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Assessment of heavy metals in soil – vegetable system: level, transfer and public risk analysis

Muhammad Tansar Abbas*[1], Hidayat Ullah[1], Irshad Ullah[2], Muhammad Farooq[3]

[1] *Institute of Chemical Sciences, Gomal University, Dera Ismail Khan 29220, Pakistan*

[2] *University of Sawabi, Department of Pharmacy KPK, Pakistan*

[3] *NCE in Physical Chemistry, University of Peshawer, Pakistan*

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Abstract

Food safety is a global hazard. The Metropolitan and Industrialization have increased the level of Contaminations in the ecosystem. The current study was conducted to find out the toxic level of HMs in soil, water and contaminated vegetables, source apportionment and their impacts on the environment. A total of seventy-five samples of water, soil, and vegetables were investigated for the determination of heavy metal concentration levels through an atomic absorption spectrophotometer (AAS 7000). The sequence of HMs concentration in *Spinacia oleracea, Brassica oleraceae, Raphanus sativus, Lactuca sativa* and *Brassica Rapa* sub-sp.rapa was observed Fe > Cu > Mn > Zn > Cd > Ni > Pb > Cr respectively. Three latent origins were extracted for HMs (Ni, Zn, Fe, Mn, Cu and Pb) having eigen value greater than one by principal component analysis. The precarious value of heavy metal transfer factor (HMTF) for Zn (13.395) was observed in *Brassica oleraceae*. The average estimated daily ingestion of metals (EDIM) was calculated which showed that all trace metals are in acceptable limit. While the value of HRI was observed > 1 for Pb, Cd and Fe in targeted vegetables which showed that vegetables are not safe for human health.

* **Corresponding Author:** Muhammad Tansar Abbas ✉ mtansar5@gmail.com

Introduction

Fresh water is essential for healthy life. Unfortunately, increases in industrialization and food commodities have decreased the water availability at global level. Due to deficiency of clean water farmer used wastewater for irrigation purpose. The presence of toxic metals as a pollutant in soil and in floras is a universal challenge for the world. Heavy metals are unsafe for humans if they exceed the tolerable limits (Gebeyehu and Bayissa, 2020). The extreme level of toxic metals in ecosystem is due to spreading of metropolitan and industrial demand in progressive countries (Wong *et al.,* 2003) .The accumulation and transfer of poisonous metals in soil is due to irrigation channels. The accretion and contacts of trace metals in soil over a long period have serious threats to environment and living community (Gupta *et al.,* 2019). The existence of heavy metals in the blood is like a ligand because they can attach with different organ and can cause cancer, neurological and skin diseases which are harmful to health. The fundamental content of human diet is vegetable because it contains necessary nutrients like vitamins and minerals which are helpful for properly growth and can alleviate from numerous diseases (Aysha *et al.,* 2017). Unfortunately, the cultivation of vegetables in polluted soil can cause serious threats for users because it can build up contaminated metals from soil. The existence of toxic metals in vegetables is a major issue for the safety of human health. The ingestion of vegetables by human can cause the buildup of heavy metals through food chain process as a result a prolonged disease occurred (Tembo *et al.,* 2006). Vegetables build up toxic metals only from polluted soils in spite from their parts which are contaminated by impure air (Haiyan and Stuanes, 2003). It is reported that the ingestion of Pb, Cd and Cu polluted vegetables on daily basis having maximum hazards of extreme intestinal cancer (Edogbo *et al.,* 2020). In various previous research papers it is well documented that the major pollutant in vegetables is toxic metals in the world when it is properly grownup in infected soil (Vetrimurugan *et al.,* 2017). The normal use of polluted water in Pakistan is 0.88 10^9 m^3 annually from which 0.1510^{9}m^{3}is openly released to canals for irrigation purpose (Murtaza and Zia, 2012).In Pakistan the use of sewage water for harvesting is an old practice in urban areas due to industrial area, household activities and city rainfall. From the previous studies it is noted that at least 26% of vegetables production in urban areas of Pakistan is due to wastewater (Sarwar *et al.,* 2020). The contaminated water coming from industries and domestic activities is a major source for cultivation of vegetables in peri urban areas (Hembrom *et al.,* 2020).Muzaffargarh is an industrial area where a lot of industries are present like textile, sugar , thermal power and Parco respectively. The effluents coming from industries and domestic activities are used for cultivation of vegetables since 1920. The people living in selected community are still unaware from the threats of contaminated vegetables therefore this study designed with following indicators like heavy metal transfer factor (HMTF), metal daily ingestion (MDI) and health risk indices (HRI) for determination of health hazard related with polluted vegetables. For identification of probable sources of toxic metals in selected area various statistical tools like PCA (principal component analysis), Cluster analysis (CA) and correlation analysis are being used. The consequences in targeted study will give a new path for making strategies in reducing health hazards associated with toxic metals.

Materials and methods

Sampling region

Muzaffargarh is a well-known district of Punjab due to agricultural production. It is positioned at 30° 45 'N latitude and 71° 48' E Longitude. It is located between two famed rivers Chenab and Indus. The extreme recorded temperature is 54 °C and the lowest recorded temperature is –1 ºC. The average rainfall is 127 millimeters. Its whole zone is 830 thousand hectares from it 440 thousand hectares is used for cultivation. Its total populace is 3.56 million.

Sample collection

The total of seventy five samples of water, soil and vegetables were collected randomly from urban area of

Muzaffargarh between the months of December, 2021.vegetables along their soil were collected from the top surface of soil 0-20 cm inside with the help of soil auger and kept in polythene bags. The water samples having capacity 500 mL was collected from irrigation sources and kept in plastic bottles. The collected soil, vegetable and water samples having capacity 0.5 kg,500 ml respectively was transported to institute of chemical sciences laboratory Gomal University DI Khan, Pakistan for physicochemical and HMs analysis.

Sample preparation

For heavy metal analysis, 1 g desiccated sample of each plant and soil was digested by 15 mL mixture ratio of (5:1:1) HNO_3, H_2SO_4 and $HClO_4$ at 80 °C to attain a clear solution (Rai *et al.,* 2019) . The digested solution was filtered through Whatman's filter paper and diluted to 25 mL flask with double deionized water. The physicochemical characteristics of soil such as pH, organic matter and electrical conductivity were measured by portable meter. Sewage water samples were taken from point source having capacity 500 mL in plastic bottles and treated with 5 mL conc. Nitric acid to minimize the bacterial degradation of HMs. Heavy metals were determined by atomic absorption spectrophotometer (AAS-7000).

Transfer factor (TF)

The heavy metal transfer factor is determined by calculating heavy metal contaminations in vegetable and soil. It is described with subsequent equation.

TF= C plant /C soil (Jolly *et al.,* 2013)

Daily metal ingestion (DMI)

The data regarding ingestion of vegetables in normal routine in adult and child was preliminary achieved from normal inspection. It is designated with subsequent equation.

DMI = C metal × C factor × D food ingestion / B mean weight (Rattan *et al.,* 2005)

Wherever $C_{\text{intake metal food}}$, C_{factor}, and $B_{\text{mean weight}}$ designate contaminated metal endorsement in vegetables, daily ingestion of vegetables, transformation element (0.0850) and normal body weight, respectively.

Health risk indices (HRI)

For the assessment of threat in the selected community the indicator health risk index is used. For this identification the data regarding daily ingestion of metal and oral reference dose is necessary. It is described by following mathematical form;

HRI = DIM/ Rfd (Kumar *et al.,* 2016)

Quality control and chemometric study

The whole chemicals used in entire research were brought from Merck Company of Germany. All the apparatus were completely rinsed with 10% $HNO_3$and finally with distilled water to avoid from contaminations. To make the sensitivity of instrument reliable the stock solutions were used at each interval of five samples.

Statistical analysis

The latest version of IBM SPSS software was used in various statistical tools like principal component analysis (PCA). Cluster analysis (CA) and correlation analysis to find out source apportionment of toxic metals.

Results and discussion

Physicochemical and HMs analysis of soil

The Physicochemical study of polluted soil is presented in Table 1 .The movement ability of toxic metals in soil is mainly affected by its parameters like pH, electrical conductivity (EC), and organic matter content (OM) (Edogbo *et al.,* 2020). The numerical value of pH and EC in soil is ranged 8.4 – 11.4, 396 – 510 (µs/cm) respectively. The percent value of organic matter ranged from (1.8 – 2.4) % in soil respectively. The level of HMs (Pb, Cr, Cd, Ni, Zn, Cu, Fe, Mn) concentration in soil ranged 1.96- 4.67, 1.56- 2.96, 1.64- 3.93, 1.67- 2.25, 1.09-2.34, 1.15-2.08, 1.37- 2.35, 1.16-1.59 (mg/Kg) respectively. The extreme value of pH and EC in soil indicates that it is not suitable for better crop production (Shahid *et al.,* 2021). The same trend of pH and EC in soil was observed by former researcher in their work (Ullah *et al.,* 2018).

Distribution of HMs in water and vegetables

The toxic level of heavy metal concentration in soil and wastewater is presented in Tables 1 and 2. The finding shows extreme disparity in concentrations of wholly examined metals in water and vegetables. The observed level of HMs (Pb, Cr, Cd, Ni, Zn, Cu, Fe, Mn) in wastewater was found 0.68-1.45, 9.15-15.37, 0.66-1.11, 0.33-0.77, 0.10-0.25, 0.12-0.20, 0.45-0.85, 0.14-0.35 mg/kg respectively. While the level of HMs in Spinacia oleracea , Brassica oleraceae ,Raphanus sativus, Lactuca sativa and Brassica rapa subsp.rapa is observed 4.76-15.34 (Pb) , 5.42-13.65 (Cr) , 6.24-16.75 (Cd) , 10-16.45 (Ni) ,7.86-23 (Cu) , 7.86-22 (Zn) , 10.50- 27.50 (Fe) and 2.13-22.75 (Mn) mg/Kg respectively (Fig. 1).

The sequence of HMs in vegetables was found Fe > Cu > Mn > Zn > Cd > Ni > Pb > Cr respectively. Maximum value of Fe (22.75) mg/Kg was observed in spinach. The extreme value of iron can cause stomach ulcer. The results of current work are compatible with the work done in Nigeria (Ali *et al.,* 2021).

Table 1. Physicochemical analysis and HMs analysis of soil.

Variables	Range	Minimum	Maximum	Mean	S.D
pH	3	8.4	11.4	9.9	2.12
EC (µs/cm)	114	396	510	453	80.61
OM %	0.6	1.8	2.4	2.1	0.42
Pb	2.71	1.96	4.67	2.99	1.21
Cr	1.4	1.56	2.96	2.17	0.62
Cd	2.29	1.64	3.93	2.52	0.89
Ni	0.58	1.67	2.25	1.95	0.25
Zn	1.25	1.09	2.34	1.48	0.51
Cu	0.93	1.15	2.08	1.44	0.37
Fe	0.98	1.37	2.35	1.73	0.39
Mn	0.43	1.16	1.59	1.36	0.15

Table 2. Heavy metals analysis of wastewater.

HMs	Range	Minimum	Maximum	Mean	S.D
Pb	0.77	0.68	1.45	1.21	0.31
Cr	6.22	9.15	15.37	12.05	2.54
Cd	0.45	0.66	1.11	0.95	0.17
Ni	0.44	0.33	0.77	0.52	0.16
Zn	0.15	0.1	0.25	0.16	0.06
Cu	0.08	0.12	0.2	0.17	0.03
Fe	0.4	0.45	0.85	0.65	0.15
Mn	0.21	0.14	0.35	0.25	0.08

Transfer factor (TF)

Heavy metal transfer factor (HMTF) mainly describes the buildup of toxic metals in plant. The buildup of trace metals in floras mostly depends on soil physicochemical characteristics like pH, texture, organic matter and moistness (Gupta *et al.,* 2019). The data regarding TF of selected vegetables is presented in Table 3 respectively. The sequence of TF in various vegetables was observed Zn > Fe > Cu > Mn > Ni > Cr > Cd > Pb respectively.

Table 3. Heavy metal transfer factor (HMTF) from soil to selected vegetables.

Vegetables	Pb	Cr	Cd	Ni	Zn	Cu	Fe	Mn
spinach	3.928	4.911	4.447	6.506	11.157	9.936	10.329	9.148
Cabbage	2.268	4.27	4.344	6.188	13.395	12.174	12.716	10.995
Radish	3.573	5.315	4.608	7.479	9.203	8.0538	12.069	10.76
Lettuce	3.985	4.435	3.06	6.608	8.915	10.854	9.563	11.447
Turnip	3.767	6.078	4.373	7.356	9.904	10.046	9.966	11.165

Table 4. Daily intake of metal (DIM) and health risk index (HRI) of vegetables.

Vegetables	Pb		Cr		Cd		Ni	
DIM	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Spinach	6.16E-03	7.08E-03	5.59E-03	6.43E-03	5.88E-03	6.76E-03	6.66E-03	7.66E-03
Cabbage	3.56E-03	4.09E-03	4.86E-03	5.59E-03	5.74E-03	6.60E-03	6.34E-03	7.28E-03
Radish	5.60E-03	6.44E-03	6.05E-03	6.96E-03	6.09E-03	7.00E-03	7.66E-03	8.80E-03
Lettuce	6.25E-03	7.19E-03	5.05E-03	5.81E-03	4.05E-03	4.65E-03	6.77E-03	7.78E-03
Turnip	5.91E-03	6.79E-03	6.92E-03	7.95E-03	5.78E-03	6.65E-03	7.53E-03	8.66E-03
HRI	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Spinach	1.54E+00	1.77E+00	3.73E-03	4.28E-03	5.88E+00	6.76E+00	3.33E-01	3.83E-01
Cabbage	8.90E-01	1.02E+00	3.24E-03	3.73E-03	5.74E+00	6.60E+00	3.17E-01	3.64E-01
Radish	1.40E+00	1.61E+00	4.03E-03	4.64E-03	6.09E+00	7.00E+00	3.83E-01	4.40E-01
Lettuce	1.56E+00	1.80E+00	3.37E-03	3.87E-03	4.05E+00	4.65E+00	3.38E-01	3.89E-01
Turnip	1.48E+00	1.70E+00	4.61E-03	5.30E-03	5.78E+00	6.65E+00	3.77E-01	4.33E-01
	Zn		Cu		Fe		Mn	
DIM	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Spinach	8.70E-03	1.00E-02	7.55E-03	8.68E-03	9.41E-03	1.08E-02	6.54E-03	7.51E-03
Cabbage	1.04E-02	1.20E-02	9.25E-03	1.06E-02	1.16E-02	1.33E-02	7.86E-03	9.03E-03
Radish	7.17E-03	8.25E-03	6.12E-03	7.03E-03	1.10E-02	1.26E-02	7.69E-03	8.84E-03
Lettuce	6.95E-03	7.99E-03	8.25E-03	9.48E-03	8.71E-03	1.00E-02	8.18E-03	9.40E-03
Turnip	7.72E-03	8.88E-03	7.63E-03	8.77E-03	9.08E-03	1.04E-02	7.98E-03	9.17E-03
HRI	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Spinach	2.90E-02	3.33E-02	1.89E-01	2.17E-01	1.34E+00	1.54E+00	1.98E-01	2.28E-01
Cabbage	3.48E-02	4.00E-02	2.31E-01	2.66E-01	1.65E+00	1.90E+00	2.38E-01	2.74E-01
Radish	2.39E-02	2.75E-02	1.53E-01	1.76E-01	1.57E+00	1.81E+00	2.33E-01	2.68E-01
Lettuce	2.32E-02	2.66E-02	2.06E-01	2.37E-01	1.24E+00	1.43E+00	2.48E-01	2.85E-01
Turnip	2.57E-02	2.96E-02	1.91E-01	2.19E-01	1.30E+00	1.49E+00	2.42E-01	2.78E-01

The highest TF value of (13.395) Zn is observed in cabbage. The same type of results obtained by earlier researcher in their work (Al-Hamarneh *et al.,* 2016).

Former investigators have pointed out that vegetables having capability to own extreme TF due to functioning of soil electrolyte in metal accumulation (Jolly, Islam and Akbar, 2013).

Correlation matrix

For the determination of correlation in different metals correlation analysis are being used (Zhang *et al.,* 2017). Table 6 represents the findings of Pearson correlation analysis. From the vegetable data it is observed that Zn is strongly correlated ($P < 0.01$) with Pb (0.518). The correlation level of Cu ($P < 0.01$) was strongly observed with Zn (0.785), while the correlation level (P < 0.01) of Fe and Mn was observed with Cu (0.662, 0.769) respectively.

Correlation analysis of water shows strong correlation of Ni with Cr(0.986) , Zn with Cr (0.989) at (P < 0.01) while the correlation level (P< 0.05) of Zn was strongly observed with Ni (0.957) respectively. Correlation analysis of soil shows strong correlation level (P < 0.05) of Zn with Pb (0.885). From the correlation matrix of water, soil and vegetables it is greatly observed that there is a strong degree of correlation with each other.

Table 5. Principal component analysis (PCA) of water and vegetables.

HMs	Initial Eigenvalues			Extraction Sums of Squared Loadings			Water	
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	PC-1	PC-2
Pb	5.025	62.812	62.812	5.025	62.812	62.812	-0.234	0.878
Cr	1.663	20.786	83.598	1.663	20.786	83.598	0.875	0.466
Cd	0.768	9.597	93.195				-0.309	-0.669
Ni	0.544	6.805	100				0.924	0.347
Zn	2.16E-16	2.70E-15	100				0.812	0.579
Cu	9.50E-17	1.19E-15	100				0.866	0.1
Fe	-2.32E-19	-2.90E-18	100				0.938	-0.178
Mn	-1.76E-16	-2.20E-15	100				0.424	0.72

HMs	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotated component matrix		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Pc-1	Pc-2	Pc-3
Pb	3.49	43.623	43.623	3.49	43.623	43.623	-0.4	0.71	-0.251
Cr	1.463	18.282	61.905	1.463	18.282	61.905	0.066	0.825	0.041
Cd	1.199	14.986	76.891	1.199	14.986	76.891	-0.175	-0.161	0.846
Ni	0.792	9.9	86.791				0.453	0.317	0.503
Zn	0.494	6.169	92.96				0.727	-0.482	-0.089
Cu	0.387	4.838	97.797				0.891	-0.151	-0.297
Fe	0.15	1.87	99.668				0.877	-0.171	0.294
Mn	0.027	0.332	100				0.909	0.147	-0.017

Table 6. Correlation analysis of water and vegetables.

HMs	Pb	Cr	Cd	Ni	Zn	Cu	Fe	Mn
Pb	1							
Cr	0.26	1						
Cd	-0.283	-0.516	1					
Ni	0.147	.986**	-0.409	1				
Zn	0.344	.989**	-0.617	.957*	1			
Cu	0.015	0.853	-0.307	0.847	0.794	1		
Fe	-0.454	0.708	-0.186	0.797	0.639	0.649	1	
Mn	0.413	0.658	-0.591	0.64	0.727	0.172	0.432	1

HMs	Pb	Cr	Cd	Ni	Zn	Cu	Fe	Mn
Pb	1							
Cr	0.382	1						
Cd	-0.155	-0.021	1					
Ni	-0.029	0.061	0.06	1				
Zn	-.518**	-0.274	-0.061	0.153	1			
Cu	-0.308	-0.077	-0.264	0.144	.785**	1		
Fe	-.562**	-0.058	0.088	.429*	.640**	.662**	1	
Mn	-0.268	0.149	-0.133	0.329	.456*	.815**	.769**	1

Cluster Analysis (CA) and Principal Component Analysis (PCA) of Heavy metals in selected vegetables irrigated with Wastewater

To examine the similarities and influence of toxic metals concentration principal component analysis (PCA) is being used. From the Table 5 it is observed that initial two principal components influence 76.891 % of the cumulative variance having Eigen value greater than one.

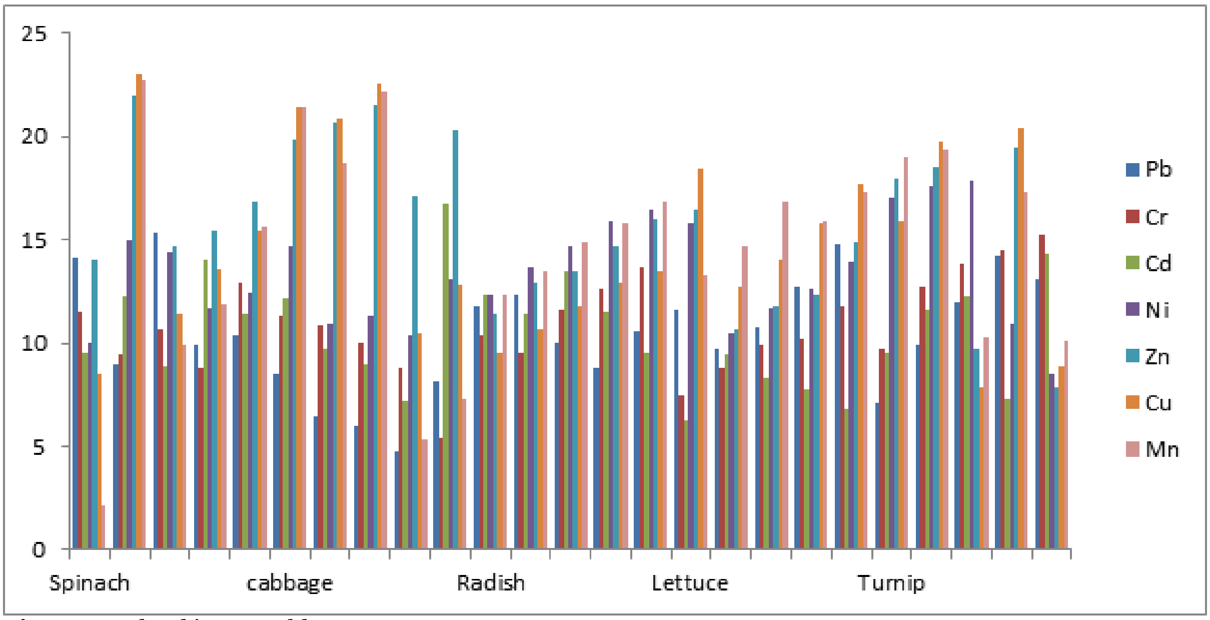

Fig. 1. HMs level in vegetables.

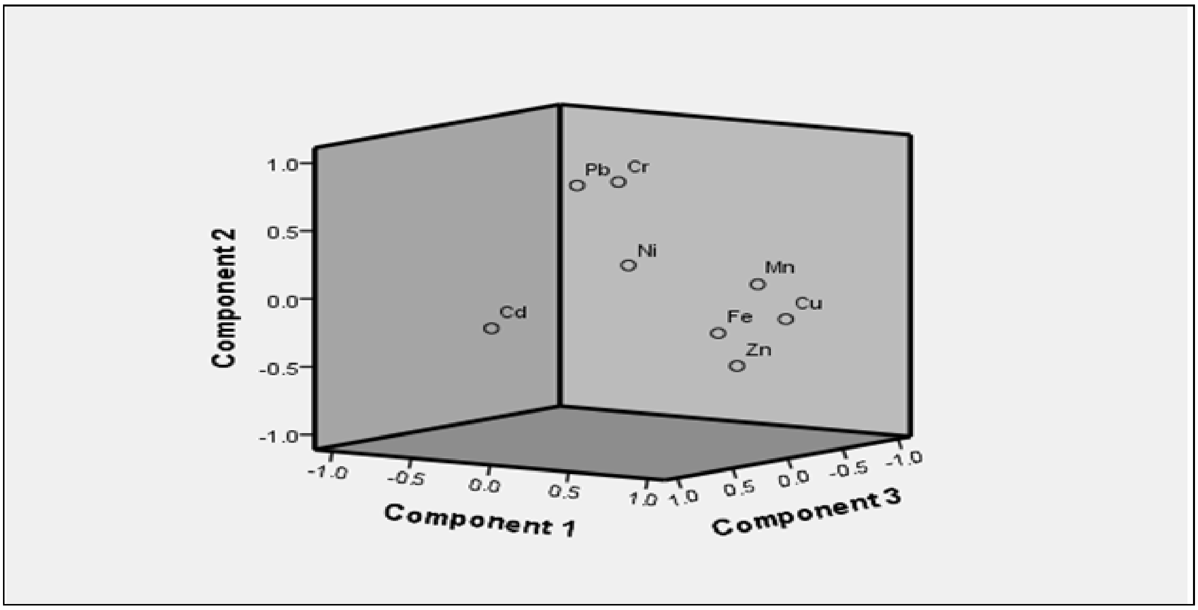

Fig. 2. PCA of vegetables.

The influence of PC-1 was greatly observed 43.623 % of cumulative variance having numerical value Cu (0.891), Zn (0.727), Fe (0.877) and Mn (0.909) respectively. The influence of PC-2 describes major loading of Cr (0.825) and Pb (0.710) with 18.282 % of the total variance respectively. While the influence of PC-3 shows major loading of Cd (0.846) having cumulative variance 14.986 % respectively. The findings of PCA of water (Fig. 3) show that the influence of pc-1 is 62.81% having major loading of Cr (0.875), Ni (0.924), Cu (0.866) and Fe (0.938) respectively.

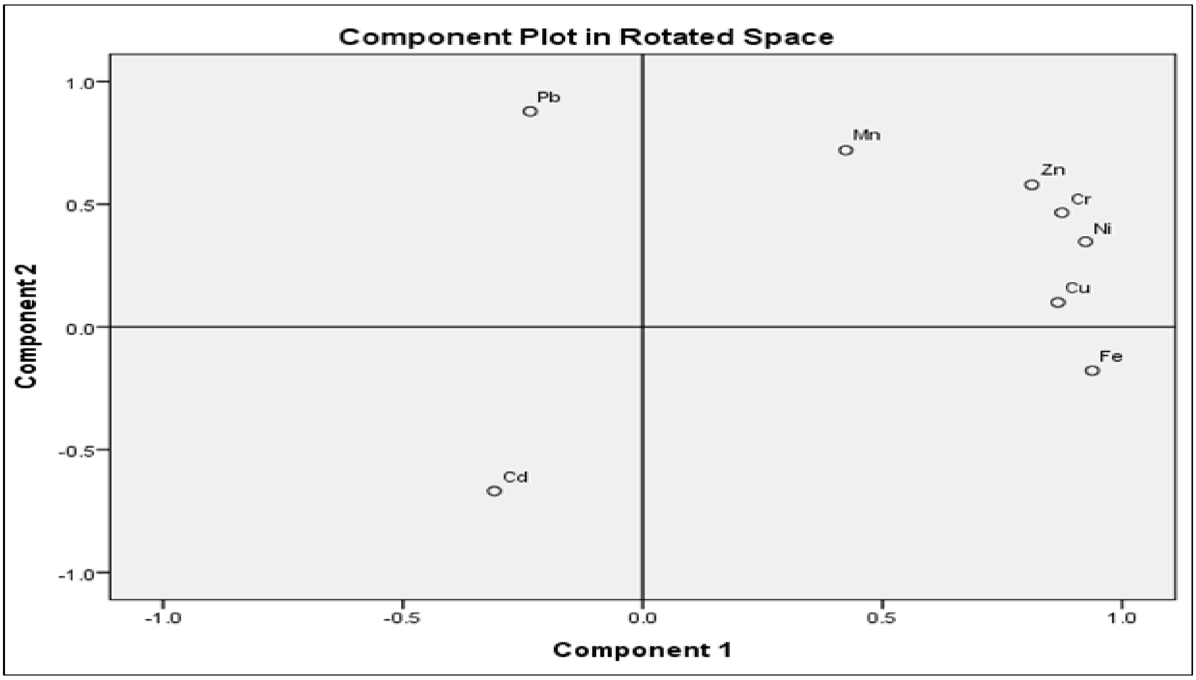

Fig. 3. PCA of wastewater.

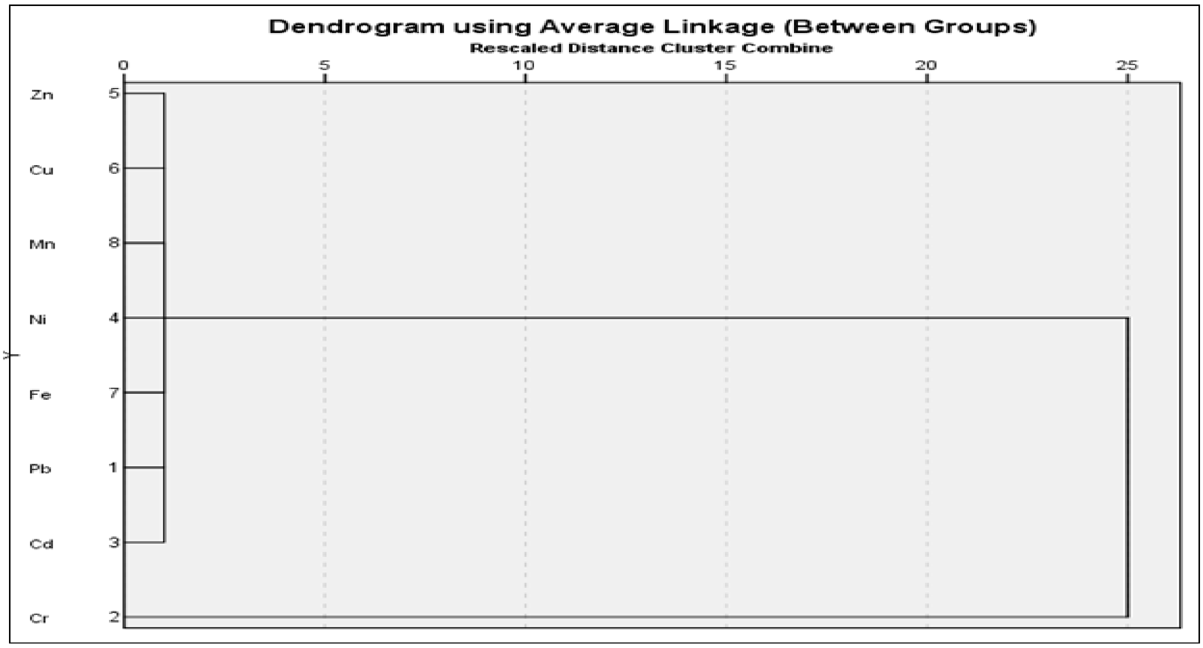

Fig. 4. Cluster analysis of water.

The influence of PC-2 is 20.786 % having major loading of Pb (0.878) respectively. PCA resolved the complex data of soil into three components. PC-1 having major loading of Pb (0.963) and Zn (0.969) shows major contribution with cumulative variance of 46.208% while PC-2, PC-3 having major loading of Ni (0.986) and Cd (0.987) shows cumulative variance of 31.079 % and 14.389 % respectively. From the PCA results (Fig. 2) it is observed that there is a major contribution of anthropogenic activities, automobile emission, and excessive usage of agrochemicals in urban area (Wang *et al.,* 2012).

Cluster analysis resolved the complex data of water into two groups' first group contain Ni and Cr while the second group contains Pb, Cd, Cu, Fe, Mn and Zn respectively. While the complex data of vegetables and soil is resolved into eight groups by cluster analysis. Dendrogram of water and soil are shown in Figures (4 & 5). From the findings of cluster analysis it is greatly observed that these groups have same source of contamination in their respective sites.

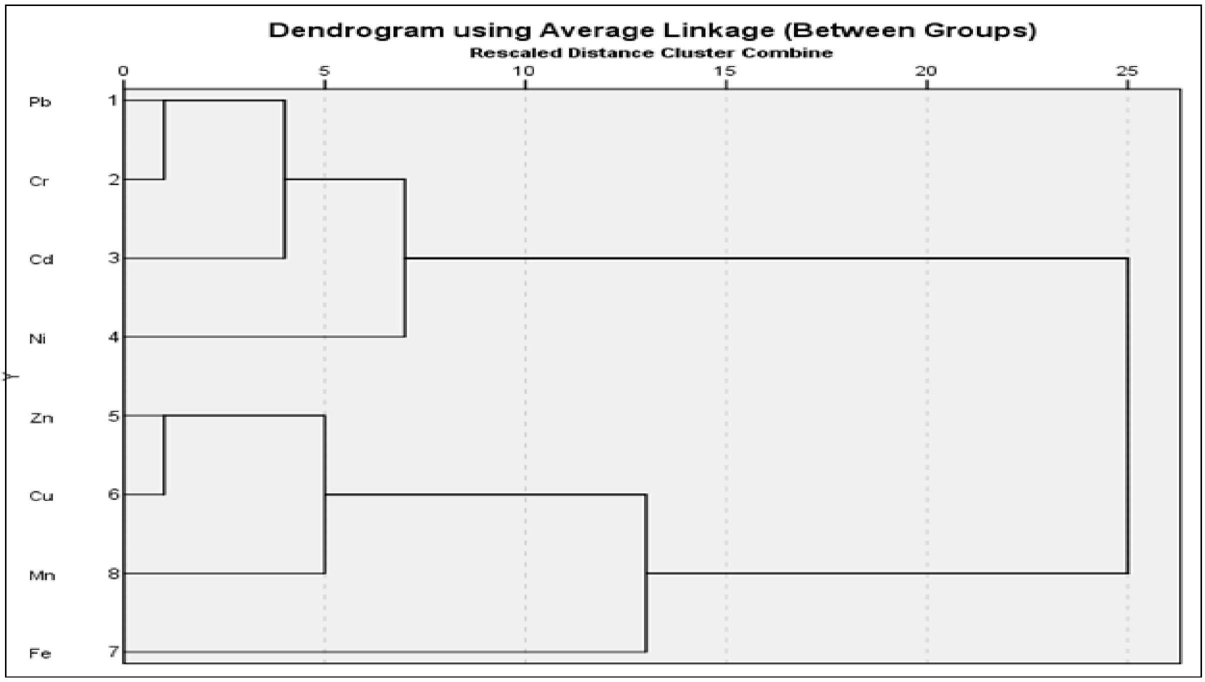

Fig. 5. Cluster analysis of vegetables.

DIM and HRI for vegetables

Table 4 describes the estimated daily metal intake and health risk indices associated with target vegetables. From the findings it is greatly observed that DIM value of HMs (Pb, Cr, Cd, Ni, Zn, Cu, Fe, Mn) in vegetables are in safe limit provided by WHO. While the HRI value of Pb, Cd and Fe exceed the recommended level in both adults and child provided by US-EPA in targeted vegetables. The findings of current work are compatible with work of earlier researcher done in peri-urban area of Swabi (Albedair and Alturiqi, 2021).

Conclusion

Vegetables are vital for health. Its safety mainly depends on irrigation channel. Extreme level of HMs (Pb, Cr, Cd, Zn, Ni, Cu, Fe, Mn) was observed in polluted soil, water and vegetables. The HRI level (in both adult and child) of Pb, Cd and Fe exceed the permissible limit in targeted vegetables. The heavy metal transfer factor (HMTF) from soil to vegetables was found higher for Zn (13.395) in cabbage. Vegetables grown in selected area are harmful to health. Therefore; it is recommended that cultivation of vegetables with sewage water in selected area should be stopped. There should be arranged an alternative channel for irrigation of vegetables. In this way we can minimize a threat associated with vegetables.

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