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A STUDY ON HEALTH RISK ASSESSMENT OF HEAVY METALS THROUGH INTAKE OF AGRICULTURAL FOOD CROPS IN DISTRICT MARDAN, PAKISTAN

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ABSTRACT

This study investigated the concentrations of different selected heavy metals like Cadmium (Cd), Chromium (Cr), Copper (Cu), Manganese (Mn), Nickel (Ni), Lead (Pb) and Zinc (Zn) in farming products (grains and vegetables), and their probable health impacts in district Mardan, Khyber Pakhtunkhwa (KPK), Pakistan. Locally grown food crops containing grains (wheat, maize, rice) and vegetables (lady finger, turnip, brinjil, tomato, bitter gourd, eddoe, potato) and human blood samples were collected haphazardly from the five selected locations named Khazana dheri, Par Hoti, Gujjar Ghari, Katlang and Takht Bhai of the area.

The samples were checked for the selected heavy metals on an Atomic Absorption Spectrometer (AAS). The findings were also matched with their acceptable parameters issued by the World Health Organization (WHO), Food and Agricultural Organisation (FAO), Pakistan Environmental Protection Agency (Pak-EPA) and State Environmental Protection Administration (SEPA). The concentrations have been detected in a descending sequence of Mn > Cu > Ni > Zn >Pb > Cr > Cd and Zn > Cr > Ni > Mn > Cu > Pb > Cd in grains and vegetables respectively. Moreover, Cd was observed beyond its safe limits in 90 % of all samples. Also, the Pb noted above its safe limits by FAO, 2001 but in limits set by SEPA, 2005. Moreover, the Chronic Daily Intake (CDI) of heavy metals by grains (3.48E-03) was enough higher than that of vegetables (1.84E-03). Additionally, the children were found more vulnerable to metal toxicity as they were recorded with a higher CDI of 2.80E-03 as compared to that of adults' CDI of 1.86E-03.

Also, the potential threats to the residents (adults and children) were noted as Zn > Cr > Mn > Cu > Ni > Pb > Cd and Zn > Cr > Cu > Ni > Mn > Pb > Cd respectively. In the end, the accumulative Hazard Index (HI) via the ingestion of selected food items was also measured. The combined Health Risk Indexes (HRIs) of each metal were obtained as Cd > Pb > Ni > Cu > Mn > Zn > Cr. With HI of grains (29.5%) and vegetables (28.6%). The cumulative HI value was valued as 4.46E+00 which is far greater than 1 indicating that although the individual HRI values were < 1 but the combined influence of all the selected heavy metals may probably induce serious health risks in people of the area.

1. Introduction

Heavy metals like lead (Pb), copper (Cu), chromium (Cr), zinc (Zn), cadmium (Cd), nickel (Ni) and manganese (Mn), are some of the key environmental pollutants and their buildup in topsoil is a great worry due to noxious effects on crop growth, food value and on human health (Rehman et al. 2017). They usually come from natural actions like the breakdown of parent rocks, or from man-made activities e.g. industrial processes, mining, wastewater irrigation, inorganic fertilizers, pesticides etc. Industries mainly impart in increasing heavy metals in the ecosystem (Mohammadi et al., 2019a). Which contaminate soil, water, and crops with heavy metals (HMs) and affect humans (Roychowdhury et al., 2003; Bundschuh et al., 2012; Khan et al., 2008; Shah et al., 2010; Sekomo et al., (2011). Their non-point sources include agro-chemicals and sewage slurry(Islam et al., 2014a; Cui et al., 2005; Zheng et al., 2007). Heavy metals are thought a principal risk to soil value (Rezapour et al., 2019). Contamination of heavy metals in soils and food harvests is a serious environmental matter due to its non-decomposable nature (Khan et al., 2010; Muhammad et al., 2011b; Radwan and Salama., 2006). Agrarian harvests particularly vegetables, grains and fruits are the main part of human food and act as buffering agents in digestion (Yang et al., 2011; Jan et al., 2010b). Their extreme ingestion via tainted food crops is associated with health hazards in humans (Turkdogan et al., 2003; Khan et al., 2008).

Studies have shown that even in minute amounts, such metals are still detrimental to organisms and may lead to many issues in human beings due to their persistent nature (Salvatore et al., 2008; Lenntech et al., 2016). Heavy metals are very noxious when accumulate in the body and may initiate strong biochemical alterations (Jan et al., 2011). Heavy metals are also damaging due to their solvable nature in water. Even a minor concentration of such metals has damaging outputs because of lacking mechanisms for their removal from the body (Krishna et al., 2009; Arora et al., 2008). Even Zn, Fe, Cu are critical for humans, but chronic troubles may arise from their excess or deficiency (Jan et al., 2011). Expiries from tumors in various countries are thought to be directly connected with the oral intake of metals like Cd, Zn, and Pb (Pasha et al., 2010).

Further, the entrance of Ni in the body can produce cardiac attacks, rashes, headaches and instability of the body (Muhammad et al., 2011). Enough quantity of Zn is extremely significant for different functions and its deficiency may result in diarrhea, depression and immune dysfunction but its upsurge can cause anemia (Muhammad et al., 2011b). Access to all such metals may collectively affect the overall growth and development of the people and eventually all the society indeed. For all these reasons, it is needed to evaluate the interaction of human beings with all these toxic metals via foodstuff ingestion because of the human’s most important position and as an uppermost utilizer in the ecosystem.

2. Materials and methods

2.1 Research area

This study was carried out in Mardan which is an administrative district of the province of Khyber Pakhtunkhwa (KPK) in Pakistan. Mardan is a part of the Peshawar basin located at the southern foothills of the lower Himalayas mountainous range of Khyber Pakhtunkhwa in Pakistan (Shah and Tariq., 2007). It lies between the latitudes 34^o 05′ to 34^o 32′ North and longitude 71^o 48′ to 72^o 25′ East (**Fig 2.1**). There are many minerals variety in these rocks comprised of constructive to ornamental nature. Its North is surrounded by the lower mountainous of Malakand Division which is a gateway to the world-famous Swat Valley.). The maximum number of rivers and tributaries flow down into the river Kabul at Nowshera. The overall climate of the area is tropical warm. The land is mostly agricultural**.** The district is well-reputed for its horticultural products like cash crops (tobacco, sugar cane, sugar beet, etc) and food crops like wheat, maize, rice, and many vegetables like brinjil, tomato, lady finger, potato, bitter gourd, turnip, cabbage, cauliflower, etc. The district occupies a total area of 1632 Km^2

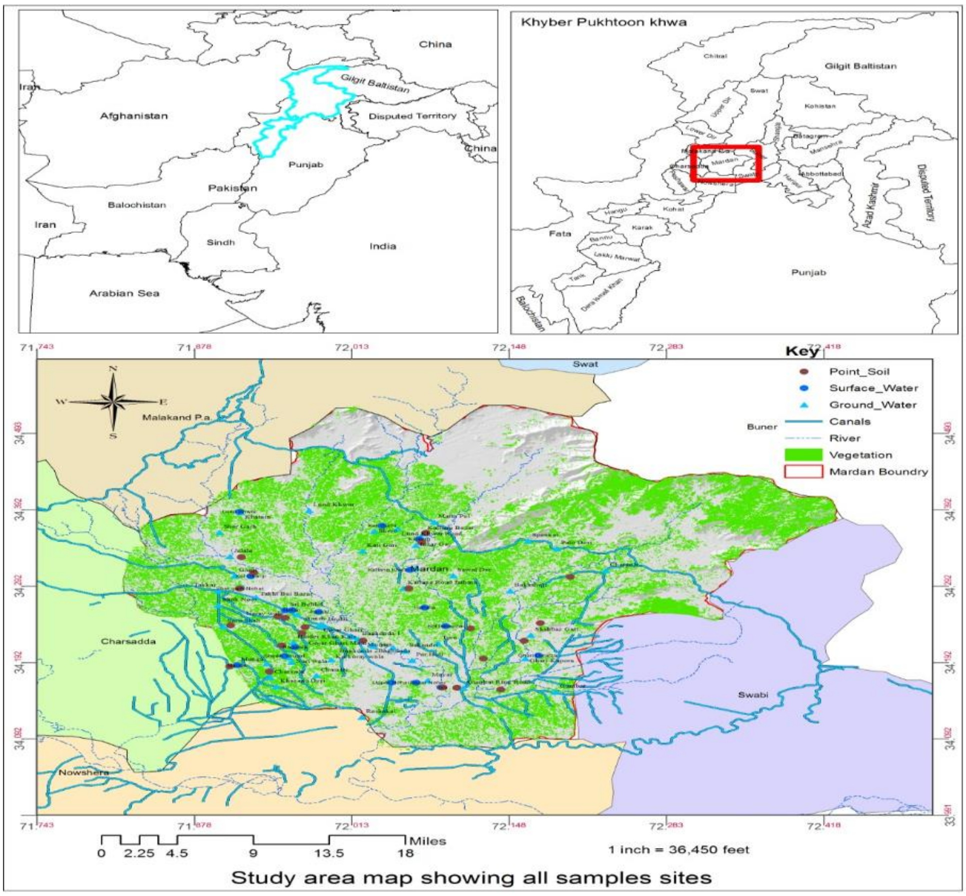


Fig- 2.1 Composite map of the study area showing sampling locations with other features

2.2 Collection and preliminary treatment of samples

2.2.1 Food Crops Sampling

Specimens were taken from locally harvested farming food crops consisting of grains (*Triticum aestivem*, *Zea mays*, *Oryza sativa*) having n =15, and vegetables (*Abelmoschus esculantus*, *Solanum tuberosum*, *Brassica rapa*, *Colocasia antiquoeum*, *Momordica charantia*, *Solanum lycopersicum and Solanum melongena*) n= 35 **(Table- 2.1**). All the samples were put in polyethylene bags and shifted to the research center where all the samples were washed with tap water and then with distalled water. After this, the eatable portions were separated and desiccated at room temperature for a day and then subsequently dried in the oven at 70-80^{o} C for about 24 hours. Further, the dried-up samples were crushed with an electric mincer and kept properly in paper bags at room temperature for the next step i.e. extraction (Jan et al., 2010b).

Table- 2.1 Food crops (n^{a} = 50) with their English, local, botanical and family names collected from the study area.

S. No	English Name	Local Name	Botanical Name	Family Name
	Grains (n = 15)			
01	Wheat	Ghanam	*Triticum aestivum*	Poaceae
02	Maize	Makhai/Jowar	*Zea mays*	Poaceae
03	Rice	Chawal	*Oryza sativa*	Poaceae
	Vegetables (n = 35)			
04	Brinjil	Baingun	*Solanum melongena*	Solanaceae
05	Tomato	Tamator	*Solanum lycopersicum*	Solanaceae
06	Lady finger	Bindhi	*Abelmoschus esculantus*	Malvaceae
07	Potato	Aloo	*Solanum tuberosum*	Solanaceae

08	Bitter gourd	Karela	*Momordica charantia*	Cucurbitaceae
09	Turnip	Shaljum	*Brassica rapa*	Brassicaceae
10	Eddoe	Kachalo	*Colocasia antiquorum*	Araceae

[a]Number of samples.

2.2.2 Collection and pretreatment of human blood specimens

Prosedure used by Jan et al., 2011 was applied for the collection of human blood specimens, from the area under study. The donors in every location were divided into two classes i.e. adults having the age 15 years and above and children less than 15 years. Blood specimens were collected from the native residents after consent. Samples (5ml) were taken with disposable syringes and putted the blood in neat polypropylene EDTA enclosed tubes to get away coagulation. Subsequently all the samples were properly tagged and moved to research laboratory in ice box for the coming stage i.e extraction.

2.3 Extraction of samples

2.3.1 Extraction of food crops samples

Extraction of food crops samples were done by procedure used by Khan et al. 2010. Samples were put in conical flasks and mix with 10 ml of concentrated Nitric acid (HNO3) and kept for a night for passive digestion. After calm digestion the samples were get heated on hot plate till almost to dryness.

After cooling the samples, we added 5 mL Perchloric acid ($HClO_4$) and heated again at different increasing temperatures till a clear extract remains. After this the samples were cooled and were filtered by using Wattman filter paper of No. 42 and made diluted with distalled water upto the final volume of 100 ml and then transferred into 100 ml specimen bottles and kept at room temprature for final examination in the Centralized Resource Laboratory, University of Peshawar for the final anlysis of the heavy metals on Atomic Absorption Spectrophotometer (Perkin Elmer AAS-700).

2.3.2 Extraction of human blood samples

Procedure used by Jan et al., 2011 was applied for extraction of human blood specimens. Here we put 3 ml of blood specimens in conical flasks and added a combine volume of 15 ml HNO_3 and $HCLO_4$ prepared in 4:1 ratio respectively. After keeping over night for cool digestion, the samples were heated on hot plate till a transparent concentrate remains. Then after cooling, the concentrate was filtered and thined upto the last volume of 100 ml by mixing with condensed water and were preserved at normal temperature for final step i.e. the analysis step.

2.4 Reagents and analytical procedures

Chemicals of high grade and a high spectroscopic precision of 99.9% (Merck, Germany) were applied for the specimens' extraction. For every metal, standards solutions were made from thier stock solution by diluting their relevant 1000 mg/L certified standards (Fluka Busch Switzerland). The concentration of substantial metals in all the specimens were investigated by Atomic Absorption Spectrophotometer (Perkins Elmer-700) fixed by graphite furnance, at the the Centralized Resourced Laboratory (CRL) in Geology Dept. University of Peshawar **(Table-2.2).**

The precision and accuracy of our process were cross tested by employing the sample-free chemicals and Standard Reference Materials (SRMs) for the selected metals. Also for the quality of our data, every sample batch was examined three times in the standard optimum state with a 95% of assurance level. Accuracy and consistency of analysis were ensured by analyzing blank and standard solution after every 10 samples for verification

Table- 2.2 Instrumental analytical conditions for the analysis of heavy metals

Metal	Acetylene (L/min)	Air (L/min)	Wavelemgth (nm)	Slit width (nm)	Lamp current (Ma)	Detection limit (mg/L)
Cd	2.0	17.0	228.8	0.7	4	0.0008
Cr	2.5	17.0	357.9	0.7	25	0.0030
Cu	2.0	17.0	324.8	0.7	15	0.0015
Mn	2.0	17.0	279.5	0.2	20	0.0015
Ni	2.0	17.0	232.0	0.2	25	0.0060
Pb	2.0	17.0	283.3	0.7	30	0.0150
Zn	2.0	17.0	213.9	0.7	15	0.0015

spectrometer function. The absorption and delay interval of spectrometer was 5 seconds. While the detection limit of this spectrometer for Cu, Mn, Pb, Cr, Zn, Ni, and Cd were 0.0015, 0.0015, 0.0150, 0.0030, 0.0015, 0.0060, 0.0008mg/L correspondingly.

2.5 Data analysis for different indicators

2.5.1 Daily Intake of Metals (DIM)

The average daily intake of food crops both for adults and for children was estimated alongwith the blood samples collection. The average daily intake of metals (DIM) was calculated by using the following Eq. 02 (Khan et al., 2010; Jan et al., 2010b).

$DIM = C_{metal} \times C_{factor} \times D_{food\ intake} / BW_{average\ weight}$ Eq. 02

where C_{metal}, C_{factor}, $D_{food\ intake}$ and $BW_{average\ weight}$, indicate the HM concentrations in plants/crops (mg/kg), conversion factor, average daily intake of food crops and consumers'body weight, correspondingly. The transfiguration value of 0.085 were utilized to change wet wight of crops into the dried ones. While the typical everyday ingestion of foodstuff for the children and adults were thought and taken to be as 0.232kg/day and 0.345kg/day correspondingly (Khan et al., 2008) and likewise the normal weight of the children and adults were taken as 32.7kg and 73kg individually (Jan et al., 2010b).

2.5.2 Health risk index (HRI)

For the calculation of the prolonged health hazard value to the public of the area via consuming unhealthy food crops, their HRI was calculated for each metal by applying the Eq. 03 (Jan et al., 2010b and Khan et al., 2008)

$HRI = DIM / RfD$ Eq- 03

In the above equation the DIM, HRI and RfD, denote the daily intake of metals, health risk index and reference doses for the metals respectively. Moreover the reference doze (RfD) standards for Cr, Zn, Ni, Cu, Mn, Pb, Cd and Pb were put as 1.5, 3.0E-01, 2.0E-02, 3.7E-02, 1.4E-01, 4.0E-03, 5.0E-04mg/kg/day correspondingly (Shah et al., 2012). The inhabitants are thought to be safe when HRI value is less than 1.(Khan et al., 2008).

2.5.3 Accummulative hazard index (HI)

The aggreagte prolonged health hazards for many toxic metals is spoken as Hazard Index (HI) and were measured in the study by using below equation 04, (Haung et al., 2008; Cao et al., 2014; Bermudez et al., 2011; Khan et al.,2014)

$HI = HRI_{Cr} + HRI_{Cd} + \ldots + HRI_{n}$ Eq. 04

Here the HI represent the cumulative hazard index while the HRI reflects the estimated health risk index values for each metals.

When the HI value is greater than 1 then the people are said to be unsafe due to the combined hazards of all the heavy metals ingested (Khan et al., 2013a).

2.6 Statistical calculations of the data

The data was analyzed by using Microsoft Excel 2010 and Sigma Plot 10.0 for processing and analysis of data to accomplish our results visually in easily understandable form. The measurements and calculations were extracted and conveyed in the respect of range, standard deviation and mean values of the data. Moreover, different location maps for the investigated area was prepared through Arc-GIS software.

3. Results and discussion

3.1 Concentrtion of heavy metals in food crops

Table- 3.1 outlines the mean concentrations of heavy metals in farming food crops collected from all the five sites which are varied across the area. Overall the intensities of these metals location wise, crop wise and metal wise were found in the order of Gujjar ghari > Takht bhai > Par hoti > Katlang > Khazana deri; Triticum aestivum > Solanum melongena > Zea mays > Oryza sativa > Abelmoschus esculantus > Solanum tuberosum > Colocasia antiquorum > Brassica rapa > Momordica charantia > Solanum lycopersicum and Zn > Mn > Cr > Ni > Cu > Pb > Cd, respectively.

The mean concentration of Cd in all the crops (grain and vegetables) was found as 0.64 mg/kg **(Table- 3.1)** which is lesser than the values reported by Khan et al. (2010) and higher than Kumar et al. (2016). It mostly exceeded its permissible limits set by SEPA (2005) sourced from Khan et al. (2010). Cd was observed lowest (0.05 mg/kg) in Abelmoschus esculantus at Katlang and highest (1.75 mg/kg) in Zea mays at Gujar ghari. On the other hand the mean concentration of Cr and Ni were almost found with same value of 2.61 mg/kg and 2.59 mg/kg respectively, which is greater than as reported by Shaheen et al. (2016).

Table- 3.1Selected elemental intensities (mg/kg) in food harvest (n^a = 50) assembled in the study area

Food Harvest	Cd	Cr	Cu	Mn	Ni	Pb	Zn
Grains (n = 15)							
Triticum aestivum	0.64±0.16	2.99±0.57	4.56±0.99	9.28±3.54	2.37±0.79	0.98±0.38	12.85±3.54
Zea mays	0.82±0.61	2.69±0.82	1.92±1.42	5.26±2.03	2.55±0.97	1.02±0.40	11.75±3.69
Oryza sativa	1.01±0.51	2.73±1.72	3.05±1.07	4.39±1.44	2.98±1.22	1.17±0.63	9.39±2.43
Vegetables (n = 35)							
Solanum melongena	0.92±.41	7.63±1.24	3.05±1.39	3.32±2.29	2.98±1.84	6.23±1.38	5.44±1.07
Solanum lycopersicum	0.41±0.70	1.55±0.65	0.82±0.23	1.29±0.85	2.22±0.39	0.88±0.41	6.54±1.47
Abelmoschus esculantus	0.49±0.40	1.80±0.56	1.46±0.36	2.94±0.91	3.27±1.20	0.77±0.55	10.62±3.65
Solanum tuberosum	0.47±0.32	1.95±1.10	2.11±0.60	2.20±0.26	2.354±0.69	1.19±0.53	9.74±4.99
Momordica charantia	0.72±034	1.69±0.72	0.95±0.52	1.15±0.81	2.60±1.55	0.70±0.71	6.80±2.59
Brassica rapa	0.18±0.13	1.36±0.94	0.82±0.41	1.12±0.43	2.29±0.84	0.81±0.48	8.48±1.77
Colocasia antiquorum	0.47±0.55	1.78±0.59	3.28±0.30	3.17±1.63	2.40±0.84	0.81±0.67	7.27±2.35

[a] Number of samples

While the mean concentration of Cu was found 2.2 mg/kg which is lesser than that of Shaheen et al. (2016) and Amin et al. (2013).

The mean concentration of Cu was found within the safe limits set by FAO/WHO, 2011, sourced from Shaheen et al. (2016).

Morover, Mn, the second most measured toxic metal in this study covers a vide range of variation in food crops with a mean concentration of 3.41 mg/kg.

The mean observed Mn was manifold below of that reported by Kumar et al.

(2016) and Shaheen et al.

(2016). Also the concentrations of Pb were measured with a mean value of 1.19 mg/kg **(Table- 3.1)**, which exceeded its maximum permissible limits (MPL) set by FAO, 2001 but comes within the limits set by SEPA (2005) sourced from Khan et al.

2010, which is 09 mg/kg. The mean concentration noted by Islam et al.

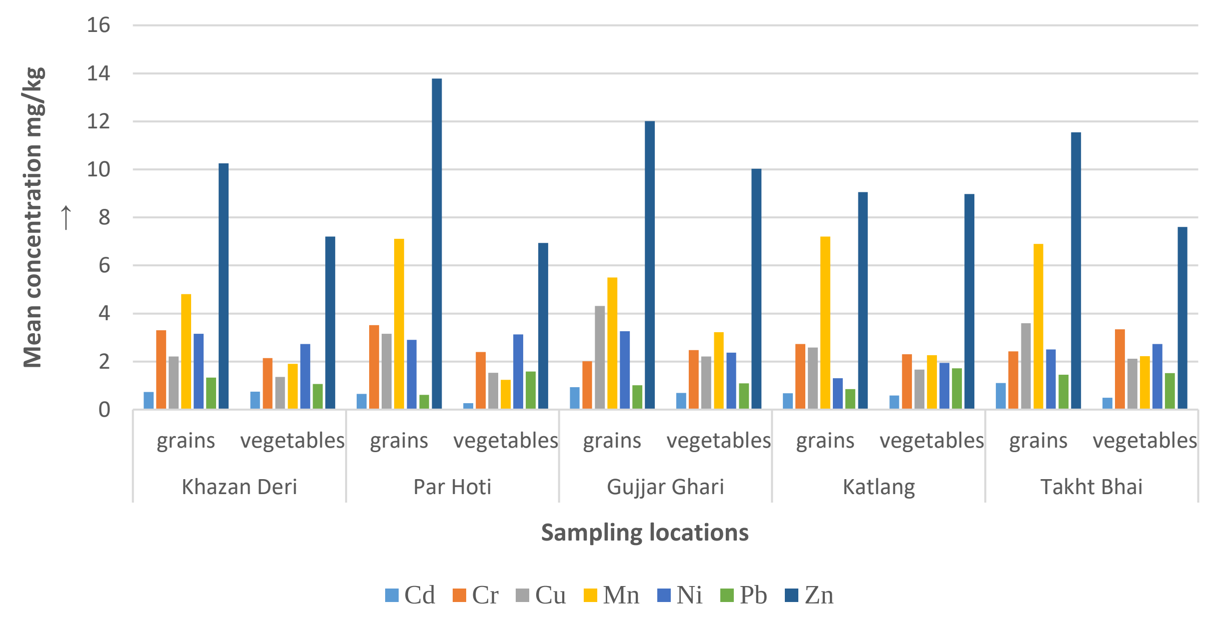
(2015) was found higher than ours. Lastly Zn was measured with a mean concentration of 8.8 mg/kg; having its lowest amount (1.51 mg/kg) in Solanum tuberosum at Khazana deri area and its highest value of 16.4 mg/kg at Par hoti in Triticum aestivum. Here the obtained concentration of Zn was less than that recorded by Amin et al. (2013) and higher than that reported by Khan et al. (2013). **Fig- 3.1** shows a flow chart of higher metal content in grains as compared to vegetables. Therefore it is very clear that concentration also vary throughout the area. Specifically the lower plain and fertile area showed greater metal contents as compared to the upper somewhat less polluted locations.

This difference might be linked with different man-made practices carried out in the area, like extensive and intensive farming, industrial activities, poor land use plans and shortage of surface irrigation water; which may push the farmers to use the wastewater for irrigation purposes mostly in pri-urban fields in the areas like Par hoti, Mayar, Toru, Bughdada etc. Such poor practices spoil the soil natural chemical balance.

Table-3.2 enlists the whole summary of the heavy metals concentration in all the sites. Overall the elements' occurrence for the foodstuffs were noted in the order of Zn > Mn > Cr > Ni > Cu > Pb > Cd with the overall mean of 8.88, 3.41, 2.61, 2.60, 2.20, 1.45 and 0.64 mg/kg respectively.

Here Cd, Cr and Ni

Fig 3.1 **Mean concentration (mg/kg) of heavy metals in crops**



were mostly observed with a higher concentration than their permissible limits which may accumulate in the consumers of the area.

Table- 3.2 Summary of the heavy metals concentration (mg/kg) in agrarian food items obtained from the investigated locations

Foodstuff	Statistics	Cd	Cr	Cu	Mn	Ni	Pb	Zn
Grains (n=15)	Mean	0.82±0.46	2.8±1.07	4.06±0.85	6.82±2.93	3.90±0.59	3.18±2.35	13.29±1.94
	Range	0.15-1.75	0.35-5.15	3.10-6.05	3.85-12.80	3.20-4.90	1.35-7.75	1.70-16.40
Vegetables (n= 35)	Mean	0.56±0.41	2.53±2.25	1.40±0.67	1.95±1.00	2.04±0.64	0.71±0.37	7.00±2.10
	Range	0.05-1.45	0.40-9.20	0.35-2.65	0.25-3.65	0.25-3.00	0.05-1.30	1.51-10.45

3.2 Concentration of heavy metals in human blood specimens

Table- 3.3 covers a transitory about the concentration of heavy metals in the human blood collected from the study area..

The concentrations were found in the adult blood samples in the order of Zn > Cr > Mn > Cu > Ni > Pb > Cd with the calculated mean concentration of 0.654± 0.015, 0.594± 0.142, 0.53± 0.08, 0.506±0.122, 0.384±0.103, 0.366± 0.051 and 0.292±0.08 mg/kg, respectively. While in children we noted as Zn > Cr > Cu > Ni > Mn > Pb > Cd with values of

Table- 3.3 Intenstities (mg/L) of toxic elements's addition in the blood specimens (n= 80) of consumers from the area.

Sites	Donors	Indicators	Cd	Cr	Cu	Mn	Ni	Pb	Zn
Khazana Deri	Adult (08)	Range	0.12-0.60	0.11-0.97	0.28-0.91	0.23-0.93	0.05-0.73	0.08-0.56	0.32-1.12
		Mean	0.34±0.16	0.48±0.39	0.36±0.25	0.55±0.26	0.22±0.23	0.31±0.18	0.65±0.32
	Children (08)	Range	0.08-0.60	0.31-0.94	0.17-1.03	0.20-1.06	0.06-0.93	0.12-0.86	0.25-1.15
		Mean	0.24±0.18	0.49±0.24	0.41±0.30	0.38±0.29	0.35±0.30	0.44±0.25	0.70±0.38
Par Hoti	Adult (08)	Range	0.03-0.43	0.24-1.36	0.09-0.86	0.14-0.90	0.21-0.66	0.12-0.93	0.30-1.29
		Mean	0.17±0.13	0.76±0.43	0.45±0.27	0.50±0.24	0.39±0.19	0.40±0.32	0.68±0.39
	Children (08)	Range	0.04-0.45	0.11-1.27	0.07-0.96	0.21-0.78	0.07-1.06	0.03-0.70	0.24-1.29
		Mean	0.29±0.12	0.57±0.49	0.34±0.29	0.39±0.18	0.41±0.33	0.28±0.23	0.72±0.47
Gujar Ghari	Adult (08)	Range	0.16-0.46	0.25-1.16	0.28-1.06	0.12-1.06	0.08-1.13	0.20-0.76	0.12-1.28
		Mean	0.29±0.10	0.73±0.39	0.69±0.24	0.40±0.31	0.45±0.37	0.43±0.21	0.65±0.46
	Children (08)	Range	0.12-0.53	0.24-0.93	0.13-0.83	0.16-1.01	0.22-1.17	0.02-0.52	0.24-0.72
		Mean	0.32±0.14	0.41±0.28	0.42±0.27	0.44±0.29	0.48±0.32	0.27±0.17	0.42±0.18
Katlang	Adult (08)	Range	0.09-0.67	0.21-0.83	0.14-1.16	0.05-1.121	0.18-0.90	0.16-0.56	0.02-1.43
		Mean	0.40±0.22	0.43±0.20	0.54±0.36	0.58±0.39	0.49±0.26	0.32±0.11	0.65±0.52
	Children (08)	Range	0.05-0.69	0.12-1.31	0.03-0.73	0.035-0.66	0.19-0.83	0.07-0.35	0.21-1.13
		Mean	0.31±0.17	0.54±0.45	0.30±0.25	0.35±0.20	0.36±0.19	0.22±0.12	0.57±0.33
Takht Bhai	Adult (08)	Range	0.08-0.63	0.13-1.33	0.22-0.86	0.28-1.05	0.04-0.86	0.09-0.75	0.13-1.26
		Mean	0.26±0.18	0.57±0.48	0.49±0.25	0.63±0.28	0.37±0.26	0.37±0.22	0.64±0.39
	Children (08)	Range	0.06-0.71	0.12-0.86	0.11-0.83	0.06-0.93	0.12-0.73	0.12-0.76	0.22-1.16
		Mean	0.28±0.19	0.38±0.22	0.53±0.30	0.51±0.34	0.40±0.22	0.38±0.24	0.68±0.36

0.609±0.264, 0.560±0.0.361, 0.540±0.143, 0.50±0.336, 0.490±0.323, 0.412±0.33 and 0.38±0.323 mg/kg respectively. The Cd concentration for adults was noted as 0.34±0.16, 0.17±0.13, 0.29±0.10, 0.40±0.22 and 0.26±0.18 mg/L and for children Cd was observed as 0.24±0.18, 0.29±0.12, 0.32±0.14, 0.31±0.17 and 0.28±0.19 mg/L in Khazana dheri, Par hoti, Gujjar ghari, Katlang and Takht bhai respectively.

As for as the Cr concentration the adult (Khazana deri) and children (Par hoti) showed the least (0.11 mg/L) buildup in their blood but on the highest (1.36 mg/L) end, Cr was observed in the adults doners of Par hoti. The Cu concentrations were noted the highest in the adult consumers of katlang while the least in kids of par hoti. Almost the same tendency was seen for Mn and Ni with the greatest accumulation in the adults of katlang and in the children of gujjar ghari**.** And lastly Zn was observed with a slightly higher concentration as compared to the rest of metals.

3.3 Health risk assessment due to heavy metals intake

3.3.1 Risk valuation of heavy elements through consumption of Foodstuffs

3.3.1.1 Daily intake of metals

As one of the health risk parameters, DIM due to the consumption of contaminted food crops was calculated. **Table- 3.4** sum up the findngs of DIM of heavy metals both for children and adults.

Similar to concentrartion of metals in food crops, the CDI of heavy elements in consumers are almost found in the same proportions. Overall the mean DIM was observed as *Triticum aestivum* > *Solanum melongena* > *Zea mays* > *Oryza sativa* > *Abelmoschus esculantus* > *Solanum tuberosum* > *Colocasia antiquorum* > *Solanum lycopersicum* > *Brassica rapa* > *Momordica charantia*. While as per metals the DIM is as Zn > Mn > Cr > Ni > Cu > Pb > Cd with values of 7.26E-03, 3.65E-03, 1.74E-03, 1.31E-03, 1.11E-03, 8.49E-04 and 4.13E-04 respectively. While the rang od CDI was found as 7.22E-

05 to 9.77E-04, 5.46E-04 to 8.52E-03, 3.29E-04 to 2.75E-03, 4.50E-04 to 1.63E-02, 8.92E-04 to 1.97E-03, 2.81E-04

Table- 3.4 DIM (mg/kg/day) or Chronic Daily Intake (CDI) for each substantial element caused from consuming the studied food crops (*n*[a] = 50) cultivated in the area under study.

Food crops	Consumers		Cd	Cr	Cu	Mn	Ni	Pb	Zn
Grains (n = 15)									
Triticum aestivum	Adults	DIM	2.57E-04	1.20E-03	1.83E-03	1.08E-02	9.52E-04	3.94E-04	1.43E-02
	Children	DIM	3.86E-04	1.80E-03	2.75E-03	1.63E-02	1.43E-03	5.91E-04	2.14E-02
Zea mays	Adults	DIM	3.29E-04	1.08E-03	7.71E-04	2.92E-03	1.02E-03	4.10E-04	9.74E-03
	Children	DIM	4.95E-04	1.62E-03	1.16E-03	4.38E-03	1.54E-03	6.15E-04	1.46E-02
Oryza sativa	Adults	DIM	4.06E-04	1.10E-03	1.23E-03	1.76E-03	1.20E-03	4.70E-04	6.30E-03
	Children	DIM	6.09E-04	1.65E-03	1.84E-03	2.65E-03	1.80E-03	7.06E-04	9.46E-03
Vegetables (n = 35)									
Solanum melongena	Adults	DIM	6.51E-04	5.68E-03	1.23E-03	8.73E-03	1.20E-03	3.03E-03	3.99E-03
	Children	DIM	9.77E-04	8.52E-03	1.84E-03	1.31E-02	1.80E-03	4.54E-03	5.99E-03
Solanum lycopersicum	Adults	DIM	4.82E-04	7.03E-04	3.29E-04	5.18E-04	8.92E-04	3.54E-04	3.43E-03
	Children	DIM	7.24E-04	1.06E-03	4.94E-04	7.78E-04	1.34E-03	5.31E-04	5.15E-03
Abelmoschus esculantus	Adults	DIM	1.97E-04	1.08E-03	5.87E-04	1.38E-03	1.31E-03	3.09E-04	6.40E-03
	Children	DIM	2.96E-04	1.63E-03	8.80E-04	2.07E-03	1.97E-03	4.64E-04	9.61E-03
Solanum tuberosum	Adults	DIM	3.49E-04	7.83E-04	8.48E-04	8.84E-04	9.40E-04	7.99E-04	4.44E-03
	Children	DIM	5.25E-04	1.18E-03	1.27E-03	1.33E-03	1.41E-03	1.20E-03	6.66E-03
Momordica charantia	Adults	DIM	3.70E-04	6.79E-04	3.82E-04	4.62E-04	1.04E-03	2.81E-04	2.73E-03
	Children	DIM	5.55E-04	1.02E-03	5.73E-04	6.94E-04	1.57E-03	4.22E-04	4.10E-03
Brassica rapa	Adults	DIM	7.22E-05	5.46E-04	3.29E-04	4.50E-04	9.20E-04	3.25E-04	3.41E-03
	Children	DIM	1.09E-04	8.20E-04	4.94E-04	6.75E-04	1.38E-03	4.88E-04	5.11E-03
Colocasia antiquorum	Adults	DIM	1.89E-04	1.04E-03	1.32E-03	1.27E-03	9.64E-04	4.22E-04	3.32E-03
	Children	DIM	2.83E-04	1.56E-03	1.98E-03	1.91E-03	1.45E-03	6.33E-04	4.99E-03

[a] Number of samples

to 4.54E-03 and 2.73E-03 to 2.14E-02, for Cd, Cr, Cu, Mn, Ni, Pb and Pb in that order. **Table- 3.4** shows the CDI of grains (3.48E-03) is pretty higher than that observed for vegetables (1.84E-03). Moreover, the children were found vulnerable to metals toxicity as they exhibited higher CDI of 2.80E-03 as compared adults CDI of 1.86E-03.

5.4.1.2 Health risk indexes of metals

Likewise, the DIM, the Health risk indexes (HRI) were also calculated for all our seven listed heavy elements, by dividing their measured DIM with their corresponding reference doses (RfD). The RfD values for Cd, Cr, Cu, Mn, Ni, Pb and Zn were considered as 0.001, 1.5, 0.04, 0.033, 0.02, 0.004 and 0.30 mg/kg bw/day respectively (US-EPA, 2002; Khan et al. 2010). The HRI value below 1 is thought to be safe with no significant risk to the human health.

Table- 3.5 covers the calculated HRI values both for adults and childre. The overall combined (adults and children) HRI values for Cd, Cr, Cu, Mn, Ni, Pb and Zn were found in a series as 7.23E-02 to 9.77E-01, 3.64E-04 to 5.68E-03, 8.24E-03 to 6.87E-02, 3.21E-03 to 1.16E-01, 4.46E-02 to 7.04E-02, 1.17E-02 to 7.56E-01 and 9.11E-03 to 7.15E-02, respectively. The highest mean HRI value of 9.77E-01 was measured for Cd in children through its intake from Solanum melongena while the Cr achieved the lowest mean level of HRI i.e 3.64E-04 in adults through consumption of Brassica rapa. The HRI values in case of adults for Cd, Cr, Cu, Mn, Ni, Pb and Zn were found in a series as 7.23E-02 to 6.50E-01, 3.64E-04 to 3.78E-03, 8.24E-03 to 4.58E-02, 3.21E-03 to 7.74E-02, 4.46E-02 to 6.57E-02, 7.03E-02 to 7.56E-01 and 9.11E-03 to 4.76E-02, respectively. But for children the HRI values are somewhat higher than adults. It is obvious from **Table-3.5** and **Fig- 3.2** that Cd has reached to higher mean HRI of 9.77E-01 but also exceeded in some places beyond its standerd value of 1 in 6% of the total collected food crops samples for adult and 10% in case of children's HRI in the area. But the overall Cd observed HRI was much lower than that reported by Singh et al. (2010). The HRI values reported by Nawab et al. (2016) were found lesser and that reported by Khan et al. (2010) were found greater than the mean HRI in this study.

Table- 3.5Health Risk Index (HRI) for separate heavy metals caused by the consumption of different food crops (n^a = 50) grown in the study area

Food Crops	Consumers		Cd	Cr	Cu	Mn	Ni	Pb	Zn
Grains (n = 15)									
Triticum aestivum	Adults	HRI	2.57E-01	8.01E-04	4.58E-02	7.74E-02	4.76E-02	9.84E-02	4.76E-02
	Children	HRI	3.86E-01	1.20E-03	6.87E-02	1.16E-01	5.10E-02	1.64E-02	7.15E-02
Zea mays	Adults	HRI	3.29E-01	7.20E-04	1.93E-02	2.08E-02	5.12E-02	1.02E-01	3.25E-02
	Children	HRI	4.94E-01	1.08E-03	2.89E-02	3.13E-02	5.49E-02	1.71E-02	4.87E-02
Oryza sativa	Adults	HRI	4.05E-01	7.31E-04	3.06E-02	1.26E-02	5.99E-02	1.17E-01	2.10E-02
	Children	HRI	6.09E-01	1.10E-03	4.60E-02	1.89E-02	6.42E-02	1.96E-02	3.15E-02
Vegetables (n = 35)									
Solanum melongena	Adults	HRI	6.50E-01	3.78E-03	3.06E-02	6.24E-02	5.99E-02	7.56E-01	1.33E-02
	Children	HRI	9.77E-01	5.68E-03	4.60E-02	9.36E-02	6.42E-02	1.26E-01	2.00E-02
Solanum lycopersicum	Adults	HRI	4.82E-01	4.69E-04	8.24E-03	3.70E-03	4.46E-02	8.84E-02	1.14E-02
	Children	HRI	7.23E-01	7.04E-04	1.24E-02	5.56E-03	4.78E-02	1.47E-02	1.72E-02

Abelmoschus esculantus	Adults	HRI	1.96E-01	7.23E-04	1.47E-02	9.87E-03	6.57E-02	7.73E-02	2.13E-02
	Children	HRI	2.95E-01	1.09E-03	2.20E-02	1.48E-02	7.04E-02	1.29E-02	3.20E-02
Solanum tuberosum	Adults	HRI	3.49E-01	5.22E-04	2.12E-02	6.31E-03	4.70E-02	1.99E-01	1.48E-02
	Children	HRI	5.24E-01	7.84E-04	3.18E-02	9.48E-03	5.04E-02	3.33E-02	2.22E-02
Momordica charantia	Adults	HRI	3.69E-01	4.53E-04	9.54E-03	3.30E-03	5.22E-02	7.03E-02	9.11E-03
	Children	HRI	5.54E-01	6.79E-04	1.43E-02	4.95E-03	5.60E-02	1.17E-02	1.37E-02
Brassica rapa	Adults	HRI	7.23E-02	3.64E-04	8.24E-03	3.21E-03	4.60E-02	8.13E-02	1.14E-02
	Children	HRI	1.08E-01	5.47E-04	1.24E-02	4.82E-03	4.93E-02	1.36E-02	1.70E-02
Colocasia antiquorum	Adults	HRI	1.88E-01	6.91E-04	3.29E-02	9.10E-03	4.82E-02	1.05E-01	1.11E-02
	Children	HRI	2.83E-01	1.04E-03	4.95E-02	1.37E-02	5.17E-02	1.76E-02	1.66E-02

[a]Number of samples

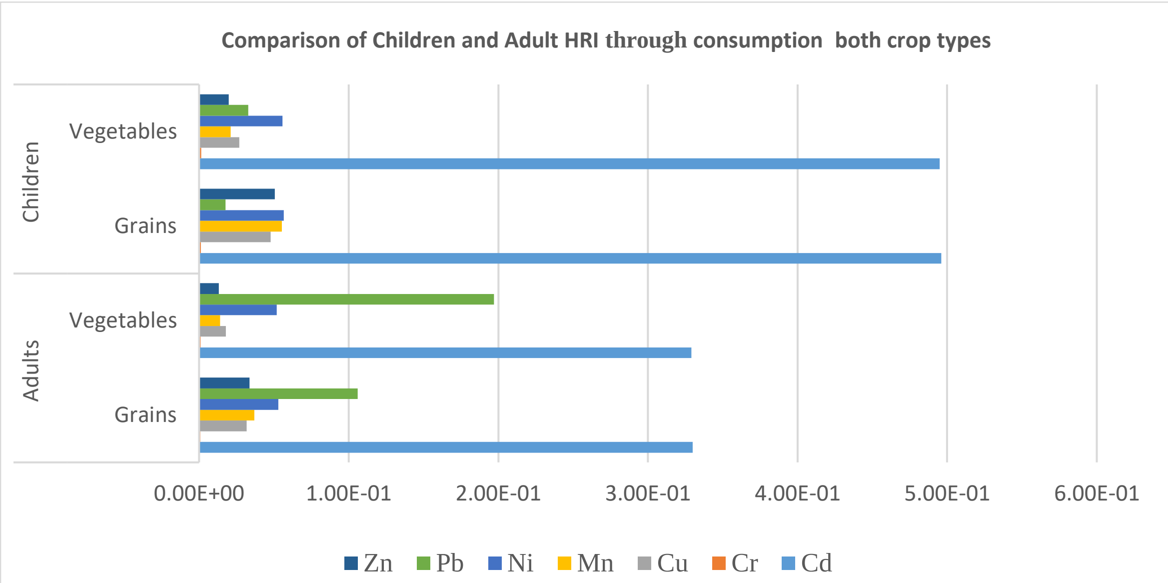


Fig. 3.2 Comparison of HRIs for Kids and Adults through intake of vegetables and grains

3.3.2 Accumulative risk assessment

The collective ingestion of heavy metals by eating the polluted food harvests might produce to a considerable long-lasting addition of noxious metals in the peoples of the area. Coming next, is a momentary debate on the joint DIMs, HRIs and the final HI findings of all nominated lethal heavy elements.

3.3.2.1 Accumulative Daily Intake of Metals (DIM)

The inclusive mean DIM of the heavy metals by consumption of agrarian food crops by the people of the study area, as per **Table- 3.6**. The whole breakup shows the flow of overall mean DIM together via using all the targeted agricultural crops and water, as Pb > Cr > Cd > Mn > Zn > Ni > Cu with a mean observed DIM like 3.05E-01, 1.34E-01, 1.24E-01, 1.03E-01, 5.55E-02, 4.67E-02 and 3.97E-02 mg/kg-day correspondingly. It is also shown that the DIM by the children are slightly greater than adults. While the sample wise input was assessed as grains (3.49E-03 mg/kg-day) > vegetables (1.84E-03 mg/kg-day).

5.4.3.2 Accumulative health risk indexes (HRIs) and hazard index (HI)

It is very important to know about the total health risk associated with the intake of toxic metals through different foodstuffs, which can be obtained by adding the HRIs values of

Table. 3.6.Calculated daily intake of metals (DIM) for analyzed toxic elements by various foodstuff (n=50) and water (n=55) ingestion of the investigated area.

Foodstuff	Consumers	Statistics	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Mean DIM	DIM
grains (n=15)	Adults	DIM	3.31E-04	1.13E-03	1.28E-03	5.16E-03	1.06E-03	4.24E-04	1.01E-02	2.78E-03	3.49E-03
	Children	DIM	4.96E-04	1.69E-03	1.92E-03	7.78E-03	1.59E-03	6.37E-04	1.52E-02	4.19E-03	
Vegetables (n= 35)	Adults	DIM	3.30E-04	1.50E-03	7.17E-04	1.96E-03	1.04E-03	7.88E-04	3.96E-03	1.47E-03	1.84E-03
	Children	DIM	4.95E-04	2.25E-03	1.08E-03	2.94E-03	1.56E-03	1.18E-03	5.94E-03	2.21E-03	
Water (n= 55)	Adults	DIM	3.53E-01	3.78E-01	1.11E-01	2.87E-01	1.31E-01	8.68E-01	1.42E-01	3.24E-01	3.41E-01
	Children	DIM	3.88E-01	4.17E-01	1.22E-01	3.16E-01	1.44E-01	9.56E-01	1.56E-01	3.57E-01	
foodstuff	**Consumers**	**Statistics**	1.24E-01	1.34E-01	3.97E-02	1.03E-01	4.67E-02	3.05E-01	5.55E-02	**1.15E-01**	

Table. 3.7.Evaluated Hazard Index(HI) and HRIs for analyzed toxic metals by various foodstuff (n=50) and water (n=55) utilization in the studied area.

Foodstuff	Consumers	Statistics	Cd	Cr	Cu	Mn	Ni	Pb	Zn	HI	Grand HI
grains (n=15)	Adults	HRI	3.30E-01	7.51E-04	3.19E-02	3.69E-02	5.29E-02	1.06E-01	3.37E-02	5.92E-01	1.32E+00
	Children	HRI	4.96E-01	1.13E-03	4.79E-02	5.54E-02	5.67E-02	1.77E-02	5.06E-02	7.25E-01	
Vegetables (n= 35)	Adults	HRI	3.29E-01	1.00E-03	1.79E-02	1.40E-02	5.19E-02	1.97E-01	1.32E-02	6.24E-01	1.28E+00
	Children	HRI	4.95E-01	1.50E-03	2.69E-02	2.10E-02	5.57E-02	3.28E-02	1.98E-02	6.53E-01	
Water (n= 65)	Adults	HRI	7.35E-01	2.40E-03	3.42E-02	2.01E-02	7.04E-02	2.29E-02	4.90E-03	8.90E-01	1.87E+00
	Children	HRI	8.09E-01	2.65E-03	3.77E-02	2.22E-02	7.75E-02	2.52E-02	5.40E-03	9.80E-01	
foodstuff	**Consumers**	**HI**	**3.19E+00**	**9.43E-03**	**1.97E-01**	**1.70E-01**	**3.65E-01**	**4.02E-01**	**1.28E-01**	**4.46E+00**	**4.46E+00**

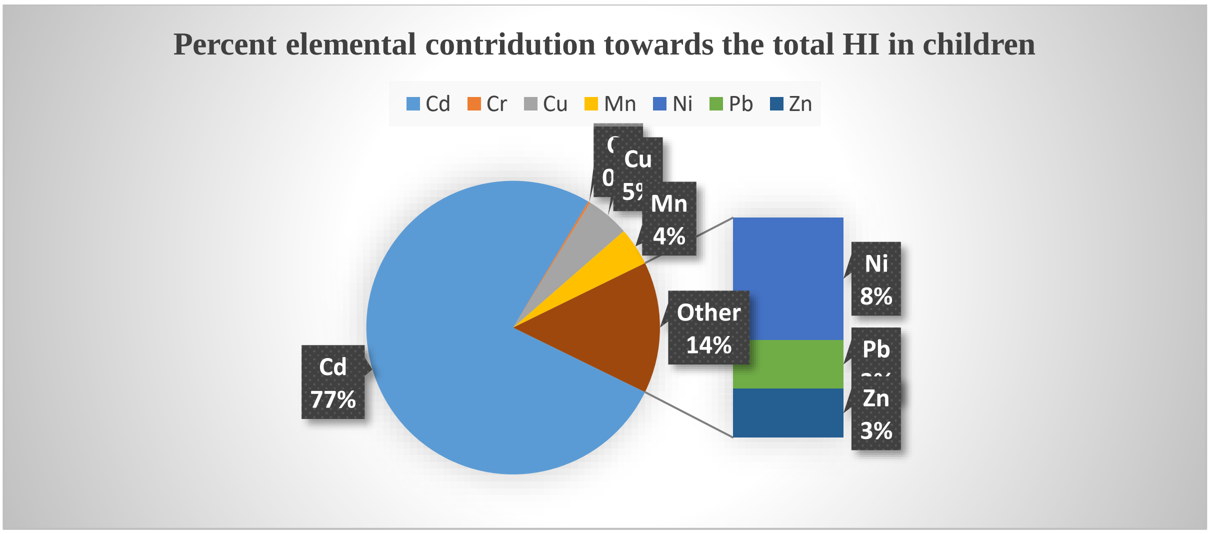


Fig 3.3Pie representation of individual share of heavy metals in total HI for Children

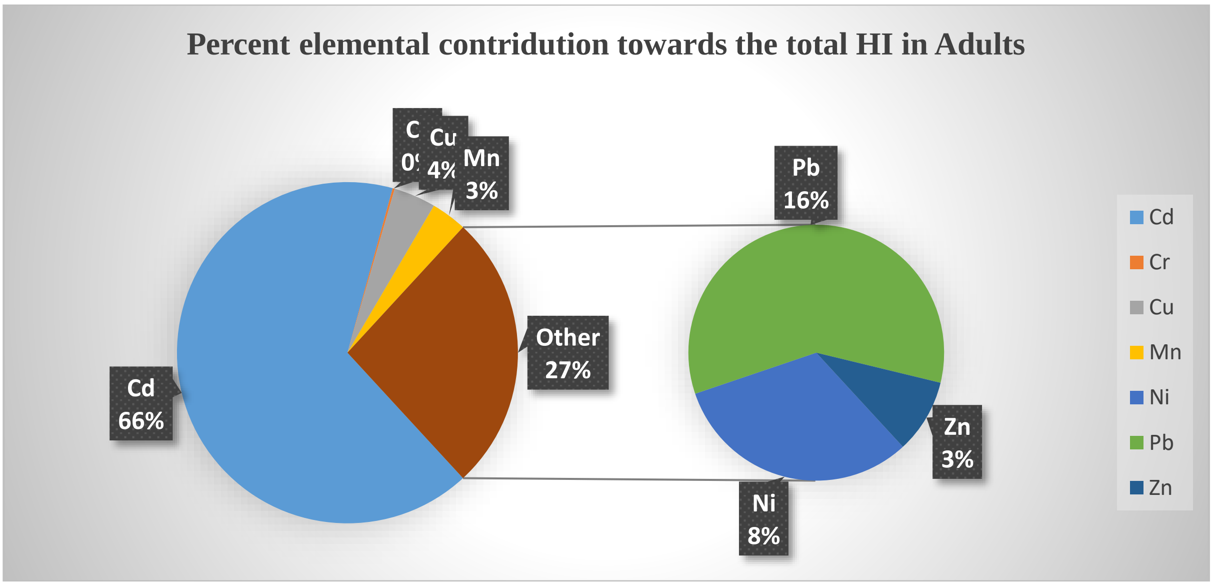


Fig- 3.4 Pie representation of individual metal contribution in total HI for Adults

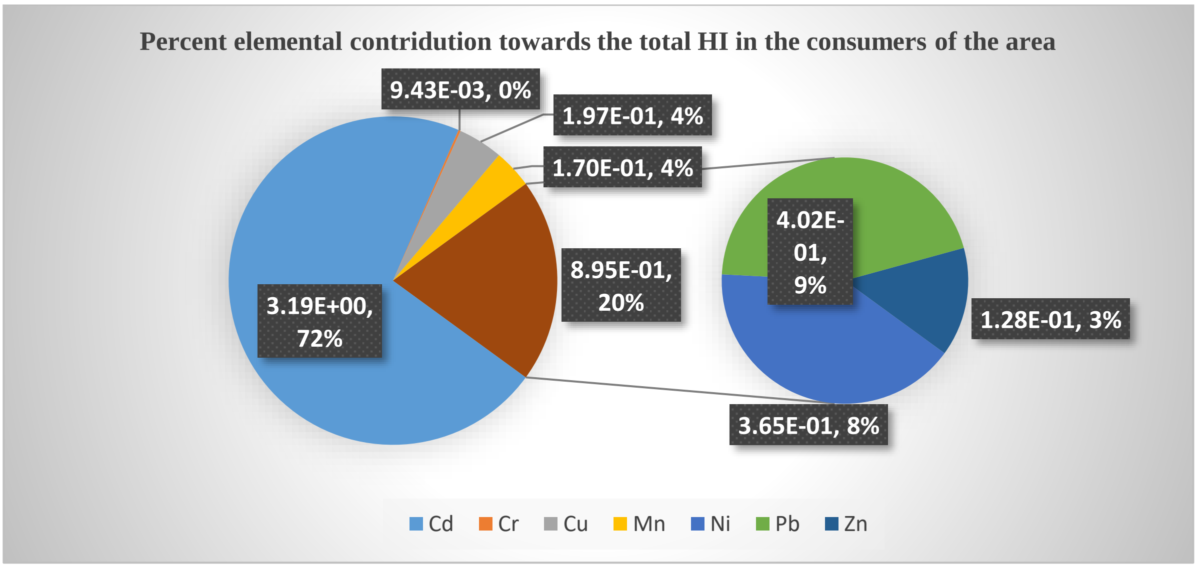


Fig- 3.5 Pie representation of the role of different metals in accumulative HI

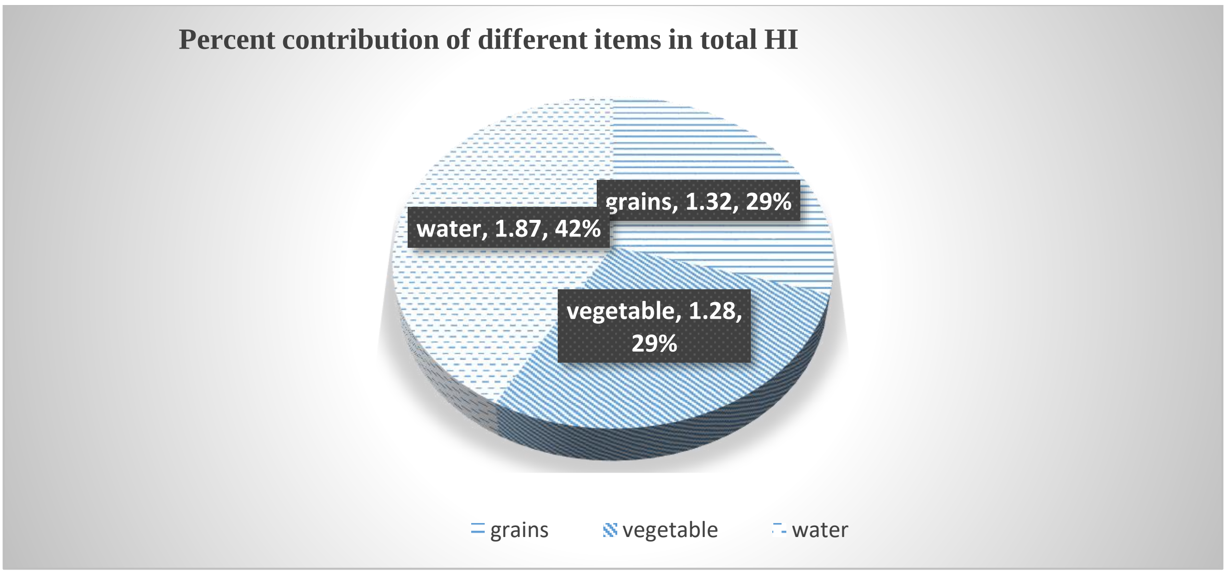


Fig- 3.63-D Pie representation of different items in accumulative HI

all the individual heavy metal/toxin taken via foodstuff. Such health risk parameter is known as hazard index (HI). The conclusive finding of the current study in the form of accumulative HRI or hazard index (HI) are clearly documented in statistical **Table 3.7** and presented visually in **Fig 3.3 to fig 3.6.** The table encapsulates the mean HRIs values for the investigated toxic elements associated with consumption of grains (n=15), vegetables (n=35) in the area. For adults' consumers of the area, the HRIs of Cd, Cr, Cu, Mn, Ni, Pb and Zn estimated in the series of 3.29E-01 to 7.35E-01, 7.51E-04 to 2.40E-03, 1.79E-02 to 3.42E-02, 1.40E-02 to 3.69E-02, 5.19E-02 to 7.04E-02, 2.29E-02 to 1.97E-01, 4.90E-03 to 3.37E-02, respectively. Similarly, the HRIs results of Cd, Cr, Cu, Mn, Ni, Pb and Zn for children were obtained in the pattern of 4.95E-01 to 8.09E-01, 1.13E-03 to 2.65E-03, 2.69E-02 to 4.79E-02, 2.10E-02 to 5.54E-02, 5.57E-02 to 7.75E-02, 1.77E-02 to 3.28E-02, 5.40E-03 to 5.06E-02, respectively.

As it is obvious from the the table **Table 3.7,** that all the estimated HRIs for the individual metal were found less than the benchmark value 1, therefore it may be stated that overall the residents of the area face no substantial health threat via consumption of grains, vegetables of the area. On the other side, the combined HRIs of each metal were observed in the oder of Cd > Pb > Ni > Cu > Mn > Zn > Cr. Moreover, the hazard index (HI) for every metal was calculated by totaling all the HRIs of each food class of the particular element to show the joint health risk raised jointly by all the selected metals. The cumulative HI value, for all the studied heavy metals, due intake of all foodstuff was valued as 4.46E+00, which is far greater than 1, indicating that although the individual HRIs value were less than 1 but the combined influence of all the selected heavy metals may possibly induce the health risks in consumers the area.

4 Conclusion

The mean concentration of Cd in all the crops (grain and vegetables) were found as 0.64 mg/kg, which mostly exceeded its permissible limits set by SEPA (2005) in about 72% of food crops. Moreover, Pb concentrations in crops was beyond its permissible limits by FAO in 78% but observed in limits set by SEPA (2005). While remaining metals were noted in safe limits. The highest DIM was obtained for Zn. HRI was largely noted less than 1, deficting that the consumption of food crops (grains and vegetables) cause no acute health risks in the area. Moreover, grains observed with considerably greater amounts of

heavy metals as compared to vegetables. Which is probably due to higher life span and greater retention time of grains in the fields. Also the intevsive use of agro-chemical almost at every stage of the crop's life; even during their production stage the farmers usually use to spray the pesticides, which are likely to absorb/adsorb to the usually exposed edible parts of vegetables. This poor practice of farmers may directly threat the consumers wellbeing.

As earlier in this study the estimated HRIs were found less than the standerd value 1, indicating the residents of the area face no significant health threat from the consumption of selected food items and drinkig water of the area. The combined HRIs of each metal were found as Cd > Pb > Ni > Cu > Mn > Zn > Cr. Additionally, the hazard index (HI) for a single metal were also calculated by adding all the HRIs of each food category of the respective metal. This was executed to show the joint health risk posed from grains (29.5%) and then the vegetables (28.6%).

The accumulative HI value, for all the evaluated heavy metals through intake of selected foodstuff was obtained greater than 1 indicating that although the individual HRIs values were less than 1 but the combined impact of all the selected heavy metals may probably exaggerate the health risks in the inhabitants of the area under study.

So its needed to mitigate the potential health risks associated with noxious substantial elemental exposure in the investigated area. The people of the area needs to improve their basic know-how about the consequences of different toxic materials. The farmers should be given the basic knowledge of modern and sustainable agriculture. The Government should bring relevant and timely legislations. The addition of heavy metals through effluents should be controled at their sources. And lastly the man-made erosion and deforestation in the upland areas needs to be controlled.

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