Human Health Risk Assessment of Heavy Metals from the Agricultural Soil in South Herzegovina

Aida ŠUKALIĆ [1]
Nihada AHMETOVIĆ [2]
Sejad MAČKIĆ [1]
Alma LETO [1]
Ahmed DŽUBUR [1]
Boris ANTUNOVIĆ [3](✉)

Summary

Contamination of agricultural soils can present a significant risk to human health through oral ingestion, particle inhalation, and dermal contact. The aims of this research were to determine the concentrations, distribution and human health risk of various heavy metals in soil samples from three agricultural areas of South Herzegovina. A total number of 32 soil samples were collected and analyzed for Lead (Pb), Cadmium (Cd), Cobalt (Co), Copper (Cu), Nickel (Ni), and Zinc (Zn). The Hazard Index (HI) was used to assess the human health risk of the study area. For the adult and children population, the HI value for dermal exposure to Cobalt (Co) was greater than one (HI>1), and non-cancerogenic effects are therefore considered as significant for human health. Our findings impose consideration of taking risk management measures in order to reduce risk for human health from Cobalt (Co).

Key words

heavy metals, soil, risk assessment

[1] Agromediterraneane Faculty, University Džemal Bijedić,
U.S.R.C. Mithad Hujdur Hujka, Mostar 88104, Bosnia and Herzegovina
[2] Faculty of Medicine, University of Tuzla, Univerzitetska 1, Tuzla 75000, Bosnia and Herzegovina
[3] Faculty of Agriculture, University of J.J. Strossmayer, V. Preloga 1, Osijek 31000, Croatia
✉ e-mail: boris.antunovic@pfos.hr

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Introduction

In recent years more attention has been paid to the contamination of land, water, air, and their entry into the food chain. Contamination of agricultural land is caused by the introduction of harmful substances by water, air or their accumulation on or in the soil at concentrations above allowed. Soil contamination with heavy elements used for agricultural purposes is the result of atmospheric precipitation, application of pesticides, artificial fertilizers and irrigation with water of poor quality (Škrbić and Đurišić-Mladenović, 2013). Agrochemical measures that contribute to the metal accumulation in the soil include: fertilization with mineral fertilizers, organic fertilizers, landscaping (application of improvers, liming, and plastering), application of pesticides and irrigation. Heavy metals accumulate in soil profiles over a longer period of time because they are not biodegradable and cause long-term consequences in terms of fertility and soil quality, while some may cause toxicity in plants, but also in humans and animals if such plants are grown and used for food production (Dellantonio *et al*., 2007, 2008; Mulligan *et al*., 2001; Nicholson *et al*., 2003). The investigated sites have a different soil composition. The type of land at the locality of Čapljina is alluvial carbonate clay soil. They occupy an area of 8.178 ha, which makes up 1.9% of the area of Herzegovina-Neretva Canton. They are located on the left and right extended valley of the Neretva River. The type of soil on the location of Mostar is brown charcoal skeleton anthropogenized pebble land. These soils are very permeable, airy and very skeletal. They do not hold water well, so soaking is one of the major agro-healing measures, which is being applied in Mostar Field.

The wider area of Stolac is covered with reddish-brown anthropogenized soils on a flysch in a complex with brown anthropogenized soils. Red soils are very shallow on the lime trees and occupy the lower and bumpy positions of the karst relief. Exposure to metals can result in negative health effects, and for this reason European and international agencies in the area of risk assessment define a set of health-based limit values and a risk assessment methodology for the purpose of protecting human health. The basic elements of risk assessment of chemical hazards include four steps: identification of hazards, characterization of hazards, assessment of exposure and characterization of risks. Methods of estimation of non-cancerous risk are usually based on the use of that part as a target hazard (HQ), the ratio between the estimated dose of the pollutant and the reference dose below which there will be no significant risks (USEPA, 2000). And if such a ratio exceeds the whole, there may be concern about possible health effects (Rupert, 2007). Heavy metals are ecologically very important because they are non-destructive; they continuously pass through the ecosystem and have a certain biogeochemical cycle. Circumference intensity is uneven over the year and depends on climatic conditions, proximity to polluting sources and biological system activities. Some of them, such as iron, manganese, copper, zinc, molybdenum, cobalt, vanadium and strontium, belong to a group of essential elements and in small quantities are necessary for many functions of the living organism. At higher concentrations, they exhibit toxic effects and if they engage in a food chain, they pose a danger to human and animal health. Some heavy metals, such as lead, nickel, arsenic and mercury, exhibit toxic effects in the case of their value above MDK in the environment. Their anthropogenic redistribution causes increased penetration in food chains and causes various disorders both in natural and anthropogenic ecosystems.

Absorption of heavy metals in the body cannot be completely avoided, but when accumulated in the body can cause very serious health problems (Singh *et al.,* 2003). The effects of heavy metals on living organisms can be carcinogenic and mutagenic. Heavy metals toxicity in children can result in neurological damage (decreased intelligence, memory loss, and learning and coordination problems) (Alkorta *et al*., 2004). Cadmium can replace zinc, calcium and potassium in the body. It retains in kidneys and livers for up to 40 years, as well as in bones. It is primarily toxic to the liver and kidneys, digestive tract, and chronic intake with insufficient calcium intake or high intake can be embedded in the bones and causing the disease, so-called *Itai-itai*, i.e., bone pain disease. Exposure to lead may occur in several ways, including inhalation of air and intake of food, water, soil or dust. Excessive exposure to lead can lead to epileptic attacks, mental retardation and behavioral disorders (Iqbal, 2012). The danger has been increased due to low mobility of lead in the environment (Lasat, 2000). Zinc is in the soil due to atmospheric rainfall, applying various measures in agriculture (fertilization, pesticide application), discharge of sewage sludge, waste from the metal industry, as well as ashes. Zinc fossil from different sources is different and depends on its chemical properties and its affinity to a particular soil, and it is therefore difficult to determine the overall level of zinc contamination (Kabata-Pendias, 2011). In humans, toxic effects due to excessive cobalt intake appear in chronic form. Cobalt inhalation can lead to bronchial asthma, and exposure to cobalt salts may have a detrimental effect on the skin, the thyroid gland, the lungs, the heart, and the bone marrow (Swennen *et al.,* 1993). The amount of nickel above permitted in the organism causes various toxic phenomena. Symptoms of nose poisoning are manifested by headache, dizziness, nausea, vomiting, chest pain and cough. Copper, besides being an essential element, can also be toxic. Acute poisoning is caused by the intake of this amount of metal above permitted, while chronic poisoning can occur and by the introduction of relatively small amounts of copper over a longer period of time. Symptoms of acute poisoning include abdominal pain, nausea, vomiting and diarrhoea, and in extreme cases death may also occur. Chronic poisoning usually results from the introduction of contaminated water and food as a result of their contact with the copper pipe or the food and cooking utensils. Risk assessment for human health is a process used to assess the health effects that may be the result of exposure to carcinogenic and non-cancerous chemicals. The risk assessment process consists of four basic steps: hazard identification, exposure estimation, toxicity (dose-response) and risk characterization. Identification of hazards basically aims to investigate the chemicals present at any site, their concentrations and spatial distribution. Pb, Cd, Co, Ni, Cu and Zn are identified as potential hazards to the community in the test field.

Material and methods

During the years 2015 and 2016, the level of soil contamination with heavy metals; such as, lead, cadmium, cobalt, nickel, zinc, iron, and copper was investigated in three selected locations (Mostar, Stolac, Čapljina) on which two cultivars of nectarine were cultivated. The pH in KCl at the Stolac site ranged from 4.36 to 5.63, and in H_2O from 6.32 to 6.94. The pH in KCl at the Mostar site ranged from 7.34 to 7.54, and in H_2O from 7.94 to 7.97. At the Čapljina site, the pH in KCl ranged from 7.88 to 8.18, and in H_2O from 6.46 to 8.41. An assessment of the risk to human health exposed by heavy metals from the soil was carried out. The work material implies all input-inputs in the surveyed sites. The sample plants had different surfaces, were of different types of soil, profiles and exposures. The irrigation system of the "drop-in-cap" type is used in the observed sites. In the locality of Mostar and Stolac, two varieties of nectarines, *Caldesi 2000* and *Big Top*, are located on the same parcel, and on the location of Capljina they are planted on different parcels which are about 1000 m apart. A total of 32 samples of land have been collected. The chemical analyzes included the analysis of heavy metals in the total form of lead, cadmium, copper, cobalt, nickel and zinc, and were made in the laboratory of the Institute for Agropedology in Sarajevo. Research work consists of three parts: field research, laboratory research and risk assessment. Descriptive statistics were produced for all the collected results.

Sampling

Sampling of soil was performed with two depths of 0-30 cm and 30-60 cm. The samples were prepared by crushing, packing in plastic bags and labelling. Then, the samples were submitted to the Agropedology Institute in Sarajevo for analysis of the total content of heavy metals and all the other physical and chemical properties of the soil were examined in the laboratory of the Faculty of Agro-faculty of the University "Džemal Bijedić" In Mostar. In the locality of Mostar and Stolac, two varieties of nectarines, *Caldesi 2000* and *Big Top*, are located on the same parcel, and on the location of Čapljina they are planted on different parcels which are about 1000 m apart.

Sample analysis

Preparation of samples for instrumental analysis of heavy metal content in the soil was done by means of aqua regia, and then their content in the extract was determined by atomic absorption spectrometry (AAS). The heavy metal extraction in gold foil was carried out according to the international standard ISO11464. This standard specifies the method of extraction of trace elements by gold-plating using appropriate atomic spectrometric techniques. According to the principle of this standard, soil samples are cut into particles of less than 2 mm to digestion with aqua regia. Such grinding results in obtaining a homogeneous sample from which the sub-action is taken and increasing the efficiency of the action of acid by increasing the surface of the particles. The dried sample was then extracted with a chloride / nitric acid mixture at room temperature for 16 hours, followed by refluxing for two hours. Extract was clarified - purified (filtered) and volume supplemented with nitric acid. The International Standard ISO11047 specifies the atomic absorption spectrometry method for determining one or more elements in soil extract extracted with aqua regia obtained in accordance with ISO11466.

Risk evaluation

The purpose of exposure estimation is to measure or estimate the intensity, frequency and duration of human exposure to the unclean environment. In the study, exposure estimation is performed by calculating the acceptable daily intake (ADI) of heavy metals due to swallowing, inhalation and dermal contact between adults and children in the study area. Adults and children are separated due to their behavioural and physiological differences. Given the various adverse effects that heavy metals may have on human health, the corresponding non-cancerous risks are calculated for children and adults according to the USEPA risk assessment model:

$$ADI_{ing} = \frac{C \times IR \times EF \times ED \times CF}{BW \times AT}$$

$$ADI_{inh} = \frac{CxIR_{air} \times EF \times ED}{PEF \times BW \times AT}$$

$$ADI_{dermally} = \frac{C \times SA \times FE \times AF \times ABS \times EF \times ED \times CF}{BW \times AT}$$

In the equation ADI_{ing}, ADI_{inh}, $ADI_{dermally}$ stand for chronic daily intake. Those abbreviations stand for: $ADI_{ing\,=}$ acceptable daily for orally (mg/kg/d); $ADI_{dermally}$ = acceptable daily intake for dermal (mg/kg/d); ADI_{inh} = acceptable daily inhalation intake (mg/m^3 for cancerous and g/m^3 for non-cancerous elements). C stands for concentration of heavy metals in mg/kg in soil, IR in mg/d ingestion factor, IR_{air} in m^3/d inhalation factor, EF in days/years frequency of exposure, ED duration of exposure in year, BW body weight in kg, AT is the average dose expressed in days, SA is the skin surface exposed in cm^2, FE is the factor of the exposure of the skin to the ground, AF soil binding factor in mg/cm^2, ABS is a dermal absorption factor, CF chronic conversion factor in kg/mg. The exposure parameters used to assess health risks through various exposure paths to soil have been presented in Table 1, whilst the reference doses (RfD) in mg/kg day have been presented in Table 2 (Kamunda *et al.*, 2016).

Table1. Exposure parameters used to assess health risks through various exposure paths to soil (Kamunda *et al.*, 2016.)

Parameters	Unit	Definition	Valuation		Reference
			Children	Adults	
ABS	--	Dermal absorption factor	0.1	0.1	6
AF	mg/cm^2	Soil adhesion factor for skin	0.2	0.07	6
BW	Kg	Average weight	15	70	6
ED	Year	Exposure time	6	30	6
EF	d/year	Exposure frequency	350	350	6
FE	--	dermal exposure ratio	0.61	0.61	6
IngR	mg/d	Soil ingested factor	200	100	6
IR_{air}	m^3/d	Inhalation factor	10	20	6
SA	cm^2/event	Exposed skin surface	2.800	5.700	6
AT_{nc}	D	Average Time for Certain non-cancerous hazards	ED × 365		6
AT_{ca}	D	Average time for cancerous hazards	LT × 365		6
CF	kg/mg	Calculation factor	10^{-6}		5
PEF	m^3/kg	Soil particulate emission factor - air	1.36×10^9		6

Table2. Reference doses (RfD) in mg/kg day (Kamunda *et al.*, 2016)

	Pb	Cd	Co	Cu	Ni	Zn
Oral RfD	3.60E-03	5.00E-04	2.00E-02	3.70E-02	2.00E-02	3.00E-01
Inhal RfD		5.70E-05	5.70E-06			
Derm RfD		5.00E-04	5.70E-06	2.40E-02	5.60E-03	7.50E-02

Calculating non-cancerous risk

Non-cancerous risk was calculated using the term "hazard quotient" (HQ). HQ is the statistical term of the ratio of two variables that expresses the likelihood of an adverse effect on an individual. It is defined as the reference value of the acceptable daily intake (ADI) and chronic reference dose (RfD) in mg/kg/day of specific heavy metals as shown in the equation:

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

For n number of heavy metals, the noncancerous effects for the population are the result of the sum of all HQs for particular heavy metals. This is known under the Hazard Index (HI) and is described in the USEPA (U.S. Environmental Protection Agency, 2004). The mathematical equation is shown as follows:

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

Namely, HQk, ADIk, RfDk are the values of metals. If the HI value is less than 1, then the exposed population will not have harmful non-cancerous effects. If the value of HI is greater than 1, then there is concern about the possible risk to the health of the exposed population of non-cancerous hazards.

Results and discussion

The average concentrations of heavy metals at all locations ranged from Zn> Pb> Ni> Co> Cu> Cd to the following values in mg/kg: Pb 25.40±85.00, Cd 0.60±1.00, Co 20.60±40.60, Ni 23.80±44.20, Cu 19.00±57.70, Zn 48.80±144.20 (Tables 3 and 4).

According to the literature data (Jakšić *et al.*, 2013), in the soil samples of investigated sites, an increase in lead content ranging from 186.80-203.20 mg/kg was established, exceeding the maximum allowable amount. This research is in line with our research, as they are located in the Mostar site, where sandy land at depths of 0-30 cm was 51.8 mg/kg in 2015 and 52.1 mg/kg of lead in 2016. Also, at the location of Čapljina, the total lead content of 85 mg/kg (2015) and 84.6 mg/kg (2016) was determined over two years. In our research, the increased zinc content was found in sandy land at the site of Mostar 111 mg/kg (2015) and 109.7 mg/kg (2016). The average zinc concentrations in soil around the world vary from 60 to 80 mg/kg (Kabata-Pendias, 2011). In a study conducted in eighteen countries around the world, the concentration of total zinc concentrations ranged from 10 to 300 mg/kg with an average value of 70 mg/kg (Barak and Helmke, 1993).

Kabata-Pendias (2011) and Branković *et al.* (2016) determined the total cobalt content in the amount of 89.4 mg/kg on the site being investigated and the value was above the maximum permissible concentrations for all soil types. In our study, total cobalt content ranged from 20.6 ± 40.6 mg/kg. According to Kabata-Pendias (2011), worldwide copper concentrations range from 14 to 109 mg/kg. In our research, the copper concentrations were in the range of 19 ± 57.7 mg/kg. Total nickel content ranged from 23.8 mg/kg to Mostar site up to 44.2 mg/kg at Čapljina location. According to Stančić et al. (2015), nickel values range from 19.2 to 43.3 mg/kg of dry soil. In the soils worldwide, it is estimated that the cadmium content is around 0.41 mg/kg and Kabata-Pendias (2011) and Branković *et al.* (2016) indicate that the cadmium content of 2.5 mg/kg was determined at the investigated sites. At locations in our research, cadmium content ranged from 0.6 ± 1 mg/kg.

Non-cancerous risk of heavy metals for children and adults

The non-cancerous risk factors for children and adults are calculated on the basis of RfD, as shown in Table 2. The calculated values have been presented in Tables 5 and 6, whilst the HQ values have been presented separately for adults (Table 5 and Figure 1) and children (Table 6 and Figure 2).

Table3. Heavy metal content by location (mg/kg)

Location / Heavy metal	Pb	Cd	Co	Cu	Ni	Zn
Stolac	25.4±52.3	0.6±1	27.2±43.6	39.8±57.5	31.4±33.7	90.2±108.3
Mostar	27.6±52.1	0.6±0.8	27.6±29.1	19±44.7	23.8±33.5	48.8±111
Čapljina	31.7±85	0.6±0.9	20.6±42.9	38±57.7	28.7±44.2	88±144.2

Table 4. Average concentrations of heavy metals at all locations (mg/kg)

	Pb	Cd	Co	Cu	Ni	Zn
Min	25.40	0.60	20.60	19.00	23.80	48.80
Max	85.00	1.00	43.60	57.70	44.20	144.20
Average	44.30	0.76	31.42	44.20	34.53	97.03

Table 5. Average daily intake (ADI) in mg/kg/d from the soil for adults

Heavy metal / The way of intake	Pb	Cd	Co	Cu	Ni	Zn
Ingestion	6.09E-05	1.03E-06	4.30E-05	6.06E-05	4.73E-05	1.33E-04
Inhalation	9.34E-09	1.59E-10	6.62E-09	9.32E-09	7.28E-09	2.04E-08
Dermal	1.50E-04	2.56E-06	1.07E-04*	1.50E-04	1.17E-04	3.29E-04
Total	2.11E-04	3.59E-06	1.50E-04	2.11E-04	1.64E-04	4.62E-04

* the value is higher than the reference doses (RfD) in mg/kg day from Table 2

Table 6. Average daily intake (ADI) in mg/kg/d from the soil for children

Heavy metal / The way of intake	Pb	Cd	Co	Cu	Ni	Zn
Ingestion	5.66E-04	9.65E-06	4.02E-04	5.65E-04	4.41E-04	1.24E-03
Inhalation	4.36E-07	7.43E-09	3.09E-07	4.35E-07	3.39E-07	9.54E-07
Dermal	7.26E-05	1.24E-06	5.15E-05*	7.24E-05	5.66E-05	1.59E-04
Total	6.39E-04	1.09E-05	4.53E-04	6.38E-04	4.98E-04	1.40E-03

* the value is higher than the reference doses (RfD) in mg/kg day from Table 2

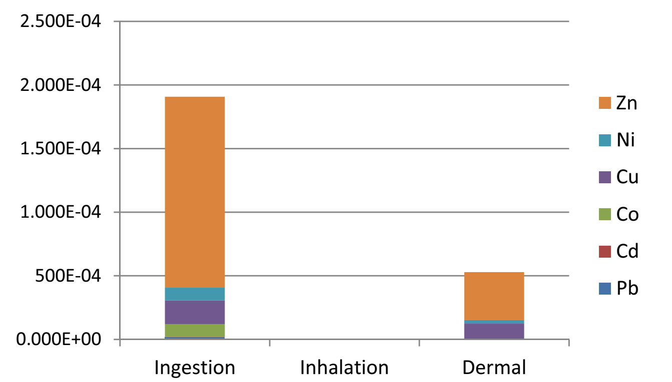

Figure 1. HQ values (adults) for heavy metals in the soil

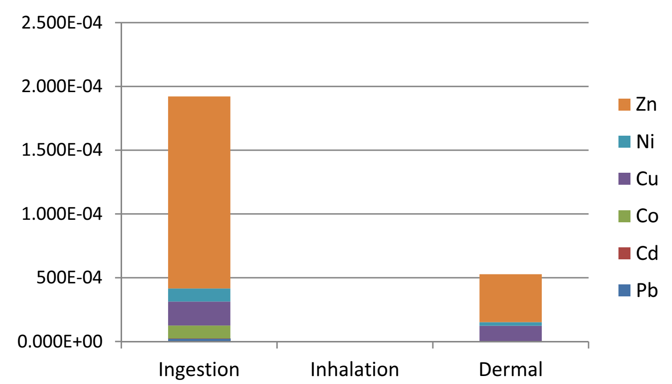

Figure 2. HQ values (children) for heavy metals in the soil

When the value of HQ and HI is less than 1, then there is no risk to human health, but if these values exceed 1, then there is concern about the risk of non-cancerous agents. HQ and HI values for adults and children for oral, dermal and inhalation pathways of heavy metals in organisms were below 1, meaning there is no risk to human health due to oral, inhaled and dermal exposures investigated with heavy metals from the soil, except for Co. The measured HQ values (adults) for all lead input paths are $1.83x10^{-3}$, for cadmium $5.30x10^{-4}$, for copper $3.06x10^{-2}$, for nickel $1.29x10^{-2}$, and for zinc 0.188. The measured HQ values (children) for all lead input paths are $2.08x10^{-3}$, for cadmium $5.34x10^{-4}$, for copper $3.08x10^{-2}$, for nickel $1.30x10^{-2}$, and for zinc 0.188. HI value >1 for Co for dermal intake for adults (1.01) and children (1.02) is meaning there is a potential risk to human health. In Luo *et al.* research (2012), it is expressed concern for the non-cancerous risk of oral lead for children, even though the value of HI is lower than 1. Also, Zheng *et al.* (2009) suggest a potential risk to children from lead inhalation exposure when its value is HI>1. Oluseye Ogunkunle *et al.* (2013) explored the potential non-cancerous risk for adults and children's health due to exposure to heavy metals in Nigeria in 2013. The risk assessment results indicated that the greatest risk to adult's and children's health is mainly related to cadmium and chromium.

Conclusion

Based on the conducted research into the content of heavy metals in soil and the assessment of human health risks from non-cancerous agents, it can be concluded that measured lead values at the location of Mostar in 2015 and 2016 were above the maximum permissible concentrations. At the location of Čapljina (*Caldesi 2000* site), the lead content in the soil was determined above the maximum permissible concentrations in both investigated years. At the site of Čapljina (*Big Top*), it was found that the content of all the heavy metals studied in the soil was in line with the limit values. The measured value of HI dermal intake Co for adults is 1.01. The measured value of HI dermal intake Co for children is 1.02. Based on the calculated values of HI> 1, there is a potential non-cancerous risk for adults' and children's health by dermal injection of cobalt. The obtained results point to the need for monitoring of agricultural land in this area as well as in the whole Bosnia and Herzegovina in order to reduce the potential risk of non-cancerous agents and the need to reduce mineral fertilizers as potentially pollutants to agricultural soil.

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