultivars, the total hazard coefficient (TTHQ) was also calculated, where TTHQ <1 is safe, and TTHQ >1 is hazardous. To estimate the overall potential for non-carcinogenic health risk from all investigated heavy metals, HI can be calculated by the sum of the TTHQ metals of each plant cultivar [13]. Furthermore, carcinogenic risk (CR) was calculated using an equation:

$$CR = EDI\ X\ \ CSF\ oral$$

Generally, total CR values lower than 1 x 10^6 are considered to be minor, above 1 x 10^{-4} are unacceptable, and lying in the range of $1x10^{-6}$ to $1x10^{-4}$ is an acceptable range.

Quality control: The pre-cleaned tools, high-quality reagents, and standards were used, and three sample replicates were done during the AAS measurements of heavy metal content in topsoil and plant cultivars. Measured data for the total content of heavy metals were the subject of descriptive statistics. Relevant USEPA guidelines and ISO standards, and the environmental Bosnia and Herzegovina rulebook were used for performing the chemical analysis [26,33]. The calculations were done using Microsoft Excel software.

3. RESULTS AND DISCUSSION

Heavy metals topsoil analysis resulted in the following average data ranges (mg/kg): 30-100 for Zn, 40-120 for Ni, 20-70 for Pb, <1 for Cd, 30-180 for Cr and 20-40 for Cu. Generally, the results obtained from AAS measurements of selected heavy metals indicated mild to moderate pollution of the investigated land area in Zenica. Enrichment factors (EF) were also calculated (as a ratio between the content of heavy metal in polluted soil and content of heavy metal in control soil), and obtained data were in the range: 0.7-1.3 for Zn, 0.8-3.6 for Ni, 0.7 -3.7 for Pb, 1 for Cd, 0.8 -1.4 for Cr, and 0.8 -1 for Cu. EF was calculated as a ratio to assess the pollution level of selected lands, where EF < 2 indicates anthropogenic non-harmful pollution, and EF 2-5 indicates moderate pollution. **T**he results showed the highest heavy metal pollution in the Gradišće area, in the following order: Ni > Pb > Zn > Cr > Cd > Cu. Similar results were obtained in other studies [1,9-11]. Furthermore, heavy metals analysis of selected plant cultivars resulted in the following average data ranges (mg/kg): 1-14 for Zn, 11-53 for Ni, 1-53 for Pb, 1-3 for Cd, 1-44 for Cr, and 1-39 for Cu. Regular monitoring of these lands is necessary in the goal of measuring the heavy metal content related to the soil matrix and possible enrichment caused by human activities and industrial emissions.

a) Soil non-carcinogenic and carcinogenic risk assessment

The values of average daily doses for adults and children are presented in Tables 3 and 4.

Due to soil exposure, the non-carcinogenic HQ and HI for adults and children were estimated using the 95th percentile value of the total metal concentration. The obtained results of the health risk of non-carcinogenic substances for adults and children, based on maximum metal concentrations in the tested soil and exposure through three different routes (ingestion, inhalation, and through the skin), are presented in Tables 5 and 6.

The data on risk pathway and total risk for adults and children are presented in Tables 7 and 8.

Potential carcinogenic risk will occur from Cr for children, as shown by the obtained results. Similar findings were obtained in foreign works where heavily polluted lands were investigated [13, 34].

b) Plants non-carcinogenic and carcinogenic risk assessment

The data of estimated daily intake, overall THQ, and HI for adults and children are presented in Tables 9 and 10

Results (Table 9) showed that the highest EDI contributions were of Pb and Cu from corn, especially for children.

This is in accordance with other similar investigations related to long-term consumption of cereals and vegetables cultivated in the polluted areas worldwide [3,6,13,35]. Corn is a plant cultivar with a very high bioaccumulation factor of investigated heavy metals [3, 36].

Table 3. Calculated values of ADD (mg/kg/day) of soil samples for adults

Heavy metals pathway	ADD (mg/kg/day)					
	Zn	Ni	Pb	Cd	Cr	Cu
ingestion	1.4×10^{-4}	1.7×10^{-4}	9.8×10^{-5}	1.4×10^{-6}	2.5×10^{-4}	5.6×10^{-5}
inhalation	2.8×10^{-8}	2.4×10^{-8}	1.4×10^{-8}	2×10^{-10}	3.6×10^{-8}	8×10^{-9}
dermal	3.3×10^{-5}	3.4×10^{-5}	2.3×10^{-5}	3.3×10^{-7}	5.9×10^{-5}	1.3×10^{-5}
total	3.4×10^{-3}	2×10^{-4}	1.2×10^{-4}	2.1×10^{-6}	3.1×10^{-4}	6.9×10^{-5}

Table 4. Calculated values ADD (mg/kg/day) of soil samples for children

Heavy metals pathway	ADD (mg/kg/day)					
	Zn	Ni	Pb	Cd	Cr	Cu
ingestion	6.4×10^{-4}	7.7×10^{-4}	4.5×10^{-4}	6.4×10^{-6}	3.1×10^{-3}	2.6×10^{-4}
inhalation	4.7×10^{-8}	5.6×10^{-8}	3.3×10^{-8}	4.7×10^{-10}	8.5×10^{-8}	1.9×10^{-8}
dermal	2.2×10^{-4}	2.6×10^{-4}	1.5×10^{-4}	2.2×10^{-6}	4×10^{-4}	8.8×10^{-5}
total	8.6×10^{-4}	1×10^{-3}	6.4×10^{-4}	8.6×10^{-6}	3.5×10^{-3}	3.5×10^{-4}

Table 5. Calculated HQ and HI values for adults

Different pathway	HQ					
	Zn	Ni	Pb	Cd	Cr	Cu
ingestion	4.7×10^{-4}	8.5×10^{-3}	2.7×10^{-2}	2.8×10^{-3}	8.3×10^{-2}	1.5×10^{-3}
inhalation	9.3×10^{-8}	1.2×10^{-5}	4.7×10^{-6}	3.5×10^{-6}	1.8×10^{-4}	-
dermal	4.4×10^{-4}	6.7×10^{-3}	4.6×10^{-2}	6.6×10^{-4}	8.4×10^{-3}	5.4×10^{-4}
HI	9.1×10^{-4}	1.5×10^{-2}	7.3×10^{-2}	3.5×10^{-3}	9.2×10^{-2}	2×10^{-3}

Table 6. Calculated HQ and HI values for children

Different pathway	HQ					
	Zn	Ni	Pb	Cd	Cr	Cu
ingestion	2×10^{-3}	3.9×10^{-2}	1.3×10^{-1}	1.3×10^{-2}	1	7×10^{-3}
inhalation	1.6×10^{-7}	2.8×10^{-5}	1.1×10^{-5}	8.2×10^{-6}	4.3×10^{-4}	-
dermal	2.9×10^{-3}	4.6×10^{-2}	3×10^{-1}	4.4×10^{-3}	5.7×10^{-2}	3.7×10^{-3}
HI	4.9×10^{-3}	8.5×10^{-2}	4.3×10^{-1}	1.7×10^{-2}	1.1	1.1×10^{-2}

Table 7. Calculated risk pathway and total risk for adults

Different pathway	Risk pathway		
	Pb	Cd	Cr
ingestion	8.3×10^{-7}	2.1×10^{-7}	1.3×10^{-6}
inhalation	5.9×10^{-10}	12.6×10^{-10}	1.5×10^{-8}
total tisk	8.3×10^{-7}	2.1×10^{-7}	1.3×10^{-6}

Table 8. Calculated Risk pathway and total risk for children

Different pathway	Risk pathway		
	Pb	Cd	Cr
ingestion	3.8×10^{-7}	9.6×10^{-5}	1.6×10^{-3}
inhalation	1.4×10^{-9}	2.9×10^{-9}	3.5×10^{-6}
Total Risk	3.8×10^{-7}	9.6×10^{-5}	1.6×10^{-3}

Table 9. Calculated EDI (mg/kg/day) for adults (a) and children (c)

Plant cultivar	EDI (mg/kg/day)											
	Zn		Ni		Pb		Cd		Cr		Cu	
	a	c	a	c	a	c	a	c	a	c	a	c
corn	0.001	0.007	0.020	0.096	0.032	0.152	0.002	0.010	0.010	0.048	0.031	0.147
chard	0.001	0.007	0.030	0.144	0.001	0.007	0.003	0.016	0.001	0.007	0.001	0.007

Table 10. THQ, TTHQ, and HI data for adults (a) and children (c)

Plant cultivar	THQ											
	Zn		Ni		Pb		Cd		Cr		Cu	
	a	c	a	c	a	c	a	c	a	c	a	c
corn	0.005	0.022	1.005	4.745	9.057	43.4	4.2	20.2	3.3	16	0.77	3.685
chard	0.005	0.022	1.505	7.205	0.400	1.91	6.6	31.2	0.47	2.2	0.035	0.168
TTHQ	0.010	0.044	2.510	11.950	9.457	45.31	10.8	51.4	3.77	18.2	0.805	3.853
HI adult	18.337											
HI children	42.705											

As the THQ data in Table 8 showed, potential non-carcinogenic risk posed to Cd, Pb, Cr, and Ni is seriously high for children as well as for adults for long-term consumption, especially of corn in the investigated lands. Similar results for the highest values of THQ and HI were found in the papers about health risk assessment of soil and edible plants in different countries, especially in areas near heavy industries - steel, petrochemical, etc. [5,6, 13,34].

The data of CR and total CR for adults and children are presented in Table 11.

Table 11. CR and total CR data for adults (a) and children (c)

Plant cultivar	CR					
	Cd		Pb		Cr	
	a	c	a	c	a	c
corn	3.2×10^{-2}	1.5×10^{-3}	2.7×10^{-4}	1.3×10^{-3}	5×10^{-3}	2.4×10^{-2}
chard	5×10^{-2}	2.3×10^{-3}	2×10^{-5}	5.7×10^{-5}	7×10^{-4}	3.4×10^{-3}
Total CR	8.2×10^{-2}	3.8×10^{-3}	2.9×10^{-4}	1.3×10^{-3}	5.7×10^{-3}	2.7×10^{-2}

Results showed that the potential carcinogenic risk calculated for target metals such as Cd, Pb, and Cr is unacceptable for children as well as for adults in the municipality of Zenica, as the CR values were above the safe limit of 10^{-4}.

4. CONCLUSIONS

Our study assessed both non-carcinogenic and carcinogenic health risk due to exposure to heavy metals Zn, Ni, Pb, Cd, Cr, and Cu through the soil - plant human contamination pathway next to the typical steel industry in the area of the municipality of Zenica. The soil was polluted with different metals, which was shown by the results of the chemical analysis of the selected metals and the calculated EF data. Particularly worrisome data were obtained for Cd, Ni, Pb, and Cr after the evaluation of the health risk for the soil and the tested plant cultivars through different routes of intake of these heavy metals. Our study could serve as a model for similar estimations that would deal with public health risk assessment in areas that are particularly contaminated by known industrial emitters.

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Conflict of Interest

The authors declare no conflict of interest.

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