World Journal of Dairy & Food Sciences 12 (2): 87-93, 2017
ISSN 1817-308X

DOI: 10.5829/idosi.wjdfs.2017.87.93

Health Risk of Heavy Metals in Vegetables Collected from Different Market Sites and Agricultural Fields of Katihar, Bihar, India

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Abstract: The present study was carried out to monitor and assess the levels of Cu, Zn, Cd and Pb in different 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 47.83, 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, respectively and was observed maximum in cauliflower followed by brinjal, spinach and coriander. The average daily intakes of Cd, Pb, Cu and Zn, by adults in vegetables were 5.38, 1.20, 1.3 and 3.267 % 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.

Key words: Soil · Vegetables · Heavy metals · Daily intake · Health risk index · PFA

INTRODUCTION

Vegetables are important part of human diet and are known to have positive effect on human health [1, 2]. Anthropogenic and natural process are responsible for increasing the levels of heavy metals in the agricultural soil [3, 4].The use of pesticides, fungicides, fertilizers and agro chemicals by farmers for high production of vegetables may increase the concentration of heavy metals in soil which are taken by aerial parts of plant. 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 [5, 6].

The leafy part of vegetables was found to be contaminated by heavy metals more frequently than the stem and root parts of plant [1, 7].This is due to higher transpiration rate to maintain the growth and moisture content of these plants [8]. The major sources of metals are wastewater untreated or partially treated industrial effluents, municipal wastes and vehicles [9, 10]. 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 [11].

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 [12-14]. The assessment of heavy metal concentration in the vegetables from the market sites are being carried in some developed countries and there are very few published data in India, however not available in the literature of Katihar city. Thus the aims of the research work were 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.

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MATERIALS AND METHODS

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. We have selected two study sites 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). On the basis of the production capacity of these vegetables and transportation to the market, we have selected 24 sites in the agricultural fields and five locations in the market. 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 (16, 17, 18, 19, 21, 22 and 24) 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. The name and number of market sites and agricultural fields are listed in Table 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 ^{0}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*. [15].

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 on 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.* [16].

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 [17].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).

Estimation of Daily Intake of Heavy Metals (DIM): The daily intake of heavy metals by people through consumption of vegetables was calculated by using following formula, Chary *et al.* [18].

$$DIM\ (\mu g\ day\text{-}1) = \frac{C\ metal \times D\ food\ int\ ake \times C\ factor}{B\ averageweight} \tag{1}$$

Where,
C_{metal} = Heavy metal concentration in plant (mg kg^{-1}), $D_{food\ intake}$ = Daily intake of vegetable (gm day^{-1} $person^{-1}$), $B_{average\ weight}$ = Average body weight (kg), C_{factor} = Conversion factor (0.085).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 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.

Health Risk Index (HRI): The value of HRI depends upon the daily intake of metals and reference dose, which was computed as described by Jan *et al*. [19]. If the value of HRI is less than 1 then the exposed population said to be safe [20].

$$HRI = \frac{DIM}{RfD} \tag{2}$$

Where,
DIM = Daily intake of heavy metals (µg day^{-1}), Rf_D -Reference dose, Rf_D value for Cu, Pb, Cd and Zn is 0.04, 0.004, 0.001 and 0.30 (mg/kg bw /day) respectively [20].

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	Coriander	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							
		Brinjal		Cauliflower		Spinach		Coriander	
Sites name	no.	Sites name	no	Sites name	no	Sites name	no	Sites name	no
New market	I	Belwa	1	Varmaili	8	Sarinda	15	Boodhnagar	20
Bhagwan chowk	II	Govind chowk	2	Bastol	9	Sarbasa	16	Daheria	21
Chalisa hatia	III	Hajipur	3	Ranipatra	10	Sonoli	17	Kuretha	22
Mirchaibari	IV	Sarif ganj	4	Dalaon	11	Haplaganj	18	Serinia	23
Mahamood chowk	V	Hawai adda	5	Rampur	12	Chattabari	19	Pharthali	24
		Sirsa	6	Pranpur	13				
		Mania	7	Rhotara	14				

Statistical Analysis: All statistical analysis were performed on lenovo ™ computer using the Microsoft EXCEL and Word 2003 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.

RESULTS AND DISCUSSIONS

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.* [21].Whereas upper limits of Cd, Pb, Cu and Zn were below the upper permissible limits as recommended by Prevention of food Adulteration (PFA) standard (6, 500, 270 and 300 µg/g) as guided by Awasthi [22]. 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. 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 [23, 24].

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

Statistical data	Pb	Cd	Cu	Zn
Min.	0.10	0.40	10.2	80.20
Max.	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
$?_n$	26.13	28.89	400.9	25.24.4
σ_{n-1}	0.494	0.561	4.648	13.815

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

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. The high accumulation of heavy metals in the test vegetables collected from agriculture fields at 5, 7, 12, 14, 21 and 22 may be due to their location near to national highway and sites 16, 17, 18 and 24 may be due to being very close to brick kiln industries. 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. 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. 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), 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). 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 [25] and Sharma *et al*. [26] and Degheim *et al.* [27].

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

	Agricultural (production) fields				Market sites			
Metals/Data	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
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
$?_n$	5.50	6.36	9.01	9.440	5.81	6.19	12.12	12.07
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	0.4735	0.868	0.934	0.236	0.558
$?_n$	7.29	7.35	6.48	7.780	8.58	9.35	7.55	7.72
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	20.928	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
$?_n$	138.6	146.5	113.4	100.6	120.1	127.5	123.7	111.00
Znic								
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	118.486	118.52	119.1	120.00	126.18	130.34	126.4
s_{n-1}	15.656	22.62	16.926	16.279	19.509	19.494	20.76	19.29
$?_n$	764.3	829.4	592.6	595.5	600.00	630.9	651.7	630.7

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

Table 5: DIM (µg day-1) in vegetables collected from market sites and agricultural fields

	Agricultural (production) fields				Market sites			
Vegetables	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn
Brinjal	0.303	0.402	7.643	42.145	0.448	0.662	9.272	46.32
Cauliflower	0.351	0.405	8.078	45.735	0.912	0.722	9.843	48.71
Spinach	0.695	0.500	8.754	45.748	0.936	0.583	9.549	58.31
Coriander	0.729	0.601	8.801	45.973	0.932	0.596	9.534	48.69
Total	2.078	1.908	33.27	178.6	3.226	2.563	138.19	202.03

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

Table 6: HRI for heavy metals in vegetables collected from market and agricultural fields

	Agricultural (production) fields				Market sites			
Vegetables	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn
Brinjal	0.303	0.101	0.191	0.140	0.448	0.165	0.232	0.154
Cauliflower	0.351	0.101	0.202	0.152	0.912	0.180	0.246	0.162
Spinach	0.695	0.125	0.218	0.152	0.936	0.146	0.239	0.194
Coriander	0.729	0.150	0.220	0.153	0.932	0.149	0.238	0.162
Mean	0.519	0.119	0.415	0.149	0.807	0.160	0.238	0.168

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 [28].

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 cauliflower followed by brinjal, spinach and coriander. The accumulation of Pb, Cd and Cu were high 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, [29].

Daily intake of heavy metals (DIM): The DMI (µg day^{-1}) values through consumption of test vegetables are given in Table 5. The results revealed that average daily intakes of Cd, Pb, Cu and Zn, by adults in vegetables collected from agricultural fields and market sites were found to be 0.518, 0.477, 8.319 & 44.9 µg/ day and 0.807, 0.641, 9.536 & 50.51 µg/ day respectively. The average DIM was in order of: $DIM_{Zn} > DIM_{Cu} > DIM_{Cd} > DIM_{Pb}$. The results show agreement with previous studies showing levels of heavy metals in edible part of food crops irrigated continuously with wastewater [30, 31], 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*. [32] and Dick *et al.* [33] 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 [34]. The data also revealed that consumption of vegetables collected from market to daily intake of Cu, Zn, Cd and Pb were of 5.38,1.2, 1.3 and 3.71% of provisional tolerable daily intake (PTDI) respectively. Thus the consumption of average amount of these contaminated vegetables does not pose health risk for consumers [35].

Human health Risk Index (HRI): The HRI calculated for heavy metals for consumption for adults (Table 6) revealed that HRI for all individual vegetables were lower than 1.0 for Cd, Pb, Zn and Cu. The mean HRI values for consumers showed the following order Cd (0.807) > Cu (0.238) > Zn (0.168) > Pb (0.160). Our results are in agreement with those reported by Khan *et al.* [36], Jan *et al.* [19] and lower than those reported by Gupta *et al.* [37] (=6.25). The HRI (less than 1) of the study area suggest that all vegetables collected from agricultural lands and from market sites were almost safe for consumer.

CONCLUSIONS

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 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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