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Human health risk assessment of lead in nectarines

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Abstract: The aim of the paper was to determine the level of soil contamination with lead and perform risk assessment by calculating the *Hazard Quotient Index (HQI).* The research was carried out during 2015 and 2016 at three locations in Herzegovina (Mostar, Čapljina, and Stolac), where nectarine cultivars ('Big Top' and 'Caldesi 2000') were cultivated. At the Mostar and Čapljina locations, the total content of lead in the soil exceeded MPC values in both years of research. Estimated daily intake (EDI) and estimated weekly intake (EWI) values of lead in the studied nectarine varieties showed no acute or chronic risk for human health.

Keywords: lead, soil contamination, nectarines, risk assessment.

Introduction

In recent years, increasing attention has been paid to the contamination of soil, water and air and the consequential entry of contaminants into the food

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chain. Contamination of agricultural soil is caused by the introduction of harmful substances via water and air or their accumulation on or in the soil at concentrations above the permitted levels. Hazardous matter in agricultural soil may be present at concentrations that may temporarily or permanently call into question its fundamental role as a favorable habitat for plants. Hazardous substances include: heavy metals (cadmium, mercury, nickel, cobalt, lead, chromium, zinc, copper, arsenic), polycyclic aromatic hydrocarbons and other harmful substances that are introduced into the soil, which, due to their improper or excessive and untimely application, can lead to a number of adverse environmental impacts. For individual elements, the range between the amount that is optimal for the organism and the amount that is harmful is very narrow. Elements such as lead, cadmium, mercury, arsenic, and molybdenum exhibit toxicity in large quantities and are classified as classical inorganic contaminants (Mačkić and Ahmetović 2012). Exposure to metals can result in negative health effects, and it is for these reasons that European and international food safety agencies have specified 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: hazard identification, hazard characterization, exposure estimation and risk characterization. The risk assessment method for chemical contaminants is defined by WHO (*World Health Organization*), US EPA (*Environmental Protection Agency*) and EFSA (*European Food Safety Authority*) (Dorne *et al.,* 2011).

Materials and Methods

Studies were carried out on already established nectarine plantations in Čapljina, Stolac, and Mostar, at three microlocations differing in pedological, climatic and exposure terms. Analyses of total lead content in the soil and lead concentrations in nectarine seed samples were performed in laboratories of the Federal Institute of Agriculture and Agropedology Institute in Sarajevo. Standard modern equipment was used for the analyses. Soil sampling was carried out at three locations in Mostar, Stolac, and Čapljina at two depths of 0-30 cm and 30-60 cm. In Mostar and Stolac, two nectarine varieties, 'Caldesi 2000' and 'Big Top', were grown on the same plot, whereas at the location in Čapljina, they were planted on different plots which were about 1000 m apart. Laboratory surveys included physical and chemical analyses of 32 soil samples, determination of total lead content (Pb) and chemical analyses of 18 nectarine fruit samples for lead content (Pb) (by pressure digestion and inductively coupled plasma mass spectrometry (ICP-MS)). Risk assessment included: estimation of daily and weekly consumption of nectarines, estimation of daily intake of lead by nectarine consumption, estimation of weekly lead intake by nectarine consumption and the *Hazard Quotient Index (HQI).*

Determination of heavy metal content in soil

Samples for instrumental analyses of the content of heavy metals in the soil were prepared by means of aqua regia, and then their content in the extract was determined by atomic absorption spectrometry (AAS). The extraction of the heavy metals in gold foil was carried out according to the international standard ISO11464. The Bylaw Act currently in force in the Federation of Bosnia and Herzegovina which is directly related to the research topic is the Guidance on Determination of Permissible Quantities of Harmful and Hazardous Substances in the Soil and Methods of Their Examination (Official Gazette FBiH, No. 72/09). Limit values of soil pollutants in their total form are set only for agricultural soils, whereas limits for other soils have not been legally established yet.

Table 1. Limit values of heavy metals in the soil in total form

Element	Limit values (mg/kg)		
	Sandy soil	Sandy loam soil	Hard ground
Cadmium (Cd)	0.5	1	1.5
Copper (Cu)	50	65	80
Lead (Pb)	50	80	100
Zinc (Zn)	100	150	200
Cobalt (Co)	30	45	60
Nickel (Ni)	40	40	50
Iron (Fe)*	3.2%	3.5%	5%

Source: Instruction on Determination of Permitted Amounts of Harmful and Hazardous Substances in Soil and methods of their examination (Official Gazette FBiH, No. 72/09) * (content of iron by H.Resulović and H. Čustović (2002))

Determination of the content of heavy metals in nectarine fruit

The lead analyte was assessed by the method for determining trace elements (lead, cadmium, and arsenic) in fruits and vegetables using the ICP - MS method (BAS EN 15763: 2011) after microwave digestion (BAS EN 13805: 2005). The content of heavy metals in nectarine fruit in relation to MPC has been defined according to the Ordinance on Maximum Permitted Quantities for Certain Contaminants in Food (Official Gazette BiH No. 68/14), according to which the MPC value for lead is 10 mg/kg.

Estimation of daily and weekly consumption of nectarines

In assessing daily and weekly nectarine consumption, the EFSA *Comprehensive European Food Consumption Database* (EFSA 2011) was used. The value for the average daily consumption of nectarines (Category: Peaches –

Prunus persica) for adult consumers was used: 166.48 g daily for acute intake or 73.35 g daily (513.45 g weekly) for chronic intake.

Estimation of daily and weekly heavy metal intake through nectarine consumption

For the calculation of average daily and weekly intake, the formulas recommended by the US EPA were used: EDI (µg / kg body weight per day) = C x Ud / BW and EWI (µg / kg body weight per week) = C x Uw / BW, where: C = mean lead concentrations in test specimens (µg / kg) = average daily intake of nectarines (g / day), U_d = average daily intake of nectarines (g/daily), Uw = average weekly intake of nectarines (g / weekly), BW = average body weight (kg).

Estimated HQI (Hazard Quotient Index)

The HQI and risk level were calculated by comparing the established values of EDI and EWI with health-based toxicological values for lead in food –TDI and TWI. The TWI value of 25 µg / kg BW recommended by EFSA was used (EFSA 2010). Values for TDI were taken as equivalent TWI, namely: 3.57 µg / kg BW.

Results and discussion

Based on the mechanical components of the soil, soil texture analysis was performed according to the USDA classification. The soil was classified as sandy loam at Stolac and Čapljina locations (‘Caldesi 2000’ plantation), sandy at the Mostar site, and clay soil at Čapljina (‘Big Top’). In 2015 and 2016, the average lead concentrations ranged from 25.40 to 85 mg / kg. Lead values above MPC were measured in Mostar (sandy site), at depths up to 30 cm, as well as in Čapljina (sand dune), at 30-60 cm depth. All values are shown in Table 2.

Table 2. Content of lead in the soil

Location/depth (cm)	Type	Soil texture	Pb (mg/kg)	
			2015.	2016.
Stolac / 0-30	*BT and C 2000	Sandy loam	25.4	26.9
Stolac / 30-60	BT and C 2000	Sandy loam	52.3	50.5
Mostar / 0-30	BT and C 2000	Sandy soil	51.8	52.1
Mostar / 30-60	BT and C 2000	Sandy soil	27.6	28.7
Čapljina / 0-30	BT	Clay loam	45.5	46.1
Čapljina / 30-60	BT	Clay loam	34.0	34.5
Čapljina / 0-30	C 2000	Sandy loam	31.7	32.1
Čapljina / 30-60	C 2000	Sandy loam	85	84.6

*BT – nectarine variety ‘Big Top’;*C 2000 – nectarine variety ‘Caldesi 2000’

Based on pH measurement results, the soil at the explored sites in 2015 and 2016 were classified as neutral to very alkaline. Table 3. shows values of soil acidity across years.

Table 3. Soil acidity

Location/depth (cm)	2015		Classification	2016		Classification
	pH H_2O	pH KCl		pH H_2O	pH KCl	
Stolac / 0-30	6.32	5.38	PA-N	6.94	5.8	N
Stolac / 30-60	6.32	4.36	PA-N	6.94	5.63	N
Mostar/ 0-30	7.97	7.39	MA	7.79	7.49	MA
Mostar/ 30-60	7.94	7.34	MA	7.96	7.54	MA
Čapljina / 0-30 BT	8.23	7.88	VA	8.41	8.16	VA
Čapljina / 30-60 BT	6.86	7.92	N-B	5.64	8.12	MA-B
Čapljina / 0-30 C 2000	8.11	7.95	VA	8.43	8.12	VA
Čapljina / 30-60 C 2000	8.64	7.94	VA	8.27	8.16	VA

N = neutral; PA-N = poorly acidic to neutral; MA = moderately alkaline;
VA = very alkaline; N-B = neutral to base; MA-B = moderately acidic to base

Table 4. shows lead content in 2015 in both nectarine varieties. The content of lead in 'Big Top' and 'Caldesi 2000' in 2016 at all sites was lower than the quantification limit.

Table 4. Content of Lead in 'Big Top' and 'Caldesi 2000' in 2015

Location	Lead 'Big Top'	Lead 'Caldesi 2000'
Stolac	<LOQ <LOQ <LOQ	<LOQ <LOQ <LOQ
Mostar	0.026 0.058 0.048	<LOQ <LOQ <LOQ
Čapljina	0.033 0.030 0.020	<LOQ 0.230 <LOQ
Reference values	0.10	0.10
Limit of quantification (LOQ)	0.020	0.020

Table 5. shows EDI and HQI for lead in ‘Big Top’ samples in 2015 and 2016. The average value of lead intake through the consumption of this nectarine variety ranged between 0.024 and 0.105 µg/kg body weight per day. The HQI values ranged between 0.67 and 2.94, indicating no risk to adult health from the daily intake of lead through this nectarine variety.

Table 5. HQI values (for daily intake) for Pb (‘Big Top’) in 2015 and 2016

Year	Location	mean Pb concentration (µg / kg)	EDI (µg/kg BW daily)	HQI (%)
2015	Čapljina	0.028	0.067	1.88
	Mostar	0.044	0.105	2.94
	Stolac	0.01	0.024	0.67
2016	Čapljina	0.01	0.024	0.67
	Mostar	0.01	0.024	0.67
	Stolac	0.01	0.024	0.67

Table 6. shows EWI and HQI values for lead in ‘Big Top’samples in 2015 and 2016. The average value of lead intake through this nectarine variety ranged between 0.073 and 0.323 µg / kg body weight per week. HQI values ranged from 0.29 to 1.29. This suggested that there was no risk to adult health by the weekly intake of lead through this nectarine variety.

Table 6. HQI values (for weekly entry) for Pb (‘Big Top’) in 2015 and 2016

Year	Location	mean Pb concentration (µg / kg)	EWI (µg/kg BW weekly)	HQI (%)
2015	Čapljina	0.028	0.205	0.82
	Mostar	0.044	0.323	1.29
	Stolac	0.01	0.073	0.29
2016	Čapljina	0.01	0.073	0.29
	Mostar	0.01	0.073	0.29
	Stolac	0.01	0.073	0.29

Table 7. shows EDI and HQI for lead in ‘Caldesi 2000’ in 2015 and 2016. The average value of lead intake through this nectarine ranged between 0.024 and 0.197 µg / kg body weight per day. The HQI values ranged between 0.67 and 5.52, which indicated no risk to adult health by the daily intake of lead through this nectarine variety.

Table 7. HQI values (for daily intake) for Pb ('Caldesi 2000') in 2015 and 2016

Year	Location	mean Pb concentration (µg / kg)	EDI (µg/kg BW daily)	HQI (%)
2015	Čapljina	0.083	0.197	5.52
	Mostar	0.01	0.024	0.67
	Stolac	0.01	0.024	0.67
2016	Čapljina	0.01	0.024	0.67
	Mostar	0.01	0.024	0.67
	Stolac	0.01	0.024	0.67

Table 8. shows EWI and HQI values for lead in 'Caldesi 2000' in 2015 and 2016. The average value of lead intake through this nectarine variety ranged between 0.073and 0.61 µg / kg body weight per week. The HQI values ranged from 0.29 to 2.44, indicting no risk to adult health by the weekly intake of lead through this nectarine variety.

Table 8. HQI values (for weekly intake) for Pb ('Caldesi 2000') in 2015 and 2016

Year	Location	mean Pb concentration (µg / kg)	EWI (µg/kg BW weekly)	HQI (%)
2015	Čapljina	0.083	0.61	2.44
	Mostar	0.01	0.073	0.29
	Stolac	0.01	0.073	0.29
2016	Čapljina	0.01	0.073	0.29
	Mostar	0.01	0.073	0.29
	Stolac	0.01	0.073	0.29

Elevated amounts of lead in agricultural soils should be expected near lead-processing industries as well as in soils where sewage sludge is used. On those lands lead content may range from 50 to 2600 ppm, depending on its origin (Tiller 1992, Pourrut *et al.,* 2011, Feleafel and Mirdad 2013).

In the research by Bosilj *et al.* (2015), six locations in Varaždin, Republic of Croatia, were found to have the total lead content above the maximum permissible concentration, even greater than expected. The presented results are in accordance with this study since the lead concentrations determined at the locations of Mostar and Čapljina exceeded MPC values for such types of soils.

The acidity of the soil (pH) and organic matter are significant factors governing the solubility of heavy metals in the soil. These factors are also important in the soil-plant relationship. In acidic soils, heavy metals are more soluble, more mobile and readily available to the plant as the bioavailable fraction is higher than in carbonate (alkali) soils.

According to the literature data (Jakšić *et al*., 2013), in soil samples of investigated sites, an increase in lead content ranged from 186.80 to 203.20 mg/kg, thus exceeding the maximum allowable amount. This research is consistent with our research as the lead values at 0-30 cm soil depth measured in 2015 and 2016 at the Mostar sandy soil site were above the maximum permissible concentrations. The amounts were 51.8 mg/kg in 2015 and 52.1 mg/kg in 2016, which according to (Tiller 1992, Pourrut *et al.,* 2011, Felearel and Mirdad 2013) can be explained by the fact that the crop is near the main road. Also, at the location of Čapljina during the two years, the total lead content of 85 mg/kg (2015) and 84.6 mg/kg (2016) was determined. At this site ('Caldesi 2000'), where sandy peat soil was determined, the total lead content at depths of 30-60 cm in both years was above the MPC value. The values were 85 mg/kg in 2015 and 84.6 mg/kg. This can be explained by the distribution of lead in the soil, which is positively correlated with its particle size distribution, suggesting that the percentage of lead increases with increasing percentage of clay. Research in the world has shown that lead accumulates directly beneath the surface of the soil as well as that it shows the lowest mobility of all toxic metals (Kabata-Pendias *et al.,* 2011).

Dimitrijevic *et al.* (2016) reported lead concentrations in nectarine and peach of 0.683 mg/kg. The average metal content in nectarine samples in a study in Romania was 0.13 mg/kg (lead) (Gogoaşă *et al.,* 2005). In a study on heavy metal content in food and its risk assessment to human health in India (Chandorkar and Prachi, 2013), the average daily intake of Pb was 0.69 mg/kg.

Conclusion

Based on the research on the content of lead in the soil and yields of nectarine varieties 'Big Top' and 'Caldesi 2000' at the selected locations in Stolac, Mostar and Čapljina accompanied with a human health risk assessment, the following conclusions can be drawn:

At the site of Mostar and Čapljina, the total lead content in the soil exceeded the maximum permissible concentrations in both research years. The estimation of the average daily intake and the average weekly intake of lead in 'Big Top' and 'Caldesi 2000' nectarine fruits showed that the recommended tolerance values of daily and weekly intake of lead were not exceeded. These results indicate that there was no acute or chronic risk to adult health. The results of the research indicate the need for the implementation of the Total Dietary Study in Bosnia and Herzegovina to determine dietary habits (such as type and quantity of food, frequency of consumption) in BiH as well as the need for the introduction of risk assessment into human health methodology.

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