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HEALTH RISK ASSESSMENT IN AGRICULTURAL SOILS IN A CITY OF PLJEVLJA (MONTENEGRO)

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ABSTRACT

Population exposure due to industrially contaminated sites (ICSs) is a global environmental health problem. Pljevlja municipality is one of the “black spots” in Montenegro and well known by pollution influence from several different sources. Agricultural soils in a city of Pljavlja are at the great ecological risk due to the impact of industrial pollutants, such as lead and zinc mine (LZM), coal fired power station (CFPS) and coal mine (CM).

This study aimed to present a children health risk assessment of agricultural soil in a vicinity of these three pollution sources for As, Hg, Pb, Cd, Cu, Zn and Cr. Both, non-carcinogenic (for As, Hg, Pb, Cd, Cu, Zn and Cr) and carcinogenic health risk (for As, Cd, Pb and Cr) due to the human exposure to the soil through different exposure pathways were evaluated.

The results of non-carcinogenic health risk assessment showed that Cr, Pb, Cd, and As were of concern for children population. Dermal contact was found as the main exposure pathway from LZM soil samples followed by ingestion and inhalation. On the other hand, for CFPS and CM soils, ingestion was the main exposures pathway. The carcinogenic health risk was highly controlled by Cr and As and ingestion is recognized as the main children exposure route.

Keywords: pollutants, health risk assessment, carcinogenic health risk, non-carcinogenic health risk.

INTRODUCTION

Contamination of agricultural soil by heavy metals has attracted a special attention, because metals can be taken by plants and may enter the food chain, and therefore, humans can also be exposed to this kind of contamination (Huang et al., 2007). Some heavy metals such as Co, Cu, Fe, Mn, Ni and Zn are essential for the proper function of human body, but when they exceed certain concentrations they become extremely toxic since they are non-biodegradable and tend to accumulate in human body leading to serious health problems (Zoroddu et al., 2019). Contrary Pb, Cd, Hg and As, do not have any beneficial effect on organisms and are therefore very harmful for human health even in small concentration (Rahman, & Singh, 2019).

Since different industrial activities inevitably lead to soil pollution in order to protect human health, it is necessary to comprehensively assess the health risks posed by heavy metals in the environment. Direct exposure of humans to polluted soil may present the health risk since toxic heavy metals may be easily transferred from soil to human body via ingestion, inhalation or dermal contact (Hua et al., 2020).

Thus the aim of this paper was to access non-carcinogenic and carcinogenic children health risk in Pljevlja municipality (Montenegro). Pljevlja is the industrially and agriculturally important town in north Montenegro, located in a valley surrounded by hills and mountains up to 2000 meters altitude. Industrial activity (mine of lead and zinc (LZM), coal fire power station (CFPS) and mine of coal (CM)) in this town present a serious risk for human health since they cause a continual environmental pollution by heavy metals.

MATERIALS END METHODS

Soil samples were taken from maximum 20 cm depth, from agricultural fields near households in November 2019 in Pljevlja municipality. The households are located near three sources of pollution, Mine of lead and zinc (MLZ), Coal fired power station (CFPS) and Coal mine (CM). For the purpose of determination of heavy metal content in soil samples, wet soil was dried at 105°C and sieved through mesh 0.25µm. 3g of dried and sieved samples were digested with 25ml of aqua regia, using reflux and heating source (EPA method 3050B). Samples were filtrated through Whatman No. 41 and flasks were washed with 5ml of concentrated HCl and adjusted to final volume of 50ml with distilled water. Concentrations of heavy metals were determined using inductively coupled plasma technique (ICP-OES), manufactured by Spectro Arcos with hydride technique for As determination.

ASSESSMENT OF HEALTH RISK

Humans could be exposed to contaminants from soil via oral and inhalation intake and through dermal exposure. Non-carcinogenic and carcinogenic risk of children through each of the three exposure pathways to soil were calculated using the methodology proposed by US Environmental Protection Agency (US Environmental Protection Agency [USEPA], 1989).

The average daily doses (*ADDs*) (mg/kg day) of potentially toxic metals via ingestion (ADD_{ing}), inhalation (ADD_{inh}) and dermal contact (ADD_{derm}) for both children were estimated using following Eqs:

$$ADD_{ingetion} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

$$ADD_{inhalation} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad (2)$$

$$ADD_{dermal} = \frac{C \times SA \times SAF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (3)$$

where C is concentration of the contaminant in soil; IngR is ingestion rate of soil; EF is exposure frequency; ED is exposure duration; BW is average body weight; AT is averaging time; InhR is inhalation rate; PEF is particle emission factor; SA is surface area of the skin that contacts the soil; SAF is skin adherence factor for soil; ABS is dermal absorption factor (chemical specific). Factors used in the risk assessment equations are given in Table 1.

Non-carcinogenic effects of each heavy metal were assessed using the hazard quotient (HQ) and hazard index (HI) according Eq. 4 and 5 while carcinogenic effect was assessed using the carcinogenic risk (CR) and total carcinogenic risk (TCR) by Eq. 6 and 7.

$$HQ_i = \frac{ADD_i}{RfD_i} \quad (4)$$

$$HI = \sum HQ_i \quad (5)$$

$$CR_i = ADD_i \cdot SF \tag{6}$$

$$TCR = \sum CR_i \tag{7}$$

were RfD is reference dose which presents the maximum daily dose of each individual metal from a specific exposure pathway, for adults and children, that is believed not to lead to an appreciable risk of deleterious effects to sensitive individuals during a lifetime; CR_i is cancer risk of each individual heavy metal, *SF* is slope factor for carcinogenic exposure and *TCR* is total cancer risk. Reference values of some parameters for health risk assessment of heavy metals in soils are given in Table 2.

For HI ≤1, it is believed that there is no significant risk of non-carcinogenic effects while for HI > 1, it means that there is a great chance of non-carcinogenic effects. According USEPA regulatory the tolerable cancer risk is in the range $1{\cdot}10^{-6} - 1{\cdot}10^{-4}$ (USEPA, 2015).

Table 1. Factors used in the risk assessment equations.

Factor	Value	
	Children	Adults
IngR; (mg/day)	200	100
InhR; (m^3/day)	7.6	20
EF; (days/year)	350	350
ED; (years)	6	24
BW; (kg)	15	70
AT; (days)	365·ED	365·ED
PEF; (m^3/kg)	$1.36{\cdot}1^9$	$1.36{\cdot}10^9$
SA; (cm^2)	2800	5700
SAF; (mg/cm^2 day^1)	0.2	0.7
ABS (for all elements except for As)	0.001	0.001
ABS (for As)	0.03	0.03

Table 2. RfDs values ($mg{\cdot}kg^{-1}{\cdot}day^{-1}$) and slope factor ($mg{\cdot}\ kg^{-1}{\cdot}day^{-1}$).

Metal	RfD_{ing}	RfD_{inh}	RfD_{der}
As	3.00E-04	3.00E-04	1.23E-04
Hg	3.00E-03	8.57E-05	2.10E-05
Pb	3.50E-03	3.52E-03	5.25E-04
Cd	1.00E-03	1.00E-03	1.00E-05
Cd	4.00E-02	4.20E-02	1.20E-02
Cu	3.00E-01	3.00E-01	6.00E-02
Zn	3.00E-03	2.86E-05	6.00E-05
Cr	3.00E-04	3.00E-04	1.23E-04
Metal	SF_{ing}	SF_{inh}	SF_{der}
$As_{(cancer)}$	1.50E+00	1.51E+01	1.50
$Cd_{(cancer)}$	3.80E-01	6.30E+00	3.80E-01
$Cr_{(cancer)}$	5.01E-01	4.20E+01	20.00

RESULTS AND DISCUSSION

Concentration of heavy metals in agricultural soil

The average concentration of heavy metals in agricultural soil samples collected in LZM, CFPS and Cm zones in Pljevlja municipality is given in Fig.1. As can be seen Zn is the metal with the highest concentration in all soil samples. The highest concentrations of heavy metals were observed in the soil collected near the LZM and area ranked in descending order as Zn > Pb> Cu> Cr> As> Cd> Hg. Furthermore, the average concentration of heavy metals in CFPS and CM sample groups are ranked in a descending order, as Zn > Cu > Pb> Cr> As> Cd> Hg for both sampling areas.

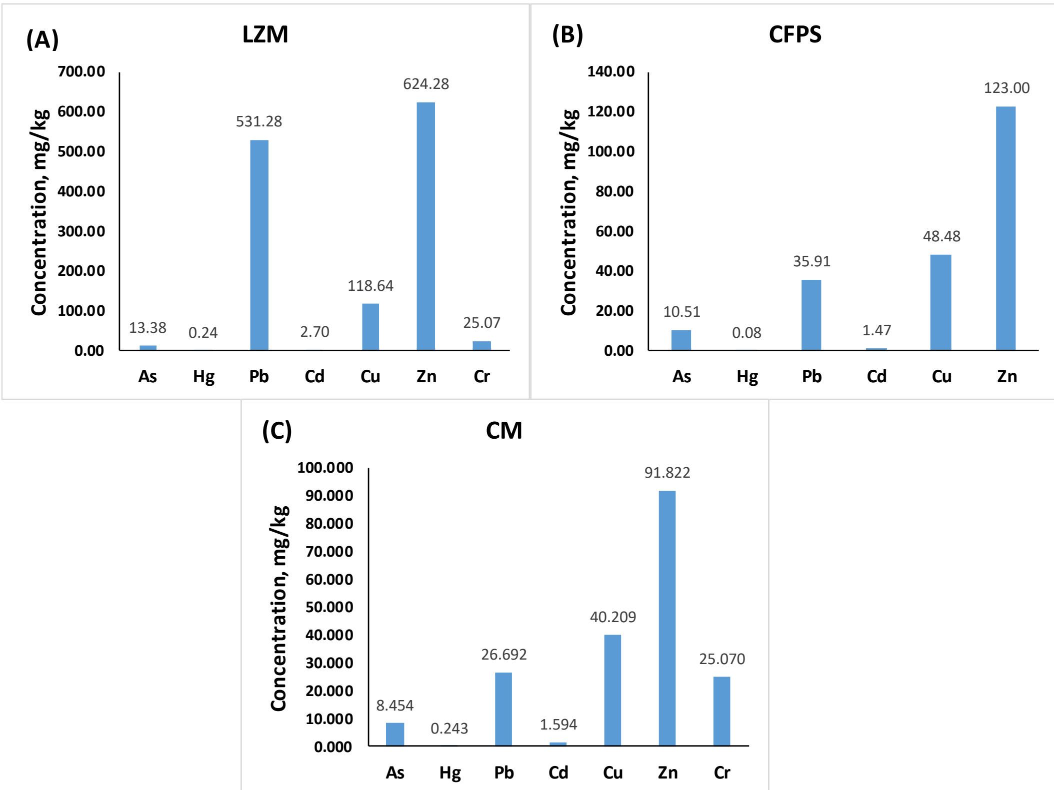

Figure 1. Average heavy metal concentration in agricultural soil in LZM, CFPS and CM zones.

NON-CARCINOGENIC AND CANCEROGENIC HEALTH RISK ASSESSMENT

The non-carcinogenic and carcinogenic health risks caused by the exposure of soil from industrial pollutants for children (aged bellow 18 year) via ingestion, inhalation and dermal contact were given in Fig. 2 (A, C, E). The highest non-carcinogenic health risk for children was observed in LZM study area followed by HI values for CFPS and CM areas.

The values of non-carcinogenic hazard indexes for Hg, Cu and Zn (Fig. 2. (A, C, E)) were all lower than one, in all study areas indicating that there no non-carcinogenic risk for adults and children through soil exposure. On the other hand, hazard indexes for Cr, Pb, Cd, and As were of concern since HI values for these metals were observed to be higher than one.

Among all investigated metals, Cr is the largest contributor to the health risk since the highest HI values were observed for this metal in all investigated areas. As shown in Fig. 2A, the hazard indexes for adults and children in LZM study area, exceeded one for Cr, Pb, Cd and As. As for

CFPS and CM study areas (Fig 2C and 2E), the HI values higher than one were observed for Cr, As and Cd.

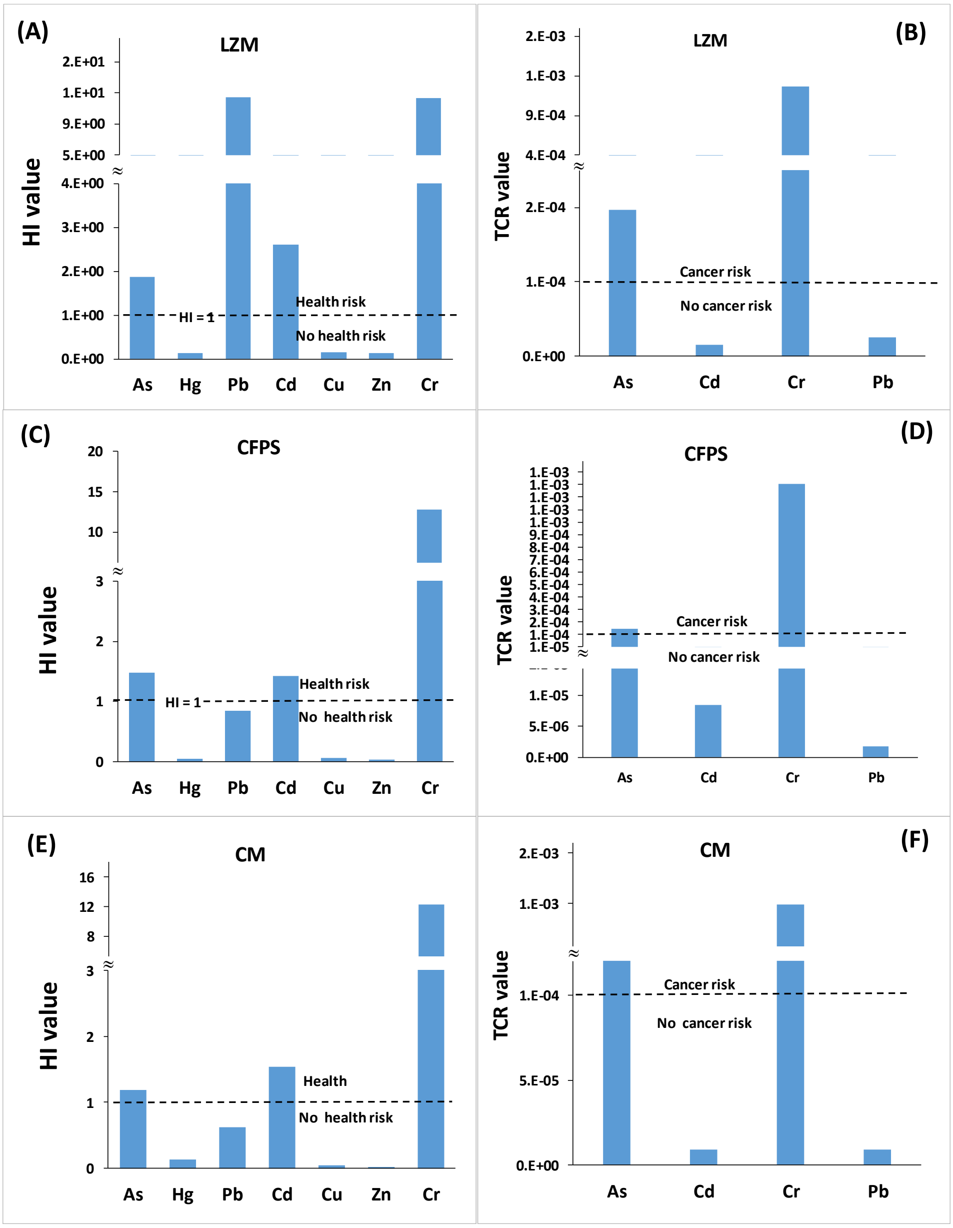

Figure 2. Contribution of different metals to non-carcinogenic hazard index (A, C, E) and to total carcinogenic risk (B, D, F) for adults and children for different soil samples.

Among the three different pathways to soil exposure for adults in LZM area (Fig. 3A), dermal contact was recognized as the main health risk followed by ingestion and inhalation. On the other hand, in CFPS and CM areas (Figs. 3B and 3C), ingestion was the main health risk followed by dermal contact and exposure by inhalation.

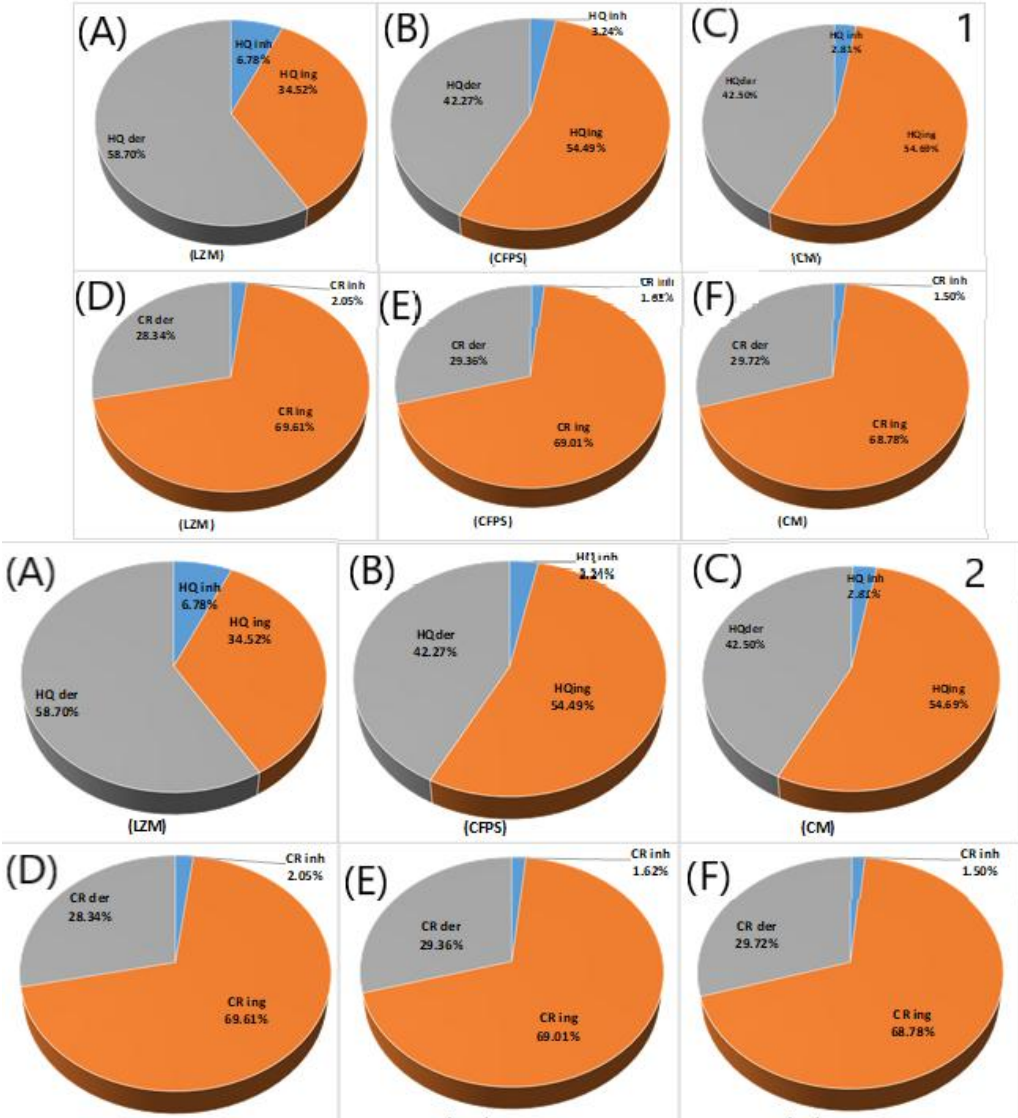

Figure 3. Contribution of exposure pathways to non-carcinogenic health rick for adults 1 (A, B, C) and for children 2(A, B, C) and to cancerogenic health risk for adults 1(D, E, F) and children 2(D, F) for different study areas.

The assessment of results of carcinogenic risk analysis are given in Fig 2 (B, D, F). It is evident that for Cd and Pb, TCR values were within acceptable limits (between $1{\cdot}10^{-6}$ and $1{\cdot}10^{-4}$) indicating an acceptable risk to human health. In all investigated areas, TCR values for Cr and As have exceeded the value of $1{\cdot}10^{-4}$ indicating the carcinogenic risk.

Further, from the results presented in Fig. 3, it can be seen that health risk posed by carcinogenic elements (1D, E, F) via ingestion is the highest, followed by dermal contact and inhalation pathway, in all investigated areas for adults as well as for children (2D, E, F).

CONCLUSIONS

The results obtained in this study have shown that children in LZM area in Pljevlja municipality are faced with a highest non-carcinogenic and carcinogenic health risk with respect to CFPS and CM zones. The children in Pljevlja municipality are not faced with carcinogenic health risk through the soil exposure with respect to Cu, Zn and Hg content in soil, while the presence of Cr, As, Cd and Pb present potential non-carcinogenic health risk.

The human health risk assessment revealed that dermal contact is the main exposure pathway in LZM zones, while in CFPS and CM zones, dermal contact is recognized as the main exposure pathway.

The total carcinogenic risk values for children in investigated locations were higher than the acceptable level ($1 \cdot 10^{-4}$) for Cr and As, thus the chance of having cancer through long-term exposure could be inevitable. On the other hand, there is no cancer risk with respect to Cd and Pb content in agricultural soil.

LITERATURE

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