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Risk Assessment of Heavy Metals via Consumption of Contaminated Vegetables Collected from Different Agricultural Fields and Market Sites

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Abstract: The present study was conducted with an aim to compare heavy metals (Cu, Zn, Cd and Pb) accumulation potential of some of the commonly grown vegetables like brinjal, cauliflower, spinach and coriander collected from different agricultural (production) and market sites of Katihar city. The accumulation of Cd, Pb, Cu and Zn in test vegetables was higher in market sites then those at all agricultural lands and elevated by 47.84, 64.84, 21.3, and 9.91% in brinjal, 36.19, 78.09, 21.83, and 6.50% in cauliflower, 34.52, 49.50, 9.1, and 9.97% in spinach and 27.86, 47.05, 10.34, and 6.13% in coriander, respectively and was observed maximum in brinjal (143.89%) followed by cauliflower (142.61%), spinach (103.09%), and coriander (91.38%). The population load index (PLI), transfer factor (TF), daily intake of metals (DIM) and health risk index (HRI) were also studied. The maximum value of PLI was found for Zn (35.06%) and minimum for Pb (0.178%) in soil collected from production sites. The TF of heavy metals in vegetable collected from market sites was found to be higher than vegetable collected from production sites and could be one of the possible reasons for health risk in human via their consumption. The average daily intake of Cd, Pb, Cu and Zn, by adults in vegetables collected from market sites were 5.38, 1.20, 4.606 and 0.336% of provisional tolerable daily intake. The HRI value of all individual vegetables was below 1.0. Therefore, it is suggested that regular monitoring of heavy metals in vegetables is essential to prevent excessive build-up of heavy metals in the food chain and appropriate precautions should also be taken at the time of transportation and marketing of vegetables.

Keywords: Soil, Vegetables, Heavy Metals, Population Load Index, Transfer Factor, Daily Intake, Health Risk Index

1. Introduction

Vegetables are important part of human diet throughout the world, as they are rich sources of essential important nutrients, such as carbohydrates, proteins, vitamins, minerals, trace elements as well as antioxidants and metabolites, [1, 2]. They also play an important role to control various chronic diseases by acting as buffering media for acid substances formed during digestion of food [2]. The accumulation of heavy metals in plant vary with factors such as climate, soil properties, atmospheric deposition, plant species, and soil to plant factors of metals [3, 4]. *These toxic metals not only inhibit root growth but can also hamper physiological processes including uptake of nutrients. The absorption of heavy metals from the soil depends on different factors such as pH, organic matter, soil metal availability, cation exchange capacity, plant species, plant growth stages and presence of other metals present in soil [4].* The major sources of metals are wastewater untreated or partially treated industrial effluents, municipal wastes and vehicles [5, 6]. Aerosols also contribute to high levels of toxic metals on the surface of vegetables during the production, transportation, and marketing that depend upon various factors such as level of the pollutants, especially dust in air, nature of road, traffic loads and period of exposure or duration to which the vegetables are exported for marketing[7]. During harvesting, transportation and marketing of vegetables, emissions of heavy metals from industries and vehicles may be deposited on the surface of vegetables [8]. Atmospheric deposition can significantly elevate the levels of heavy metals in vegetables sold in the market were also reported by Sharma *et al.* (2008a, b) [9, 10].

Some metals such as Mn, Cu, Zn, Co, Mo and Fe are essential for human life in appropriate concentration as they catalyse enzymatic activities in human body, but in excess they become poisonous [11]. They get magnified with rising tropic level and ultimately get accumulated in human beings where they cause chronic and acute ailments and even death [12]. Some of these heavy metals form complexes with carboxylic (-COOH), amino (-NH_2), imino (>NH) and thiol (-SH) groups present in the proteins and they disturb the activity of the proteins to catalyse the function of enzymes [13]. The new biological complex molecules thus formed lose their function which result in break down or cell damage [13, 14]. Three heavy metals of greatest health concern are Cd, Pb and Hg. There is no biological need of any of them. The increase in environmental pollution caused by toxic metals is of great concern because of their carcinogenic properties, their non-biodegradability and bio-accumulation [15]. Continuous human consumption of contaminated vegetables may cause accumulation of heavy metals in the kidney and liver and can cause disturbance of biochemical process and cause cardiovascular, kidney, liver, nervous and bone diseases [16-18].

The assessment of heavy metal concentration in the vegetables from the market sites are being carried on in some developed countries and there are very few published data in India, however not available in the literature of Katihar city. Thus the aim of the research work was to monitor and assess the concentration of heavy metals (Cu, Zn, Cd and Pb) in some selected vegetables collected from different agricultural (production) fields and market sites of Katihar city, Bihar, India and also to estimate health hazard through their consumption.

2. Materials and Methods

2.1. Site Description

Katihar is an agricultural district that covers 3056 square km areas and located at 25.53°N and 87.58°E. Kathiar is too small but has large number of small scale industries. The wastewater and disposal of sewage water are drained to the agricultural land where these are used for growing crops and vegetables. Two study sites were selected i.e., market site (in between 0-5 km) and agricultural land (in between 5-15 km from the city centre). Freshly samples of some commonly grown vegetables i.e., brinjal (*Solanum melongena*), cauliflower (*Brassica oleracea* L.), spinach (*Spinacia oleracea* L.) and coriander (*Coriandrum sativum*) were collected simultaneously from the market sites and agricultural fields around Katihar city, Bihar, India during October 2014 to March 2015 (Table 1). The name and number of market sites and agricultural fields are listed in Table 2.

Table 1. *Green vegetable samples collected from agricultural fields and market sites.*

S. N	Common name	Vernacular name	Botanical name	Part used	Family
1	Brinjal	Baigan	*Solanum melongena*	Whole fruit	Solanaceae
2	Cauliflower	Phool Gobi	*Brassica oleracea* L	Fruity flowers	Brassicaceae
3	Spinach	Palak	*Spinacia oleraceae* L	Leaves	Amaranthaceae
4	Coriande	Dhania	*Coriandrum sativum*	Leaves	Apiaceae

Table 2. *Name and no. of market sites and agricultural (production) fields in the study area.*

Market sites		Agricultural (production) fields							
All Test Vegetable		Brinjal		Cauliflower		Spinach		Coriander	
Site name	No.	Site name	No.	Site name	No.	Site name	No.	Site name	No.
New Marker	I	Belwa	1	Varmali	8	Sarinda	15	Boodnagar	20
Bhagwan Chock	II	Govind Chock	2	Bastol	9	Sarbasa	16	Daheria	21
Chalisa Hatia	III	Hajipur	3	Ranipatra	10	Sonoli	17	Kuretha	22
Mirchaibari	IV	Sarifganj	4	Dalan	11	Haplaganj	18	Sernia	23
Mahamood Chock	V	Hawaiadda	5	Rampur	12	Chattabari	19	Pharthali	24
---------------------	----	Sirsa	6	Pranpur	13	------------	----	--------------	----
---------------------	----	Mania	7	Rhotra	14	------------	----	--------------	----

On the basis of the production capacity of these vegetables and transportation to the market, 24 sites were selected in the agricultural fields and five locations in the market (Figure 1). Among these agricultural fields 11 sites (3, 4, 5, 7, 10, 11, 12, 14, 21, 22 and 24) are located vicinity to the national highway (NH) and 7 sites (6, 13, 16, 17, 18, 19 and 23) are located close to brick kilns industries. Three locations (III, IV and V) in market are located in the dense populated area and heavy traffic on a narrow road and three (I, III and V) are also close to industrial, commercial and residential areas.

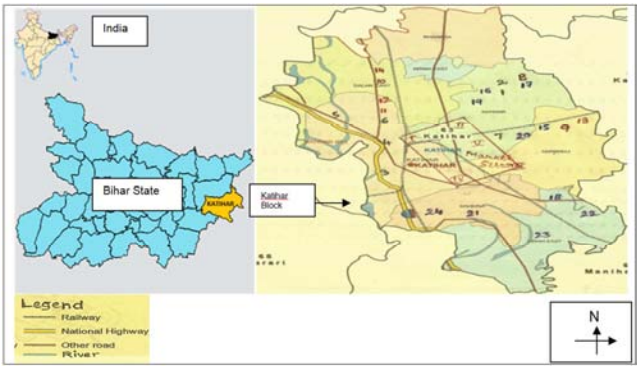

Figure 1. *Study area and location of sampling points.*

2.2. Soil Sampling

Soil samples were collected from 24 agricultural fields by digging a monolith of 10cm x 10cm x 10cm size by using plastic scooper. Each field was first subdivided into five parts, then all collected soil samples were mixed together to form composite soil sample from each field. Non-soil particles were removed from each sample, dried at 40°C for 48 h and ground into fine powder and then sieved through 2 mm nylon sieve. After this each sample was transferred into air tight polyethylene bag and brought into laboratory for analysis as described by Lei *et al*. (2008) [19].

2.3. Vegetable Sampling

The vegetables selected for heavy metal analysis were brinjal, cauliflower, spinach and coriander. Brinjal and cauliflower were collected from 7, 7 sites; spinach and coriander were collected from 5, 5 sites from agricultural fields respectively during the growing season and simultaneously from five (I, II, III, IV and V) market sites. All the collected vegetables (one intact inflorescence head of cauliflower and 1kg each of brinjal, spinach and coriander) were washed with tap water to remove the soil particles and then uneatable parts were removed. The edible part was sliced into pieces and dried separately on sheet of filter paper, then dried in oven at 75°C for 24 h and then crushed and sieved at room temperature and digested by using the method described by Jamail *et al.* (2009) [20].

2.4. Digestion of Samples

1gm of each sample of soil and vegetable was placed in 100 mL beaker separately and digested with 15 mL of tri-acid mixture i.e. HNO_3, $HClO_4$ and H_2SO_4 at 5:1:1 ratio at 80°C on an oven plate till the solution becomes transparent [21]. The solution thus obtained was filtered and each filtrate was made to 50 mL by mixing deionised water and subjected to atomic absorption spectrophotometer for analysis for heavy metal (Pb, Cd, Cu and Zn).

3. Data Analysis

3.1. Pollution Load Index (PLI)

The degree of soil pollution for each metal was computed as pollution load index by using following modified equation, (Liu *et al*., 2005) [22].

$$PLI = C_{Soil} / C_{Reference} \quad (1)$$

Where, C_{Soil} and $C_{Reference}$ represent the metal concentration in the wastewater irrigated and reference soil respectively.

3.2. Transfer Factor (TF)

The metal transfer factor from soil to plant was determined by using following formula, (Cui *et al*., 2005) [23].

$$TF = C_{Plant} / C_{Soil} \quad (2)$$

Where, C_{plant} and C_{soil} represent the metal concentration in plant fresh weigh and in soil dry weigh respectively.

3.3. Daily Intake of Metals (DIM)

The daily intake of heavy metal was calculated by using following formula, (Chary *et al*., 2008) [24].

$$DIM = C_{metal} \times D_{food\ intake} \times C_{factor} / B_{average\ body\ weight} \quad (3)$$

Where C_{metal}, $D_{food\ intake}$, $B_{average\ weight}$ and C_{factor} represent heavy metal concentration in plant (mg kg^{-1}), daily intake of vegetable (gm day^{-1} person $^{-1}$), average body weight (kg) and conversion factor (0.085) respectively. The conversion factor was used to convert fresh green vegetables weight to dry weight, as described by Rattan *et al*. (2005) [25]. The average body weight was considered to be 55 kg by conducting survey of 100 adult (male and female) people from study areas in each was determined by formal interview conducted with people of study areas during period of sampling. The average daily vegetable in take for adult was considered to be 250 gm day^{-1} person $^{-1}$ which was determined by formal interview conducted with people of study areas.

3.4. Health Risk Index (HRI)

The value of HRI depends upon the daily intake of metals (DIM) and reference oral dose (Rf_D), which was computed as described by Jan *et al*. (2010) [26]. The HRI less than 1 means exposed population said to be safe (US-EPA IRIS, 2006) [27]. Rf_D value for Cu, Pb, Cd, and Zn is 0.04, 0.004, 0.001 and 0.30 (mg/kg bw /day) respectively [27].

$$HRI = DIM / Rf_D \quad (4)$$

3.5. Statistical Analysis

All statistical analysis were performed on lenovo ™ computer using the Microsoft EXCEL and Word 2007 format. Similarly, the significance of differences between the concentrations of heavy metals in soil and vegetables were calculated by using Casio calculator (made in China) *fx-991 MS.* A probability of $p > 0.05$ was considered statically significant.

4. Results and Discussion

4.1. Accumulation of Heavy Metals in Soil Samples

Results revealed that concentration of heavy metals (µg/g) in test soil collected from different vegetable production fields ranged from 0.40 - 2.35 for Cd, 0.10 -1.57 for Pb, 10.2 – 24.3 for Cu and 80.2 -123.3 for Zn (Table 3). The upper limits of Cd, Cu and Zn in the test soil were higher than uncontaminated soil (Cd; 1.0, Cu; 15 and Zn; 100 µg/g) and lower for Pb (50.0 µg/g) as reported by Temmerman *et al.* (1984) [28]. But the upper limits of heavy metals were below the upper permissible limits as recommended by Prevention of food Adulteration (PFA) standards (6, 500, 270 and 300 µg/g, respectively for Cd, Pb, Cu and Zn) as guided by Awasthi (2000) [29]. Results also revealed that about 11 vegetable production sites, which were near to national highway and about 7 sites which were in the vicinity of brick kilns showed higher concentration of Cd, Pb, Cu and Zn than those agricultural fields which were free from special emission (Figure 2).

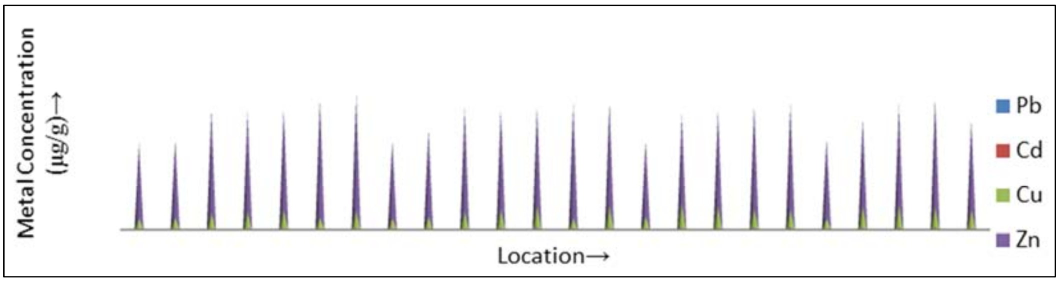

Figure 2. *Metal concentration in soil at 24 sites of Agricultural fields at study area.*

On the survey of study areas it was found that continuous irrigation of agricultural land with sewage and wastewater, use of Zn containing pesticides, Zn containing fertilizers and ash from brick kilns may be possible reasons of Zn accumulation in the soil [30, 31]. The percentage pollution load index (PLI) of each metal in soil at SWI zone compared with PFA standard (Awasthi, 2000) value was in the order of Cd (20%), Cu (6.18%), Pb (0.18%) and Zn (35.063%), (Table 3).

Table 3. *Accumulation of heavy metals (µg/g) in the soil from agricultural fields (n = 24).*

Statistical data	Pb	Cd	Cu	Zn
Minimum	0.10	0.40	10.2	80.20
Maximum	1.57	2.35	24.3	123.3
Mean	0.89	1.20	16.70	105.18
Median	1.42	1.07	16.30	113.8
Sum of total value	26.13	28.89	400.9	25.244
Simple Standard Deviation	0.494	0.561	4.648	13.815
Prevention of food Adulteration	500	6.00	270	300
Pollution Load Index	0.00178	0.20	0.06185	0.3506

4.2. Accumulation of Heavy Metals in Vegetable Samples

Results revealed that accumulation of heavy metals was found to be high in all vegetables collected from market sites compared to agricultural fields (Table 4). The maximum concentration of heavy metals (µg/g) were recorded at sites - 7 (1.25), 12 (1.35), 16 (3.1) and 24 (2.95) for Cd, at sites- 7(1.34), 14 (1.32), 17(1.47) and 22 (1.95) for Pb, at sites-5 (25.3), 12 (26.2), 16 (27.4) and 21 (24.5) for Cu, at sites- 7(130.3), 12 (142.7), 18(134.3) and 24 (134.6) for Zn in brinjal, cauliflower, spinach and coriander respectively, collected from agricultural fields as shown in Figures 3-18. The high accumulation of heavy metals in the test vegetables collected from agriculture fields at 5, 7, 12, 14, 21.22 and 24 may be due to their location near to national highway and sites 16, 17 and 18 may be due to being very close to brick kiln industries. The high concentration of heavy metals in the tested vegetables at sites 7, 12, 16 and 24 may also be ascribed due to their locations in the vicinity of brick kiln industries and also very close to national highway.

The concentration of Zn ranged from 95.2-140.1, 97.8-143.7, 99.5-147.3 and 95.3-141.4 µg/g in brinjal, cauliflower, spinach and coriander respectively collected from market sites (Table 4). In all vegetables collected from agricultural fields and market sites the accumulation of Zn was high followed by Cu, Cd and Pb than permissible limits of PFA standard (50µg/g). The maximum concentration of Cu in vegetables collected from market at site-V in cauliflower (37.7 µg/g) and in brinjal (35.4 µg/g), which were higher than permissible limit guided by PFA (30 µg/g), whereas minimum concentration of Cu was found to be 16.4 (µg/g) in brinjal at market site-I as shown in Figures 11-12.

Table 4. *Accumulation of heavy metals (µg/g) in the vegetables collected from production and market sites.*

Agricultural (production) Fields					Market Sites			
Metals	Brinjal	Cauliflower	Spinach	Coriander	Brinjal	Cauliflower	Spinach	Coriander
Cadmium								
Min.	0.45	0.45	0.86	0.82	0.58	0.68	0.98	0.98

Agricultural (production) Fields					Market Sites			
Metals	**Brinjal**	**Cauliflower**	**Spinach**	**Coriander**	**Brinjal**	**Cauliflower**	**Spinach**	**Coriander**
Max.	1.25	1.35	3.10	2.95	1.21	2.20	3.48	3.81
Median	0.95	1.01	2.21	2.12	1.21	1.24	2.71	2.71
Mean	0.786	0.909	1.802	1.888	1.162	1.238	2.424	2.414
σ_{n-1}	0.348	0.390	0.922	0.985	0.586	0.599	1.311	1.307
Σ_n	5.50	6.36	9.01	9.440	5.81	6.19	12.12	12.07
% Elevation	------	-------	-------	------	47.84	36.19	34.52	27.86
Lead								
Min.	0.20.	0.25	0.85	0.80	0.25	0.35	1.15	1.01
Max.	1.34	1.32	1.47	1.95	2.54	2.84	1.75	2.43
Median	1.25	1.25	1.47	1.95	2.03	1.85	1.75	1.66
Mean	1.041	1.05	1.01	1.05	1.716	1.87	1.51	1.544
σ_{n-1}	0.412	0.377	0.257	0.474	0.868	0.934	0.236	0.558
Σ_n	7.29	7.35	6.48	7.780	8.58	9.35	7.55	7.72
% Elevation	------	-------	-------	------	64.84	78.09	49.50	47.05
Copper								
Min.	15.3	15.8	17.5	14.8	16.4	16.7	19.7	17.3
Max.	25.3	26.2	27.4	24.5	35.4	37.7	28.5	26.3
Median	22.4	25.8	25.3	22.8	27.4	28.2	27.7	24.7
Mean	19.8	20.93	22.68	20.12	24.02	25.5	24.74	22.2
σ_{n-1}	3.612	4.121	4.047	3.885	7.658	8.09	3.66	3.575
Σ_n	138.6	146.5	113.4	100.6	120.1	127.5	123.7	111.0
% Elevation	------	------	-------	-------	21.31	21.83	9.10	10.34
Zinc								
Min.	90.3	92.2	93.1	94.8	95.2	97.8	99.5	95.3
Max.	130.3	142.7	134.3	134.6	140.1	143.7	147.3	141.4
Median	110.4	137.7	122.7	120.5	121.6	130.7	139.6	134.3
Mean	109.18	118.49	118.52	119.1	120.00	126.18	130.34	126.4
σ_{n-1}	15.656	22.62	16.926	16.28	19.509	19.494	20.76	19.29
Σ_n	764.3	829.4	592.6	595.5	600.00	630.9	651.7	630.7
% Elevation	------	-------	------	-------	9.91	6.50	9.97	6.13

Σn = sum of the total values and σ_{n-1} = sample standard deviation

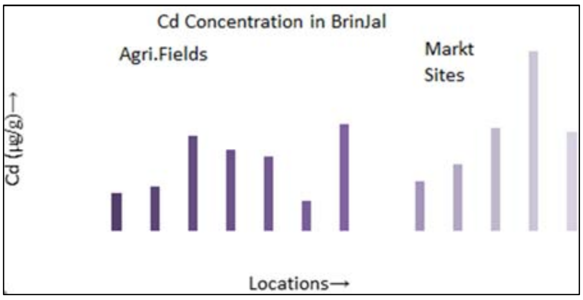

Figure 3. *Concentration of Cd in brinjal.*

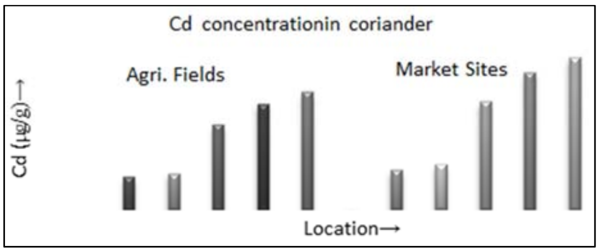

Figure 4. *Concentration of Cd in coriander.*

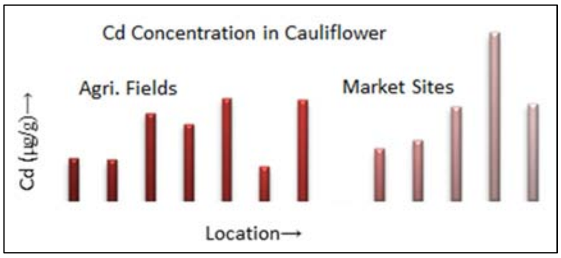

Figure 5. *Concentration of Cd in cauliflower.*

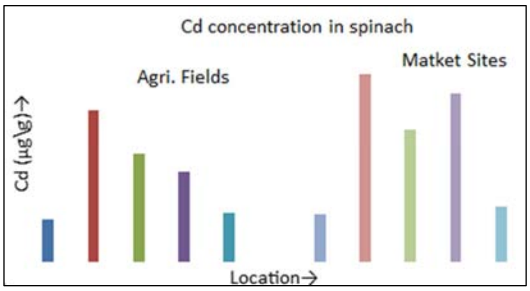

Figure 6. *Concentration of Cd in spinach.*

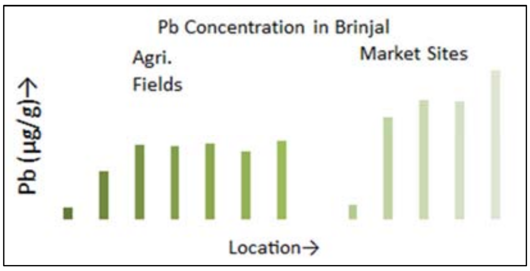

Figure 7. *Concentration of Pb in brinjal.*

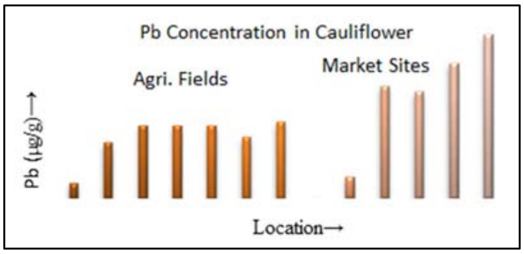

Figure 8. *Concentration of Pb in cauliflower.*

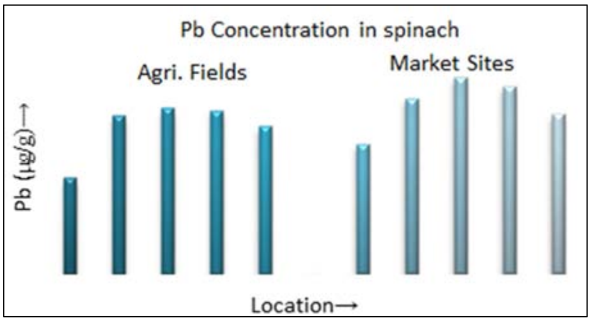

Figure 9. *Concentration of Pb in spinach.*

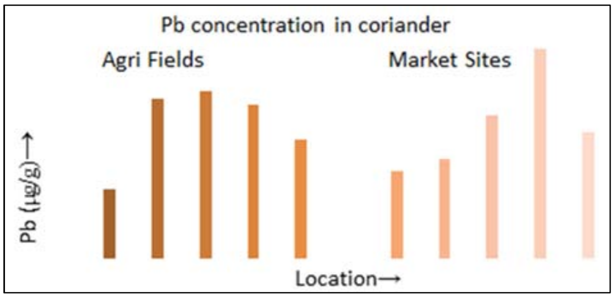

Figure 10. *Concentration of Pb coriander.*

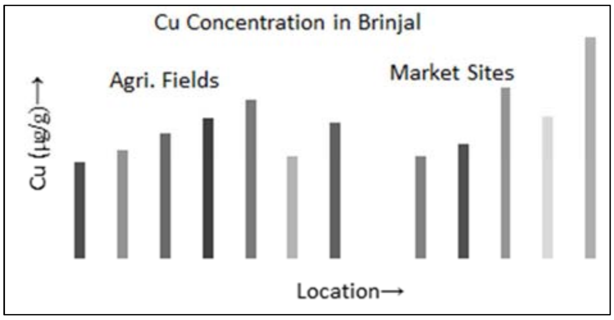

Figure 11. *Concentration of Cu in brinjal.*

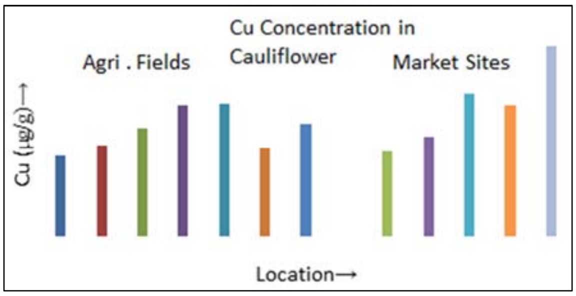

Figure 12. *Concentration of Cu in cauliflower.*

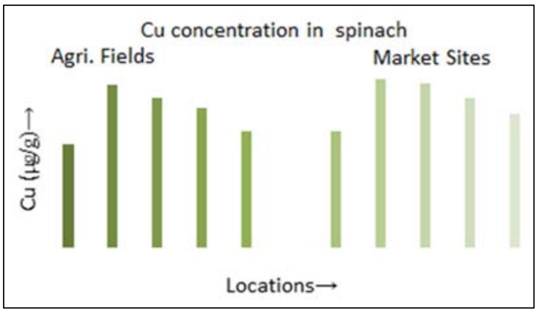

Figure 13. *Concentration of Cu in spinach.*

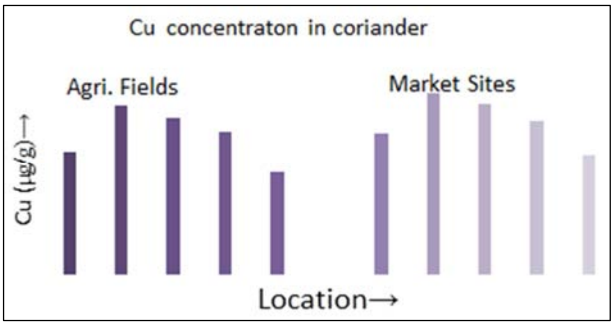

Figure 14. *Concentration of Cu in coriander.*

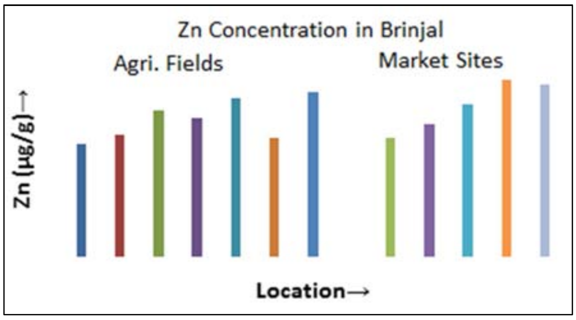

Figure 15. *Concentration of Zn in brinjal.*

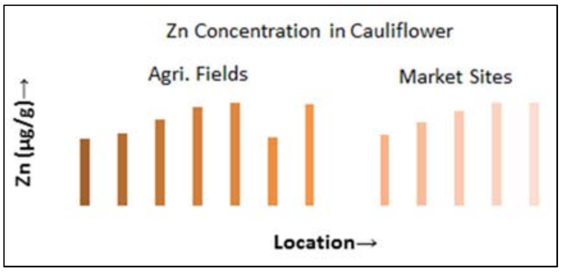

Figure 16. *Concentration of Zn in cauliflower.*

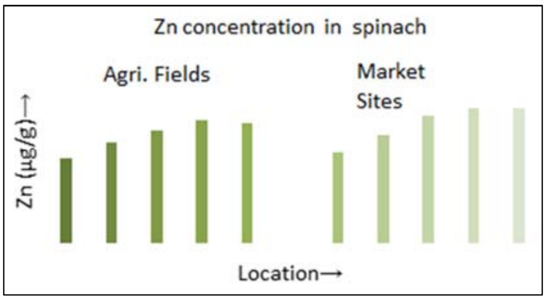

Figure 17. *Concentration of Zn in spinach.*

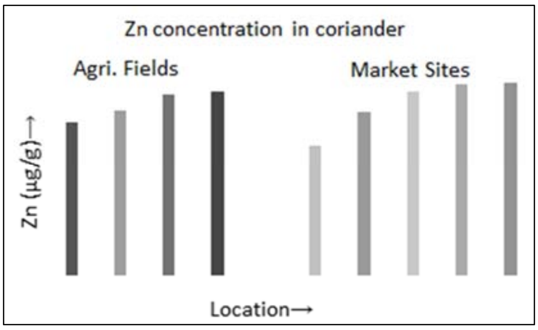

Figure 18. *Concentration of Zn in coriander.*

The maximum concentrations of Cd (µg/g) in vegetable collected from market sites were recorded in spinach at site-II (3.84), at site-III (2.71), at site-IV (3.45), in coriander at site- III (2.71), at site-IV (3.45), at site-V (3.81), in brinjal at site-IV (2.1) and in cauliflower at sites-IV (2.2) as shown in Figures 3-6, which were higher than permissible limits of PFA (1.5 µg/g). The maximum accumulation of Pb (µg/g) in vegetables collected from market at site- V in brinjal (2.54) and in cauliflower (2.84), at III in spinach (1.75) and at site-IV in coriander (2.43) as shown in Figures 7-10. Pb concentration in cauliflower and brinjal were slightly high and spinach and coriander were slightly lower than PFA standard (2.5µg/g). Based on the results of the mean concentration of all metals, Zn showed high and Pb low levels in all the vegetables collected from agricultural and market sites. Similar results reported by various researchers, Sharma and Chettri (2005) [32], Sharma *et al*. (2009) [8] and Degheim *et al.* (2004) [33]. *The absorption of heavy metals from the soil depends on different factors such as pH, organic matter, soil metal availability, cation exchange capacity, plant species, plant growth stages and season and presence of other heavy metals in soil [34]. Data as shown in Table 4 the accumulation of* Cd, Pb, Cu and Zn in the test vegetables collected from market sites and agricultural fields were compared and found that the levels of Cd, Pb, Cu and Zn increased by 47.83, 64.84, 21.3 and 9.91% in brinjal, 36.19, 78.09, 21.83 and 6.50% in cauliflower, 34.52, 49.50, 9.1 and 9.97% in spinach and 27.68, 47.05, 10.34, and 6.13% in coriander. The percent increase in the concentration of heavy metals was observed maximum in brinjal (143.88%) followed by cauliflower (142.61%), spinach (103.09%), and coriander (91.38%). The high accumulation of Pb and Cu were observed in cauliflower than other test vegetables may be due to it having a higher exposed area of inflorescence and hence greater capacity to absorb metals from atmosphere. The high accumulation of heavy metals in vegetables obtained from market sites may also be due to transportation and marketing processing of vegetables which may pose a threat to the quality of the vegetables with consequences for the health of consumers of locally produced foodstuffs, [35].

4.3. Transfer Factor (TF)

The transfer factor of Cd, Pb, Cu and Zn varied from 0.770-1.568, 0.956- 1.429, 1.158-1.358, 1.038-1.132 and 0.965-2.367, 1.576-1.717, 1.329-1.526, 1.141-1.239 for vegetable collected from production and market sites respectively (Table 5). The TF of different heavy metals shows the following order- $TF_{Cu} > TF_{Pb} > TF_{Zn} > TF_{Cd}$ and $TF_{Cd} > TF_{Pb} > TF_{Cu} > TF_{Zn} >$ in case of vegetables collected from production and market sties respectively. To investigate the human health risk index metal transfer factor from soil to plant is essential tool (Cui *et al.,* 2004). The high uptake of metals in these vegetables may be reason to high transpiration rate to sustain the growth and moisture content in leafy vegetables [36]. The transfer factor for Cd, Pb, Cu and Zn greater than one in most test vegetables indicates, greater accumulation and these metals have high transfer factor that migrate to the edible part of the vegetables [37].

Table 5. *TF of heavy metals from soil to vegetables collected from production and market sites.*

Agricultural (production) Fields						**Market Sites**				
Vegetables	**Cd**	**Pb**	**Cu**	**Zn**	**Σ_{TF}**	**Cd**	**Pb**	**Cu**	**Zn**	**Σ_{TF}**
Brinjal	0.770	0.956	1.158	1.038	3.922	0.965	1.576	1.438	1.141	5.12
Cauliflower	0.888	1.143	1.253	1.126	4.41	1.209	1.717	1.526	1.199	5.651
Spinach	1.057	1.190	1.358	1.127	4.732	2.367	1.386	1.481	1.239	6.473
Coriander	1.568	1.429	1.204	1.132	5.333	2.357	1.418	1.329	1.199	6.303
Σ_{TF}	4.283	4.718	4.973	4.423	8.397	6.898	6.097	5.774	4.778	25.547

The soil–plant transfer factor of heavy metals in vegetable collected from market sites was found to be higher than vegetable collected from production sites. High TF (> 1) for metals in vegetables collected from production sites was due to the vicinity of brick kilns or near to national highway, whereas high TF (> 1) for metals in vegetables collected from market sites was due to heavy metals deposition on the vegetables during transport and marking in more polluted urban environment of Katihar city. High TF values were observed for Cd, Pb, Cu and Zn metals with test vegetables and could be one of the possible reasons for health risk in human via their consumption

4.4. Daily Intake of Metals (DIM)

The DIM (μg day^{-1}) values through consumption of test vegetables are given in Table 6. The results revealed that daily intakes of Cd, Pb, Cu and Zn, by adults in vegetables collected from agricultural fields and market sites were found to be 2.078, 1.908, 33.27, 179.60 μg/ day and 3.228, 2.563, 38.198, 202.03 μg / day respectively. The DIM was in order of: $DIM_{Zn} > DIM_{Cu} > DIM_{Cd} > DIM_{Pb}$ for vegetables collected from both production and markets sites. The daily intake of heavy metals through consumption of test vegetables collected from market sites was found to be higher compare the vegetables collected from agricultural fields.

Table 6. *Daily intake of metals (μg day-1) in vegetables collected from production and market sites.*

Agricultural (production) Fields						Market Sites				
Vegetables	**Cd**	**Pb**	**Cu**	**Zn**	Σ_{TF}	**Cd**	**Pb**	**Cu**	**Zn**	Σ_{TF}
Brinjal	0.303	0.402	7.643	42.145	50.493	0.448	0.662	9.272	46.32	56.702
Cauliflower	0.351	0.405	8.078	45.735	54.569	0.912	0.722	9.843	48.71	60.187
Spinach	0.695	0.500	8.754	45.748	55.695	0.936	0.583	9.549	58.31	69.378
Coriander	0.729	0.601	8.801	45.973	56.109	0.932	0.596	9.534	48.69	59.752
Σ_{DIM}	2.078	1.908	33.27	179.60	216.86	3.228	2.563	38.198	202.03	246.02
% of PDTI	3.463	0.892	1.109	0.0029	--------	5.380	1.197	4.606	0.336	-------

PTDI Cd = 60 μg day^{-1}, Pb = 214 μg day^{-1}, Zn = 60 mg day^{-1} and Cu = 3 mg day^{-1}

The results show agreement with previous studies showing levels of heavy metals in edible part of food crops irrigated continuously with wastewater [38, 39], the results also showed that the present findings were lower than 54 μg/day and 412 μg /day of Pb in adult as reported by Debeca *et al*. (1987) [40] and Dick *et al.* (1987) [41] respectively and also lower than 21.6, 858.6, 426.6 and 3.7 mg /day for Cd, Cu, Pb and Zn respectively as reported by Bahemuka and Mubofu (1999) [42]. The data also revealed that consumption of vegetables collected from market to daily intake of Cd, Pb, Cu and Pb were of 5.38, 1.2, 4.606 and 0.336% of provisional tolerable daily intake (PTDI) respectively. Thus the consumption of average amount of these contaminated vegetables does not pose health risk for consumers [43].

4.5. Health Risk Index (HRI)

The HRI values for consumers showed in the following order Cd (3.228) > Cu (0.955) > Zn (0.672) > Pb (0.640) and Cd (2.078) > Cu (0.831) > Zn (0.597) > Pb (0.477) through consumption of test vegetables collected from market sites and agricultural fields respectively (Table 7). The results revealed that HRI value for all individual vegetable collected from market sites was higher compare to vegetable collected from production sites but found to be less than 1.0. The results of the study are in agreement with those reported by Khan *et al.* (2010) [44], Jan *et al.* [26] and lower than those reported by Gupta *et al.* [45] (Σ=6.25). The HRI less than 1 for Cd, Pb, Zn, and Cu through consumption of all test vegetables of the studied area suggest that all vegetables collected from agricultural lands and from market sites were almost safe for consumer.

Table 7. *HRI for heavy metals in vegetables collected from production and market sites.*

Agricultural (production) Fields						Market Sites				
Vegetables	**Cd**	**Pb**	**Cu**	**Zn**	Σ_{HRI}	**Cd**	**Pb**	**Cu**	**Zn**	Σ_{HRI}
Brinjal	0.303	0.101	0.191	0.140	0.735	0.448	0.165	0.232	0.154	0.999
Cauliflower	0.351	0.101	0.202	0.152	0.806	0.912	0.180	0.246	0.162	1.500
Spinach	0.695	0.125	0.218	0.152	1.190	0.936	0.146	0.239	0.194	1.515
Coriander	0.729	0.150	0.220	0.153	1.252	0.932	0.149	0.238	0.162	1.481
Σ_{HRI}	2.078	0.477	0.831	0.597	3.983	3.228	0.640	0.955	0.672	5.495

5. Conclusion

The present study showed that levels of heavy metals in all test vegetables collected from market sites were higher than those collected from agricultural fields. In both sites mean concentration of Cu, Zn, Cd and Pb were lower than PFA standards. The TF, DIM and HRT values for heavy metals in all test vegetables collected from market sites were also found to be higher than those collected from agricultural fields. The results also indicated the variation of accumulation of heavy metals in vegetables tested may be due to anthropogenic activities like continuous irrigation with sewage and wastewater, addition of agro chemicals, use of metal-based pesticides, traffic load, brick kiln industries around production sites and polluted urban environment of Katihar city. Dietary intake of food results in long-term low level body accumulation of heavy metals and the detrimental impart become visible only after long time exposure.

The present research work further suggested that the regular monitor of heavy metals in vegetables is essential to reduce the health risk. Vegetables should be washed properly to remove aerial contamination from surface of vegetables and appropriate precaution should be taken at the time of marketing of vegetables.

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