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SOIL - PLANT TRANSFER OF HEAVY METALS IN EIGHT WINTER VEGETABLES AND ITS IMPACT ON CONSUMER HEALTH RISK DUE TO THEIR DIETARY INTAKE

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ABSTRACT

This study was carried out to assess the consumer health risk due to intake of eight winter vegetables grown in farm soil around rural areas of Jwalapur, district Haridwar, Uttarakhand, India. The levels of cadmium, chromium, copper, iron, nickel, lead and zinc in farm soil and in edible parts of vegetables were determined using atomic absorption spectrometer. The order of accumulated metals in vegetables was zinc > iron > nickel > lead > cadmium > copper > chromium. The observed pattern of metal transfer (TF) from soil to edible parts of vegetables was zinc > iron > chromium > copper > lead > cadmium > nickel. The metal pollution index (MPI) values for all vegetables under study were found greater than unity (1.52 to 1.91). The daily intake of metals (DIM) was estimated to assess the human health risk index (HRI) due to consumption of these vegetables as dietary component. The overall HRI order was Cd (290-440) > Pb (72.5- 117.5) > Zn (16.33- 52.53) > Ni (17.0- 24.0) > Cu (5.25- 10.0) > Fe (0.33-0.70) > Cr (0.14 - 0.23).

KEY WORDS : Winter vegetables, Heavy metals, Soil - Plant Metal Transfer, MPI, DIM, HRI

INTRODUCTION

Intensive industrial operations, un-planned fast urbanization, modern agricultural practices and other anthropogenic activities have contaminated farm-soil and water bodies heavily that toxic metals and hazardous chemicals have accumulated through soil and water to food crops and vegetables and prolong consumption of which may affect human health adversely in various ways (Singh, 2005, Kulshrestha *et al.*, 2012). Further due to contamination of irrigating system, the toxic metals through agri-soil are incorporated to farm products and vegetables (Singh, 2005). Although, several useful metals in a very low level are required essentially by our body to operate numerous biochemical reactions and for various other functions, when the concentration of metals in human body exceeds their required levels the metal whether essential or not may be hazardous to the humanity (Kulshrestha *et al.*, 2012).

In south Asian countries vegetables are integral constituent of balanced diet. Varieties of seasonal vegetables and fruits fulfill the human requirements of essential micro-nutrients, vitamins, fiber, poly-phenols, sugars and antioxidants (Agrawal *et al.*, 2007; Kumar *et al.*, 2007). Most of the plants accumulate essential micronutrients and few other metals as per their metabolic needs. The accumulation of a metal in plants depends on its bio-availability in soil solution around the plant root system and soil pH which influence the metal bio-availability (Brown *et al.*, 1995). In addition to absorbing these micronutrients, certain plants selectively bio-concentrate much higher concentration of metals from contaminated soil (Hussain *et al.*, 2019). Due to increasing contamination of soil and use of waste water for irrigation, toxic metals from agri-soil are accumulated in edible parts of vegetables and other crops (Singh *et al.*, 2010) and regular consumption of such vegetables may lead to several health problems (Sharma *et al.*, 2006; Arora *et al.*, 2008).

The present investigation describes the

assessment of human health risk (HRI) due to consumption of eight winter vegetables by estimating the accumulated Cd, Cr, Cu, Fe, Ni, Pb and Zn in edible parts of the *Solanum melongena*, *Raphanus sativus*, *Daucus carota*, *Solanum tuberosum, Brassica rapa, Colocasia esculenta, Brasssica oleracea var. capitata* and *Brassica oleracea var. botrytis*. The soil samples were also analyzed for these metals. The soil -plant metal transfer factors (TF) and metal pollution index (MPI) for these vegetables were evaluated. The daily intakes of metals (DIM) on consuming vegetables under study were calculated to assess the health risk index (HRI).

METHODS AND MATERIALS

Study area

The vegetables under study were grown in agri-fields around Jwalapur areas in Haridwar district, Uttarakhand, India. The study area consists of rural and sub-urban settlements without waste water disposal facilities, that directly affect the water and soil quality (Kulshrestha *et al*., 2012).

Industrial units like rice mills, oil extraction plants, paper - pulp, gur - khand, brick kilns, stone crushers, etc are the main sources of toxic metals contamination (Kulshrestha *et al*., 2012). Farmers of the Hardwar district including study area produce most of the seasonal vegetables for domestic and commercial purpose that fulfills the requirements of adjoining areas of Dehradun district and other places of Garhwal region of Uttarakhand. Farmers of the study area usually use water from dug wells or hand pumps to irrigate their crops, while, many of these also use waste water for irrigation (Kulshrestha *et al*., 2012).

Collection of Farm Soil Samples

Collection of representative samples of farm soil were done randomly during October 2016 from five sites of the study area from a depth of 15- 20 cm and kept in separate clean zipped plastic bags. The samples were dried in open for 5-6 days and finally in electric oven (80^{0}C) for 3- 4 hours. The dried samples were finely grinded to homogenized fine particles and kept in clean and dry labeled bottles.

Collection of Vegetable Samples

Vegetable plants selected for metal uptake studies are presented in the Table 1. The collection of vegetable samples was done between November 2016 and February 2017. The matured samples of *Radish, Carrot, Potato, Turnip* and *Taro* plants were rooted out from the soil and their roots (tubers) were separated from the plants and cleaned separately 3-4 times with distilled water. Similarly, the matured samples of *Brinjal* fruit, *Cauliflower* head and *Cabbage* vegetative buds were collected and cleaned as above. All cleaned Vegetables were cut into small

Table 1. Descriptions of Vegetable Plants selected for study

S. N.	Botanical Name, Family	English Name	Parts used	Dietary importance
1.	*Solanum melongena,* Solanaceae	Brinjal	Fruit	Controls diabetes, has vitamin K, fiber, nutrients, anti-oxidants, vasodilators.
2.	*Raphanus sativus*, Brassicaceae	Radish	Tuber	Has vitamins E, A, C and B_6, anti-oxidants, fiber, Zn, K, P, Mg, Cu, Ca, Fe and Mn.
3.	*Ductus carrotus*, Apiaceae	Carrot	Tuber	Has fiber, ß Carotene, vitamin K, and anti-oxidants, lowers cholesterol.
4.	*Solanum tuberosum*, Solanaceae	Potato	Tuber	Has fiber, vitamins K, C, B_6, and Glyco-alkaloids, lowers Cholesterol in blood.
5.	*Brassica rapa,* Brassicaceae	Turnip	Tuber	Has fiber, vitamins K, A, C, E, B_6, Omega-3 acids, Mn, K, Mg, Fe, Ca, Cu and P.
6.	*Colocasia esculenta,* Araceae	Taro root (Arbi)	Tuber	Has fiber, anti-oxidants, vitamin A, protects heart, controls diabetes, immune booster.
7.	*Brasssica oleracea var. capitata*, Brassicaceae	Cabbage	Vegetative bud	Has vitamins C and K, minerals, immunity booster, reduces cataract, keeps heart healthy and lowers cholesterol level.
8.	*Brassica oleracea var. botrytis*, Brassicaceae	Cauli-flower	Head	Prevents cancer, rich in fiber, Folic acid Carotenoids, Glucosinolates, Thiamine, Riboflavin, Niacin and Pantothenic acid.

pieces and spread on filter paper sheets to air dry for 3-4 days in dust free chamber, finally oven dried for 4-5 hours at 60- 65^{0}C. The dried plant samples were grinded to powder and kept in labeled sample bottles.

Treatment of Vegetables and Soil Samples

To determine the accumulated metals in vegetables, grinded samples (1.0g) were subjected to HNO_3 – $HClO_4$ (5:1, v/v) digestion at nearly 80^{0}C for several hours following the standard methods (APHA, 2005) to get a transparent light colored liquid, which was then filtered in 100 mL volumetric flasks and filled up to the mark with double distilled water. For estimation of metal concentration in soil samples, 1.0 g of the finely grinded soil samples were digested with HNO_3 –HCl–$HClO_4$ mixture (5:1:1) for several hours to get transparent extract which was filtered and diluted to a volume of 100 m with double distilled water (Brown *et al*, 1995; Jones, 2001).

Estimation of Metals in Soil and Vegetable Samples

The concentration (mg/kg, dw) of cadmium, chromium, copper, iron, nickel, lead and zinc in the samples of soil and edible parts of vegetables were determined on Atomic Absorption Spectrometer, *Analyst 200* (Perkin Elmer), using air-acetylene flame, in accordance to standard methods (Jones, 2001; APHA, 2005). The standard stock solutions of metal ions used were from *Sigma Aldrich*, which were diluted to required concentrations for preparing working standards. Other reagents, chemicals, and solvents used were of analytical grade (Kulshrestha *et al*, 2012). Doubly- distilled water was used for all purposes. All the estimations were run in triplicate.

Soil - Plant Metal Transfer Factor (TF)

Soil - plant metal transfer factor (TF) quantifies the degree of possible risks and hazards associated with the intake of heavy metals in edible parts of the vegetables grown on contaminated soil (Jan *et al*, 2010) or due to regular use of waste water for irrigation (Liu *et al*., 2006). The TFs for individual vegetable was calculated as the ratio of the concentration of heavy metals in vegetables and in the concerned soil as dry weight (Usero *et al*., 1997).

$TF = C_{plant} / C_{soil}$ … (1), where,

$\mathbf{C}_{plant}$ and $\mathbf{C}_{soil}$ signify heavy metal concentration in plant and soil on dry weight basis.

Metal Pollution Index (MPI)

Metal pollution index (MPI) signifies the overall level of metals in edible parts of a vegetable. MPI is calculated as the geometric mean of concentration of tested metals accumulated in edible parts of vegetable under study and obtained by putting these values in following equation (Usero *et al.* 1997).

MPI (mg/ Kg) = $(Cf_1 \times Cf_2 \times Cf_3 \times Cf_4 …….. Cf_n)^{1/n}$ (2), where,

Cf_n represents the concentration of nth heavy metal in edible parts of vegetable.

Consumers Health Risk due to Vegetables Intake

The assessment of human health risk due to intake of vegetables in diet is usually done in terms of health risk index (HRI) (US -EPA. 1989), which is computed as the ratio of the daily intake of metals

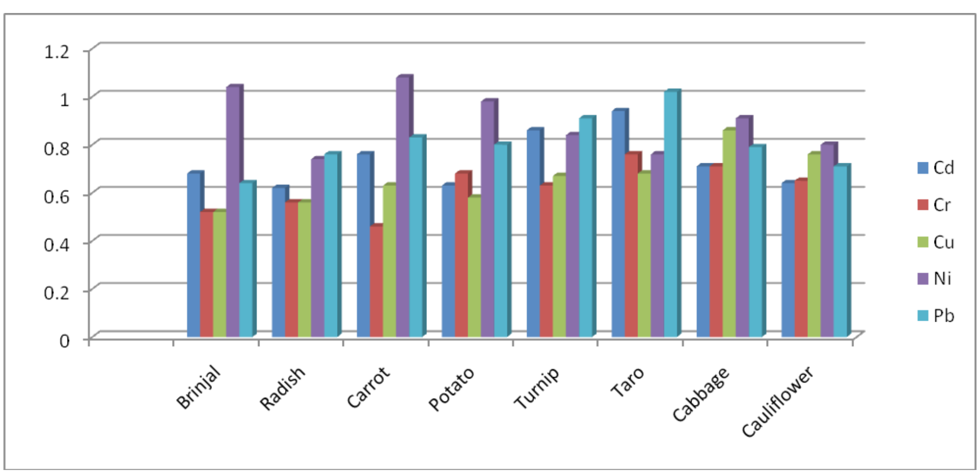

Fig. 1 (a). Accumulation of Cd, Cr, Cu, Ni and Pb in edible parts of vegetables

(DIM) due to consumption of metal contaminated vegetables under study and oral reference dose (US –EPA, 2013) by using the relation (3):

HRI = Daily Intake of Metals (DIM) / Reference Oral Dose ($R_f D$) .. (3)

The daily intake of metals (DIM) is evaluated by putting appropriate values in following relation (Chary *et al.*, 2008).

$$DIM = C_{Metal} \times C_{Factor} \times D_{Food\ Intake} / B_{Average\ Body\ Weight} \quad (4)$$

In the above relation, C_{Metal}, C_{Factor}, $D_{Food\ Intake}$ and $B_{Average\ Body\ Weight}$, signify metal concentration (mg/Kg) in edible parts of vegetable plant, conversion factor, daily intake of vegetables and average body weight of consumer, respectively. The C_{Factor} of 0.085 was used for conversion of fresh weight of vegetable to dry weight (Rattan *et al.,* 2005). The average daily intake of vegetables suggested by WHO guidelines in human adult diet is 300 to 350g per person (FAO/ WHO, 1989) and the average body weight of an adult consumer is suggested 60kg (FAO/ WHO, 1993). The oral reference dose of the metal (mg/kg/ day), Rf D (US –EPA, 1997; 2010; 2013; Jan *et al.,* 2010) is an approximation of daily tolerable exposure to which a person is expected to have without any significant risk of harmful effects during a life span (Kacholi *et al.,* 2018). The oral reference dose for cadmium, lead, copper, manganese, zinc, chromium and iron reported was 0.001, 0.004, 0.04, 0.14, 0.3, 1.5 and 15 mg/kg/day, respectively (US –EPA,1997) and for Ni was 0.02 mg/Kg/day (US- EPA, 2010).

RESULTS AND DISCUSSION

Table 2 presents the concentration of cadmium, chromium, copper, iron, nickel, lead and zinc recorded in farm soil samples as mg/Kg, dry weight of soil and in vegetable samples of *Solanum melongena*, *Raphanus sativus*, *Daucus carota*, *Solanum tuberosum*, *Brassica rapa*, *Colocasia esculenta*, *Brasssica oleracea var. capitata* and *Brassica oleracea var. botrytis* as mg/Kg, dw. The metal concentrations recorded in soil and vegetable samples under study were compared with the recommended safe limits (Awasthi, 2000; WHO, 2007).

Heavy Metals Concentrations in Farm Soil

In farm soil the concentrations of metals was, Zn (16.40±2.45) > Fe (14.59±2.67) > Ni (1.98 ±0.084) > Pb (1.72±0.042), Cd (1.69±0. 0.037 > Cu (1.36±0.038) > Cr (1.28±0.069 mg/Kg, dw). All the values were much below the recommended safe limits for soil (Awasthi, 2000; WHO, 2007).

Accumulated Metals in Vegetables

The metal accumulating potential of a plant depends on the concerned metal, its bio-availability in soil solution around the plant root system, plant species, growth stages, types of soil and soil pH (Chang *et al.*, 1984). Heavy metal transfer from farm-soil to different parts of vegetables differs greatly due to variation in metal uptake potentials of the plant as well as their accumulation in tubers and translocation to the above ground parts (Arora *et al.*, 2008). The diverse nature of metal accumulation in edible parts of vegetables could be attributed to their diverse morphological characteristics (Khan, 2013). The accumulated levels of cadmium (Table 2, Fig. 1) in different vegetables was: 0.94±0.051 (*Taro*) > 0.86±0.053 (*Turnip*) > 0.76±0.026 (*Carrot*) > 0.71±0.038 (*Cabbage*) >0.68±0.042 (*Brinjal*)

Table 2. Metal Concentrations (mg/kg on dry weight basis) found in Farm Soil and Vegetables

Metals →	Cd	Cr	Cu	Fe	Ni	Pb	Zn
Farm Soil	1.69±0.037	1.28±0.069	1.36±0.038	14.59±2.67	1.98±0.084	1.72±0.042	16.40±2.45
(Safe limits*)	(3.0-6.0)	——	(135-270)	——	(75-150)	(250-500)	(300-600)
Vegetables							
S. melongena	0.68±0.042	0.52±0.026	0.62±0.038	12.42±0.23	1.04±0.055	0.64±0.026	11.63±0.17
R. sativus	0.62±0.072	0.56±0.035	0.46±0.052	11.62±0.24	0.74±0.046	0.76±0.026	16.54±0.81
D. carrotus	0.76±0.026	0.46±0.043	0.63±0.038	10.82±0.12	1.08±0.043	0.83±0.055	10.65±0.13
S. tuberosum	0.63±0.052	0.68±0.043	0.58±0.051	14.22±0.90	0.98±0.060	0.80±0.052	21.44±0.93
B. rapa	0.86±0.053	0.63±0.060	0.67±0.043	21.71±0.97	0.84±0.051	0.91±0.053	15.16±0.94
C. esculenta	0.94±0.051	0.76±0.038	0.68±0.051	11.48±0.64	0.76±0.045	1.02±0.051	15.65±0.95
B. oleracea var. capitata	0.71±0.038	0.70±0.034	0.86±0.034	13.24±0.88	0.91±0.061	0.79±0.053	18.86±0.97
B. oleracea var. botrytis	0.64±0.034	0.65±0.043	0.76±0.069	11.64±0.61	0.80±0.051	0.71±0.061	34.24±1.03
(Safe limits*)	(1.5)	(5.0)	(30)	-	(1.5)	(2.5)	(50)

* Awasthi, 2000; WHO, 2007

>0.64±0.034 (*Cauliflower*) >0.63±0.052 (*Potato)* > 0.62±0.072 (*Radish*). Among the studied vegetables *Taro* and *Turnip* recorded comparatively higher levels of cadmium, yet in all samples cadmium was below the safe limit for vegetables (Awasthi, 2000; WHO, 2007). Cadmium is not essential for plant growth and when accumulated in agri- soil it may harm friendly soil microorganisms affecting plant growth. Cadmium exerts adverse effects when deposited in liver and kidney through food chain (Singh S. *et al.*, 2012). Chromium is essential micronutrients for plant growth (Fernando *et al*, 2012) and its levels in all the tested vegetables were within the recommended safe limit that varied between 0.46±0.043 and 0.76±0.038, lowest in *Carrot* and highest in *Taro*. The accumulated order was: *Taro* (0.76±0.038) > *Cabbage* (0.70±0.034) > Potato (0.68±0.043) > *Cauliflower* (0.65±0.043) >*Turnip* (0.63±0.060) > *Radish* (0.56±0.035) > *Brinjal* (0.52±0.026) > *Carrot* (0.46±0.043). Copper is essential for plant growth (Ramtek, *et al,* 2016) and recorded the accumulation order in vegetables as *Cabbage* (0.86±0.034) > *Cauliflower* (0.76±0.069) > *Taro* (0.68±0.051) > *Turnip* (0.67±0.043) > *Carrot* (0.63±0.038) > *Brinjal* (0.62±0.038) > *Potato* (0.58±0.051) > *Radish* (0.46±0.052, mg/Kg, dw).

The accumulated order of iron in tested vegetables was *Turnip* (21.71±0.97) > *Potato* (14.22±0.90) >*Cabbage* (13.24±0.88) > *Brinjal* (12.42±0.23) > *Cauliflower* (11.64±0.61) > *Radish* (11.62 ±0.24) > *Taro* (11.48±0.64) > *Carrot* (10.82±0.12 mg/ Kg). In all vegetable samples nickel levels were within the permitted limits (Awasthi, 2000) and the order was *Carrot* (1.08±0.043) > *Brinjal* (1.04±0.055) > Potato (0.98±0.060) > *Cabbage* (0.91±0.061) > Turnip (0.84±0.051) > *Cauliflower* (0.80±0.051) > Taro (0.76±0.045) > *Radish* (0.74±0.046). Nickel is essential for the active synthesis of urease in plant cells which acts as a catalyst in the hydrolysis of urea to ammonia and carbamic acid (Cui *et al.*, 2004). The order of accumulated lead in vegetables (Table 2, Fig.1) was Taro (1.02±0.051) > *Turnip* (0.91±0.053) > Carrot (0.83±0.055) >*Potato* (0.80±0.052) > Cabbage (0.79±0.053) > *Radish* (0.76±0.026) > *Cauliflower* (0.71±0.061) > *Brinjal* (0.64±0.026 mg/Kg, dw. The level of lead in all the samples was much below the safe limit (Awasthi, 2000). The accumulated values of zinc in vegetables were below the recommended safe limit of 50 mg/Kg, dw (Awasthi, 2000). Zinc is needed for several metabolic activities and plant growth (Ramtek *et al.,* 2016). However, much high level of zinc may cause health problems (Paula *et al.*, 2016). The order of accumulated zinc in vegetables was Cauliflower (34.24±1.03) > *Potato* (21.44±0.93) > *Cabbage* (18.86±0.97) > *Radish* (16.54±0.81) > Taro (15.65±0.95) > *Turnip* (15.16±0.94) > *Brinjal* (11.63±0.17) > *Carrot* (10.65 ±0.13 mg/Kg, dw).

Statistical Analysis

To evaluate significant differences among the metals in agri-soil and in edible parts of vegetables, *one-way ANOVA* was computed at $p<0.01$ level. The ANOVA outputs for Cd, Cr, Cu, Ni, and Pb concentrations in soil and vegetables were significant at $p<0.01$. However, the ANOVA results for Fe and Zn were not found significant at $p<0.01$ or at $p<0.05$.

Soil - Plant Metal Transfer Factor (TF)

Soil to plant metal transfer factor (TF) for individual vegetable was calculated by putting the experimental values of $\mathbf{C}_{plant}$ and $\mathbf{C}_{soil}$ equation (1) (Usero *et al*,1997) mentioned earlier. The TF values for Cd, Cr, Cu, Fe, Ni, Pb and Zn in tested vegetables are given in Table-3.

The order of TF (Table.3) was, Zn > Fe > Cr > Cu > Pb > Cd > Ni. The TF of Cd was high in *Taro* (0.56)

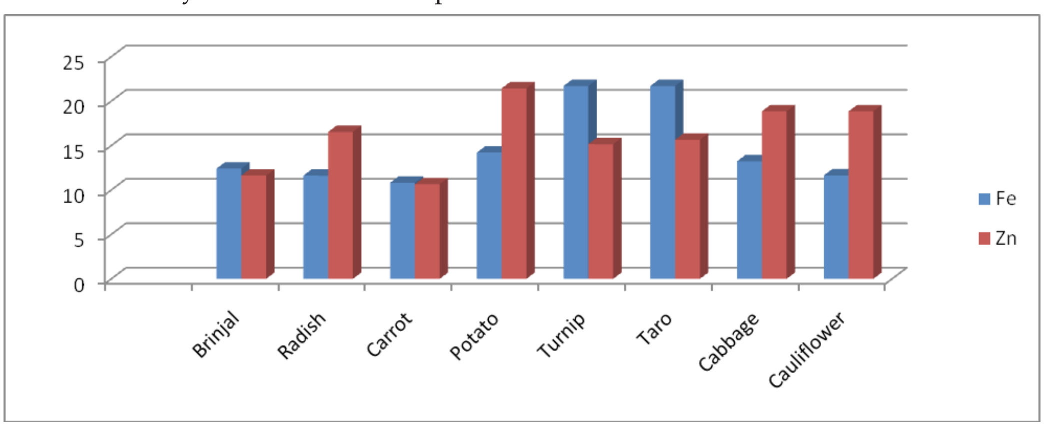

Fig. 1(b). Accumulation of Fe and Zn in edible parts of vegetables

Table 3. Soil - Plant Metal Transfer Factor (TF) of Vegetables

Vegetables/Metals	Cd	Cr	Cu	Fe	Ni	Pb	Zn
S. melongena	0.40	0.41	0.45	0.85	0.52	0.37	0.71
R. sativus	0.37	0.44	0.34	0.80	0.37	0.44	1.01
D. carrotus	0.45	0.36	0.46	0.74	0.54	0.48	0.65
S. tuberosum	0.37	0.53	0.43	0.97	0.49	0.46	1.31
B. rapa	0.51	0.49	0.49	1.49	0.42	0.53	0.92
C. esculenta	0.56	0.59	0.50	0.79	0.38	0.59	0.95
B. oleracea var. capitata	0.42	0.55	0.63	0.91	0.46	0.45	1.15
B. oleracea var. botrytis	0.38	0.51	0.56	0.80	0.40	0.41	2.09

and *Turnip* (0.51), while lowest in *Radish* and *Potato* (0.37), *Cauliflower* (0.38), *Brinjal* (0.40), *Cabbage* (0.42) and *Carrot* (0.45). Chromium exhibited high transfers in *Taro* (0.59), *Cabbage* (0.55), *Potato* (0.53), *Cauliflower* (0.51) and *turnip* (0.49) and lowest in *Carrot* (0.36), *Brinjal* (0.41) and *Radish* (0.44). The TF of Cu was highest in *Cabbage* (0.63), followed by *Cauliflower* (0.56), *Taro* (0.50) and *Turnip* (0.49) and lowest in *Radish* (0.34), and followed by *Potato* (0.43), *Brinjal* (0.45) and *Carrot* (0.46). Nickel showed high transfer in *Carrot* (0.54), followed by *Brinjal* (0.52), *Potato* (0.49) and *Cabbage* (0.46), while the lowest was in *Radish* (0.37), *Taro* (0.38) and followed by *Cauliflower* (0.40) and *Turnip* (0.42). Iron exhibited high TF in all the vegetables, highest being in *Turnip* (1.49), followed by *Potato* (0.97), *Cabbage* (0.91), *Brinjal* (0.85) and the lowest was in *Carrot* (0.74), followed by *Taro* (0.79) and *Radish* (0.80). Zinc recorded high TF in *Cauliflower* (2.09), followed by *Potato* (1.31), *Cabbage* (1.15), *Radish* (1.01), *Taro* (0.95) and *Turnip* (0.92), while the lowest was in Carrot (0.65) and Brinjal (0.71). The higher value of the TF (> 0.50) indicated the elevated status of metal in the concerned vegetable. The variations in metal transfer factors among different vegetables may be attributed to the differences in the concentration and bioavailability of metals in the farm soil (Chauhan and Chauhan, 2014) and metal uptake potential of vegetables (Cui *et al*, 2004).

Status of Metal Pollution Index (MPI)

The values of MPI were obtained by putting the metal concentrations found in vegetables, in equation (2) mentioned earlier and the results are presented in the Table 4.

$$\text{MPI (mg/ Kg)} = (Cf_1 \times Cf_2 \times Cf_3 \times Cf_4 \ldots\ldots Cf_n)^{1/n} \quad \ldots\ldots\ldots\ldots(2)$$

MPI is a reliable method for metal pollution monitoring (Samuel *et al*, 2018) and signifies the overall metal concentrations in vegetables and the possible quantum of risk to consumers (Usero *et al*, 2006; Liu *et al*. 2006; Wang *et al*. 2005). The MPI between 1 and 2 is moderate heavy metal pollution (Rashed, 2010). The computed MPI order for vegetables was *Turnip* (1.91) > *Cabbage* (1.85) > *Cauliflower* (1.84) > *Taro* (1.82) > *Potato* (1.79) > *Carrot* (1.56) > *Brinjal* (1.55) > *Radish* (1.52 mg/Kg). This order confirmed the high accumulation of tested metals in the edible parts of the vegetables and the consumer health risk may aggravate further (Ghosh, *et al*, 2013).

Assessment of Consumer Health risk Index (HRI)

(i) The daily intake of metals (DIM) was assessed by putting values of observed metal concentrations in vegetables for C_{Metal}, 0.085 for C_{Factor} and 325g for $D_{Food\ Intake}$ of an adult consumer in relation (3). The value of $D_{Food\ Intake}$ taken in this study was the mean of the WHO suggested values (300 to 350g / adult consumer) (FAO/ WHO, 1989).

$$DIM = C_{Metal} \times C_{Factor} \times D_{Food\ Intake} / B_{Average\ Body\ Weight} \quad ..(3)$$

The obtained results of daily intake of metals (DIM) are presented in Table 5.

The values of DIM depends upon the properties

Table 4. Metal pollution index (MPI) due to Cd, Cr, Cu, Fe, Ni, Pb and Zn in vegetables

S. N.	Vegetable	MPI (mg/Kg)	S. N.	Vegetable	MPI (mg/Kg)
1.	*S. melongena*	1.55	5.	*B. rapa*	1.91
2.	*R. sativus*	1.52	6.	*C. esculenta*	1.82
3.	*D. carrotus*	1.56	7.	*B. oleracea var. capitata*	1.85
4.	*S. tuberosum*	1.79	8.	*B. oleracea var. botrytis*	1.84

of soil, level of concerned metal in soil, its bioavailability and vegetable species (Kumar *et al*, 2017). The range DIM (Table 5) for Cd varied from 0.29 to 0.44, for Cr from 0.21 to 0.35, for Cu from 0.21 to 0.40, for Fe from 4.98 to 10.50, for Ni from 0.34 to 0.50, for Pb from 0.29 to 0.47 and for Zn varied from 4.90 to 15.76, mg/Kg /day/person.

(ii) The Health risk Index (HRI) values, for Cd, Cr, Cu, Fe, Ni, Pb and Zn accumulated in tested vegetables were computed as per equation (4) mentioned earlier and summarized in Table-6.

The degree of toxicity of heavy metals to consumer health depends upon their intake through vegetables (Adeel and Malik, 2014). If, HRI <1 the vegetable is assumed to be safe (USEPA IRIS., 2006), while, HRI >1 is unsafe for human health (U.S. EPA, 2002). The overall order of recorded HRI (Table 6, Fig. 2) was Cd (290-440) > Pb (72.5- 117.5) > Zn (16.33- 52.53) > Ni (17.0- 24.0) >Cu (5.25- 10.0) > Fe (0.33-0.70) > Cr (0.14 - 0.23) and are comparable with earlier results (Adeel and Malik, 2014; Ramteke *et al.*, 2016). Out of seven metals tested, Cd exhibited highest HRI values in all the vegetables that varied from 290 to 440, while all the HRI values for Cr were lowest and varied from 0.14 to 0.23. In *Brinjal*, the HRI order was Cd (300) > Pb (72.5) > Ni (24) > Zn (17.87) >Cu (7.25) > Fe (0.38) > Cr (0.16); in *Radish* the order was Cd (290) > Pb (87.5) > Zn (25.37) > Ni (17.0) > Cu (5.25) > Fe (0.36) >Cr (0.17) ; in *Carrot* the order was Cd (350) > Pb (95) > Ni (25) > Zn (16.33) > Cu (7.25) >Fe (0.33) > Cr(0.14). The HRI order in *Potato* was Cd (290) > Pb (92.5) > Zn (32.9) > Ni (22.5) > Cu (6.75) >Fe (0.44) > Cr(0.21); in *Turnip* the order was Cd (400) > Pb (105.0) > Zn (23.27) > Ni

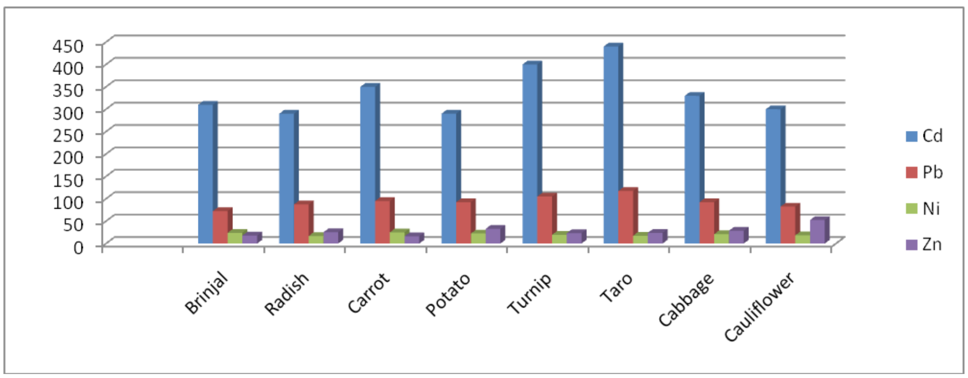

Fig. 2(a). Health risk index (HRI) due to intake of vegetables containing Cd, Pb, Ni and Zn.

Table 5. Daily intake of metals (DIM, mg/Kg/day/person) through vegetables consumption

Vegetables	Cd	Cr	Cu	Fe	Ni	Pb	Zn
S. melongena	0.31	0.24	0.29	5.72	0.48	0.29	5.36
R. sativus	0.29	0.26	0.21	5.35	0.34	0.35	7.61
D. carrotus	0.35	0.21	0.29	4.98	0.50	0.38	4.90
S. tuberosum	0.29	0.32	0.27	6.55	0.45	0.37	9.87
B. rapa	0.40	0.29	0.31	10.50	0.39	0.42	6.98
C. esculenta	0.44	0.35	0.32	5.29	0.35	0.47	7.21
B. oleracea var. capitata	0.33	0.33	0.40	6.10	0.42	0.37	8.63
B. oleracea var. botrytis	0.30	0.29	0.35	5.35	0.37	0.33	15.76

Table 6. The estimated health risk index (HRI) values for metals in vegetables

Vegetables, Metals	Cd	Cr	Cu	Fe	Ni	Pb	Zn
S. melongena	310	0.16	7.25	0.38	24.0	72.5	17.87
R. sativus	290	0.17	5.25	0.36	17.00	87.50	25.37
D. carrotus	350	0.14	7.25	0.33	25.00	95.00	16.33
S. tuberosum	290	0.21	6.75	0.44	22.50	92.50	32.90
B. rapa	400	0.19	7.75	0.70	19.50	105.00	23.27
C. esculenta	440	0.23	8.00	0.35	17.50	117.50	24.03
B. oleracea var. capitata	330	0.22	10.00	0.41	21.00	92.50	28.77
B. oleracea var. botrytis	300	0.19	8.75	0.36	18.50	82.50	52.53

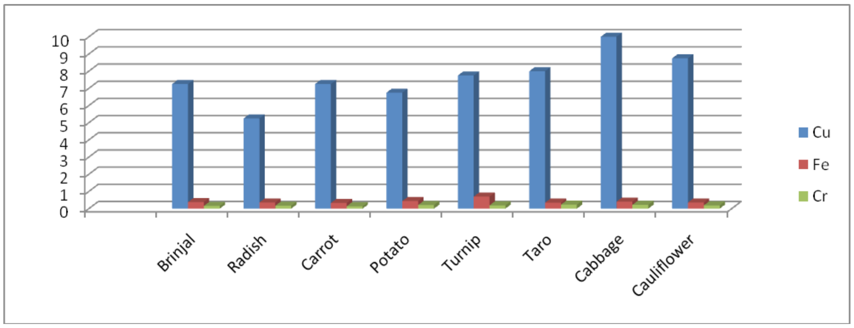

Fig. 2(b). Health risk index (HRI) due to intake of vegetables containing Cu, Fe and Cr.

(19.5) > Cu (7.75) >Fe (0.70) > Cr(0.19); in *Taro* the order was Cd (440) > Pb (117.5) > Zn (24.03) > Ni (17.5) > Cu (8.0) >Fe (0.35) > Cr (0.23); in *Cabbage the order was* Cd (330) > Pb (92.5) > Zn (28.77) > Ni (21.0) > Cu (10.0) >Fe (0.41) > Cr(0.22); and in *cauliflower* the order was Cd (300) > Pb (82.5) > Zn (52.53) > Ni (18.5) > Cu (8.75) >Fe (0.36) > Cr (0.19).

Among the studied vegetables the HRI due to Cd were much high (290-440) followed by Pb (72.5 – 117.5). Since, the recorded levels of Cd and Pb in vegetables varied from 0.62±0.072 to 0.94±0.051 and 0.64±0.026 to 1.02±0.051 mg/Kg, dw, respectively, such high values of HRI due to Cd and Pb may be partly due to much smaller RfD value for Cd (0.001) and Pb (0.004) in denominator of equation (3) used to evaluate HRI values, that gives high HRI values for Cd and Pb.

CONCLUSION

Vegetables are the most essential and integral part of our diet that is needed for proper growth of human body. Regular application of treated and untreated waste water may raise the levels of heavy metals to alarming point in farm soil; consequently these metals are accumulated in edible parts of vegetables during active growth period of the plants. This study revealed that vegetables grown in the study area accumulated Cd, Cr, Cu, Fe, Ni, Pb and Zn in significant amount due to contamination of farm soil for waste water used for irrigation. Such vegetables which have crossed the recommended safe limits for human consumption, when taken by the consumers in food diet are bound to result adverse health risks. Since, all the vegetables recorded MPI >1, these vegetables are unsafe for human consumption. The computed order of MPI for vegetables was Turnip (1.91) > Cabbage (1.85) > *Cauliflower* (1.84) > Taro (1.82) > *Potato* (1.79) > *Carrot* (1.56) > *Brinjal* (1.55) > *Radish* (1.52). Out of seven metals tested, Cd exhibited highest health risk index (HRI) in all the vegetables that varied from 290 to 440 and the lowest HRI was in all vegetables due to Cr which varied from 0.14 to 0.23. The contribution of metals in overall order of evaluated Health Risk Index (HRI) was Cd (290-440) > Pb (72.5- 117.5) > Zn (16.33- 52.53) > Ni (17.0- 24.0) > Cu (5.25- 10.00) > Fe (0.33-0.70) > Cr (0.14 - 0.23). On the basis of overall HRI, *Taro* and *Turnip* are to cause highest health risks, while Radish and *Potato* to cause the lowest health risk among studied vegetables. Therefore, regular evaluation of farm soils, water used for irrigation and cultivated vegetables for heavy metals are required, to prevent possible health risk to the consumers. Further, proper cleaning before cooking of vegetables should be a practice to avoid contamination of heavy metals through road side dust and vehicular exhaust and smoke from brick kiln and other industrial units.

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REFERENCES

Adeel, Mahmood and Malik, Riat Naseem, 2014. Human health risk assessment of heavy metals via consumption of contaminated vegetables collected from deferent irrigation sources in Lahore, Pakistan. *Arabian Journal of Chemistry*. 7 : 91-99.

Agarwal, S. B, Singh, Anita, Sharma, R. K. and Agrawal, M. 2007. Bioaccumulation of heavy metals in vegetables: A threat to human life. *Terrestrial and Aquatic Toxicology*. 1(2) : 13-22.

APHA (American Public Health Association). 2005. *Standard Methods for the Examination of Water and Wastewater*, 21st Ed, Washington, D.C.

Arora, Monu, Bala, Kiran, Rani, Shewta, Rani, Anchal, Kaur, Barinder and Mittal, Neeraj. 2008. Heavy Metal Accumulation in Vegetables irrigated with water from different sources. *Food Chem.* 111(4) : 811-815.

Awasthi, S. K. 2000. Prevention of Food Adulteration Act No 37 of 1954, Centre and State Rules as Ammended for 1999, 3rd Ed, Ashoka Law House, New Delhi.

Brown, S. L., Chaney, R. L., Angle, J. S. and Baker, A. M. 1995. Zinc and cadmium uptake by hyperaccumulator *Thlspi caerulescens* and metal tolerant *Silane vulgaris* grown on sludge amended soils, *Environ Sci Technol.* 29 : 1581- 1585.

Chang, A. C., Page, A. L., Warneke, J. E. and Grgurevic, E. 1984. Sequential extraction of soil heavy metals following a sludge Application. *J. Environ Qual.* 1 : 1133-1138.

Chary, N. S., Kamla, C. T. and Raj D. S. 2008. Assessment risk of heavy metals from consuming food grown on sewage irrigated soils and food chain transfer. *Ecotoxicology and Environment Safety.* 69 (3) : 513-524.

Chauhan, Geetanjali and Chauhan, U. K. 2014. Human health risk assessment of heavy metals via dietary intake of vegetables grown in wastewater irrigated area of Rewa, India, *International Journal of Scientific and Research Publications.* 4(9) : 1-9.

Cui, Y.J., Zhu, Y.G., Zhai, R. H., Chen, D.Y., Huang, Y. Z. and Qiu, Y. 2004. Transfer of metals from soil to vegetables in an area near a smelter in Nanning, China. *Environ Int.* 30 (6) : 785-791.

Fernando Guerra, Trevizam, Anderson Recardo, Muraoka Takashi, Mercante, Nericlenes Chaves, Canniaati-Brazaca and Solange Guidolin. 2012. Heavy metals in vegetables and potential risk for human health, *Sci. Agric.* 69(1) : 54-60.

Ghosh, Ratna, Xalxo, Reshma and Ghosh, Manik. 2013. Estimation of Heavy metals in vegetables from different market sites of tribal based Ranchi City through ICPOES and to Assess Health Risk, *Current World Environment.* 8 (3) : 435-444.

Hussain, Athar, Priyadarshi, Manjeet and Dubey, Shashikant. 2019. Experimental study on accumulation of heavy metals in vegetables irrigated with treated wastewater. *Applied Water Science.* Vol. 9, Article number: 122.

Jan, F. A. 2010. A Comparative Health Risk of Human Health Risks via Consumption of food crops grown on waste water irrigated soil (Peshawar) and relatively clean water irrigated soil (lower Dir) *Journal of Hazardous Materials.* 179 (1-3) : 612-21.

Jones, J. Benton, Jr. Laboratory Guide for Conducting Soil Tests and Plant Analysis; CRC Press, 2001.

Kacholi, David Sylvester and Sahu, Minati. 2018. Levels and Health Risk Assessment of Heavy Metals in Soil, Water, and Vegetables of Dar es Salaam. Tanzania, *Journal of Chemistry.* 2018 |Article ID 1402674

Khan, Muhammad Usman, Malik, Riffat Naseem and Said, Muhammad. 2013. Human health risk from heavy metal via food crops consumption with wastewater irrigation practices in Pakistan. *Chemosphere.* 93 (10) : 2230-2238

Kulshrestha, Shail, Awasthi, Alok and Dabral, S. K. 2012. Studies on the assessment of toxic metals present in Biological Samples (Part-1). *Int. J. Res. Chem. Environ.* 2 (1) : 195-199.

Kumar, Arvind, Seema and Kumar, Vipin. 2017. Human Health Risk of Heavy Metals in Vegetables grown in contaminated soil irrigated with Sewage Water. *American Journal of Food* Science and Nutrition. 4(4) : 23-35.

Kumar, Nirmal, J. I., Soni, Hiren and Kumar, R. N. 2007. Characterization of heavy metals from market vegetables using inductive coupled plasma analyzer. *Journal of Applied Sciences and Environmental Management.* 11 (3) : 75-79.

Liu, W., Li, X., Li, H. H. Sr. and Wan, Y. W. 2006. Heavy metal accumulation of edible vegetables cultivated in agricultural soil in the suburb of Zhengzhou City, People's Republic of China. Bulletin *of Environmental Contamination and Toxicology.* 76 : 163-170.

Paula, F. Ávila, Silva, Eduardo Ferreira da, Candeias, Carla. 2016. Health risk assessment through consumption of vegetables rich in heavy metals: the case study of the surrounding villages from Panasqueira mine, Central Portugal. *Environ Geochem Health.* 38(3) : 25 pages.

Ramteke, Shobhana, Sahu, Bharat Lal, Dahariya, Nohar Singh, Patel, Khageshwar Singh, Blazhev Borislav and Matini, Laurent, 2016. Heavy Metal Contamination of Vegetables. *Journal of Environmental Protection.* 7: 996-1004

Rashed, M. N. 2010. Monitoring of contaminated toxic and Heavy metals from mine tailings through age accumulation in some soil and weed plants in south east Egypt. *J. Hazard. Mater.* 178 : 739-746.

Rattan, R. K., Datta, S, P., Chhonkar, P. K., Sribabu, K., Singh, A. K. 2005. Long term impact of irrigation with sewage effluents on heavy metal contents in soils, crops and ground water- a case study. *Agric. Ecosyts. Environ.* 109 : 310-322.

Samuel, Teye Ametepey, Samuel, Jerry Cobbina, Felix, Jerry Akpabey, Abudu, Ballu Duwiejuah and Zita, Naangmenyele Abuntori, 2018. Health risk assessment and heavy metal contamination levels in vegetables from Tamale Metropolis, Ghana. *International Journal of Food Contamination.* 5 : 5.

Sharma, R. K., Agarwal, M. and Marshall, F. 2006. Heavy metal contamination of soil and vegetables in suburban areas of Varanasi, India, 2006. *Bulletin of Environmental Contamination and Toxicology.* 77 :

312-318.

Singh, A., Sharma, R. K., Agrawal, M. and Marshall, F. 2010. Health risk assessment of heavy metals via dietary intake of foodstuffs from the wastewater irrigated site of a dry tropical area of India. *Food Chem Toxicol.* 48 (2) : 611-19.

Singh, S., Zacharias, M., Kalpana, S. and Mishra, S. 2012. Heavy metals accumulation and distribution pattern in different vegetable crops. *Journal of Environmental Chemistry and Ecotoxicology.* 4(10): 170-177.

Singh, V. P. 2005. Toxic Metals and Environmental Issues, Sarup & Sons, New Delhi.

Technical Report, 1989. "Evaluation of certain food additives and Contaminants". Technical Report Series Geneva. 33rd Report of the Joint FAO/WHO Expert Committee on Food Additives. World Health Organization (WHO), Geneva, Switzerland.

Technical Report, 1993. "Evaluation of certain food additives and Contaminants". Technical Report Series Geneva. 41st Report of the Joint FAO/WHO Expert Committee on Food Additives. World Health Organization (WHO), Geneva, Switzerland.

US- EPA 1989d. Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part A). Washington, DC: US Environmental Protection Agency, Office of Emergency and Remedial Response; Report nr EPA/540/1-89/002.

US-EPA, 1997, Exposure Factors Handbook- General Factors. EPA/600/P-95/002Fa, Vol. I, Office of Research and Development. National Centre for Environmental Assessment. US Environmental Protection Agency, Washington DC.

US-EPA, 2002, Region 9 preliminary remediation goals. Washington, D.C.: U.S. Environmental Protection Agency; 2002.

US-EPA IRIS, 2006. United States, Environmental Protection Agency, Integrated Risk Information System. ^http://www.epa. gov/iris/substS>.

US - EPA, 2010. Risk-based Concentration Table. United State Environmental Protection Agency, Washington, DC.

US-EPA, 2013: Reference dose (R_fD): Description and use in health risk assessments, Background Document 1A, Integrated risk information system (IRIS); United States Environmental Protection Agency: Washington, DC, 15 March 2013; [http:// www epa.gov/iris/rfd.htm.]

Usero, J., Regalado, E. G. and Gracia, I. 1997.Trace metals in the bivalve mollusks Ruditapes decussatus and Ruditapesphillippinarum from the Atlantic Coast of Southern Spain. *Environ Int.* 23(3): 291-298.

Wang, X.L., Sato, T., Xing, B.S. and Tao, S. 2005. Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. *Science of the Total Environment.* 350 : 28-37.

WHO. 2007. Joint FAO/WHO Expert standards program codex Alimentation Commission. Geneva, Switzerland. [Available online http://www.who. int].