

Heavy Metal Contamination of Vegetables

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Abstract

In India, several (>50) vegetables are widely used as food due to their high nutrition values. However, vegetables in industrial area are getting contaminated with heavy metals by disturbing biological and biochemical processes in the human body. In present study, the risk of human health by heavy metals (Fe, As, Cr, Mn, Cu, Zn, Pb, Cd and Hg) through the intake of common vegetables *i.e. Solanum lycopersicum, Solanum melongena, Amaranthus tricolor* L., *Chenopodium album* L., *Spinacia oleracea* and *Coriandrum sativum* obtained from the largest coal burning basin, Korba, India is described . The concentration of Fe, As, Cr, Mn, Cu, Zn, Cd, Pb and Hg in the soils (n = 6) was ranged from 18,328 - 37,980, 85 - 105, 34 - 72, 314 - 760, 146 - 165, 126 - 164, 1.11 - 1.39, 116 - 148 and 0.11 - 0.21 mg/kg with mean value (p = 0.05) of 28,011 \pm 6582, 96 \pm 6, 57 \pm 11, 597 \pm 148, 153 \pm 5, 145 \pm 11, 1.26 \pm 0.10, 133 \pm 11 and 0.16 \pm 0.03 mg/kg, respectively. The contamination, sources and bioaccumulation, pollution and health risk indices of the heavy metals *i.e.* As, Fe, Cr, Mn, Cu, Zn, Pb, Cd and Hg in the plants are described.

Keywords

Heavy Metals, Vegetables, Contamination, Metal Pollution Index, Health Risk Index

1. Introduction

Vegetables are common diet taken by populations throughout the world, being rich sources of essential nutrients, antioxidants and metabolites by acting as buffering agents for acidic substances produced during the digestion processes [1]. However, both essential and toxic elements were absorbed by vegetables from the soil [2]. Potential human health risks *i.e.* cancer, kidney damage, etc. from consumption of contaminated vegetables with the heavy metals (HMs) were reported [3]. The heavy metals *i.e.* Cr, Mn, Fe, Ni, Cu, Zn, Cd, Pb and Hg were sig-

nificantly accumulated in commonly used vegetables *i.e.* Amaranthus tricolor L., Chenopodium album L., Spinacia oleracea, Coriandrum sativum, Solanum lycopersicum and Solanum melongena [4]-[11]. In the present work, the contamination, enrichment, health risk and sources of the heavy metals *i.e.* Fe, As, Cr, Mn, Cu, Zn, Pb, Cd and Hg in six vegetables (*i.e.* Solanum lycopersicum, Solanum melongena, Amaranthus tricolor L., Chenopodium album L., Spinacia oleracea and Coriandrum sativum) grown in soil of the largest coal basin: Korba city, Chhattisgarh, India are described.

2. Materials and Methods

2.1. Collection of Vegetables

The vegetables were collected from Urga area of Korba in January, 2013, Figure 1. Six commonly used



Figure 1. Representation of sampling location.

vegetables (*i.e. Solanum lycopersicum, Solanum melongena, Amaranthus tricolor* L., *Chenopodium album* L., *Spinacia oleracea* and *Coriandrum sativum*) rich in nutrition and minerals were selected for analysis of the heavy metal loads [12]. The vegetables were harvested from the center and four corners of the plot. The vegetables were washed with deionised water to remove any possible foliar contaminants, such as pesticides, fertilisers, dust and mud. They were then cut into small pieces using a stainless steel knife and oven dried to remove moisture at 60°C to a constant mass. The dried tissue was stored in a moisture-free atmosphere prior to further processing. The samples were then ground using a ceramic mortar and pestle to reduce the dried material to a suitable size for digestion and analysis.

2.2. Collection of Soil

The soil samples (100 g) were collected with a stainless steel hand-trowel, within 0 - 20 cm depths from the rhizospheric area in January, 2013 [13]. The trowel was carefully cleaned after each sampling exercise, to avoid cross-contamination. The soil samples were kept in the polyethylene bottle and dried in an oven at 60°C for 12 hrs. The sample was milled and sieved out particles of ≤ 1 mm for the analysis.

2.3. Analysis

The P/T MARS CEM (Varian Company) microwave digester was used for the digestion of soil and plant samples at 200°C withhold time of 15 min. The soil sample (0.25 g) was digested with aqua regia (2 mL of HNO₃, 65%, v/v + 6 mL of HCl, 37%, v/v) by using EN 13346 method. The plant sample (0.25 g) was digested with 5 mL HNO₃, 65%, v/v + 2 mL H₂O₂ by using EN 14084 method. The Varian Liberty AX Sequential ICP-AES (plasma flow: 15 L/min, auxiliary flow: 1.5 L/min, power: 1KW, PMT voltage: 650 V) was used for analysis of the metals *i.e.* Cr, Mn, Fe, Cu and Zn. A VARIAN "SpectrAA 220Z" model graphite furnace atomic absorption spectrometer (GF-AAS) equipped with a longitudinal Zeeman effect background corrector was employed for analysis of metals *i.e.* Cd and Pb. The VARIAN "SpectrAA 55B" equipped with hydride/cold vapor regenerator accessories was selected for analysis of elements *i.e.* As and Hg.

2.4. Quality Control Analysis

The standard soil (NCS DC73382, Spex Certiprep, UK) and apple leaf (NIST-1515, USA) reference materials were used for the quality control of the heavy metals. The accuracy of the results were found within $\leq \pm 2\%$ of the certified value. The relative standard deviations of the triplicate analysis (n = 3) was $\leq \pm 6\%$.

2.5. Metal Pollution Index

The Metal Pollution Index (MPI) is a geometrical mean of concentration of all tested metals in a given food stuff and computed by using following equation [14].

$$MPI(mg/kg) = (Cf_1 \times Cf_2 \times Cf_3 \times \cdots \times Cf_n)^{1/n}$$

where, Cf_n is a concentration of n^{th} metal in a given food stuff.

2.6. Health Risk Index

The Health Risk Index (HRI) was computed as the ratio of estimated exposure of tested vegetables and oral reference dose by using following equation:

$$HRI = DIM/R_f D$$

where, DIM and R_fD represent daily intake of metal and reference oral dose, respectively. The DIM was evaluated by using following equation:

$$\text{DIM} = C_{\text{metal}} \times C_{\text{factor}} \times D_{\text{food intake}} / B_{\text{average weight}}$$

where, C_{metal} , C_{factor} , $D_{\text{food intake}}$ and $B_{\text{average weight}}$ denote metal concentration in plant (mg/kg), conversion factor, daily intake of vegetable and average body mass of the consumers, respectively. The conversion factor of 0.085 was used to convert fresh vegetable weight to dry weight [15]. The average body of the consumers chosen was 60 kg. The daily intake of vegetables *i.e. Solanum lycopersicum, Solanum melongena, Amaranthus tricolor* L.,

Chenopodium album L. and *Spinacia oleracea* was 300 g. However, daily intake of *Coriandrum sativum* was 100 g. The oral reference dose for Hg, As, Cd, Pb, Cu, Mn, Zn, Cr and Fe reported was 0.0001, 0.0003, 0.001, 0.004, 0.04, 0.14, 0.3, 1.5 and 15 mg/kg/day, respectively [16].

3. Results & Discussion

3.1. Heavy Metal Concentration in Soils

Each soil sample was analyzed in triplicate, and the mean value of heavy metals *i.e.* Fe, As, Cr, Mn, Cu, Zn, Cd, Pb and Hg is presented in **Table 1**. The concentration of Fe, As, Cr, Mn, Cu, Zn, Cd, Pb and Hg in the rhizospheric soils (n = 6) was ranged from 18328 - 37980, 85 - 105, 34 - 72, 314 - 760, 146 - 165, 126 - 164, 1.11 - 1.39, 116 - 148 and 0.11 - 0.21 mg/ kg with mean value (p = 0.05) of 28,011 \pm 6582, 96 \pm 6, 57 \pm 11, 597 \pm 148, 153 \pm 5, 145 \pm 11, 1.26 \pm 0.10, 133 \pm 11 and 0.16 \pm 0.03 mg/kg, respectively. The background level of Al, Fe, Mn, Cr, Cu, Zn, Cd, Pb, As and Hg in the earth crust reported was 81,530, 39,200, 775, 92, 28, 67, 0.09, 17, 4.8 and 0.05 mg/kg [17]. The mean enrichment factor (E_f) value with respect to Al for Fe, Mn, Cr, Cu, Zn, Cd, Pb, As and Hg was found to be 1.0, 1.1, 0.8, 8.4, 2.9, 19, 11, 27 and 4.0, respectively. Three metals *i.e.* Cd, Pb and As were enriched extremely in the soil of the studied area. The HMs concentration in the surface soil of the Korba basin was found to be higher than other coal burning regions of the country, probably due to higher exploitation of coals [18] [19].

3.2. Heavy Metal Concentration in Vegetables

Five species of each vegetable was analyzed, and their mean value at 95% confidence limit is summarized in **Table 2**. *Solanum lycopersicum* (tomato) is a staple common vegetable consumed by all classes of Indian citizens. Tomatoes are often a significant part of the human diet and are also abundant sources of antioxidants. The mean concentration value of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg (n = 5) was 0.56 ± 0.06 , 500 ± 23 , 2.9 ± 0.6 , 71 ± 11 , 25 ± 8 , 29 ± 10 , 0.42 ± 0.11 , 1.1 ± 0.4 and 0.07 ± 0.03 mg/kg, respectively. The safe limits reported for As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg in the vegetables were 0.1, 425, 2.3, 500, 40, 100, 0.1, 0.2 and 0.03 mg/kg, respectively. The concentration of As, Cd, Pb and Hg was found to be higher than the safe limits. The content of metals *i.e.* Cr, Zn, Cd and Cu in the tomato of the studied area was observed to be comparable to the values reported in the Varanasi city, India [20]. *Solanum melongena* (Brinjal), an angiospermic family member of Solanaceae, is a common and popular vegetable crop grown in the subtropics and tropics, even in pollutant contaminated regions. The mean concentration value of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg (n = 5) was 0.78 ± 0.09 , 192 ± 27 , 1.0 ± 0.4 , 17 ± 9 , 29 ± 11 , 24 ± 7 , 0.25 ± 0.07 , 0.7 ± 0.03 and 0.08 ± 0.04 mg/kg, respectively. The concentration of four metals: As, Cd, Pb and Hg in the brinjal was found to be higher than the safe limits. The concentration of four metals: As, Cd, Pb and Hg in the brinjal was found to be higher than the safe limits. The concentration of four metals: As, Cd, Pb and Hg in the brinjal was found to be higher than the safe limits. The content of Fe, Mn, Cu and Zn in the brinjal of the studied area was found to be comparable the values reported in the other parts of country [21] [22].

Amaranthus species are cultivated and consumed as leafy vegetables in many parts of the world. Amaranthus are excellent raw in salads, used as a steamed vegetable, and included in soups and stews. The mean concentration value of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg (n = 5) was found to be 2.08 ± 0.22 , 2255 ± 116 , 10 ± 3 , 200 ± 16 , 36 ± 4 , 106 ± 15 , 0.67 ± 0.11 , 0.7 ± 0.04 and 0.14 ± 0.05 mg/kg, respectively. The content of six metals *i.e.* As, Fe, Cr, Cd, Pb and Hg was found higher than the safe limit of 0.1, 425, 2.3, 0.1, 0.2 and 0.03 mg/kg,

S. No.	Sample No.	Fe	As	Cr	Mn	Cu	Zn	Cd	Pb	Hg
1	S1	$24{,}235\pm1020$	103 ± 11	49 ± 6	712 ± 58	165 ± 9	159 ± 8	1.11 ± 0.09	147 ± 10	0.21 ± 0.4
2	S2	$33,\!865\pm1424$	98 ± 9	59 ± 7	749 ± 62	150 ± 8	136 ± 7	1.31 ± 0.10	136 ± 9	0.17 ± 0.03
3	S 3	$33{,}678 \pm 1437$	105 ± 12	62 ± 8	618 ± 52	146 ± 7	140 ± 6	1.39 ± 0.11	133 ± 8	0.11 ± 0.02
4	S4	$19{,}980 \pm 946$	85 ± 8	67 ± 8	428 ± 46	153 ± 9	164 ± 7	1.23 ± 0.12	116 ± 7	0.19 ± 0.03
5	S5	$37{,}980 \pm 1547$	91 ± 7	72 ± 9	760 ± 65	156 ± 8	144 ± 6	1.38 ± 0.13	148 ± 10	0.14 ± 0.03
6	S6	$18{,}328\pm878$	93 ± 6	34 ± 5	314 ± 34	149 ± 7	126 ± 6	1.13 ± 0.11	119 ± 8	0.12 ± 0.02

Table 1. Mean value (n = 5) of heavy metal content in rhizospheric soil, mg/kg.

S = Soil sample.

Table 2. Mean value $(n = 5)$ of heavy metal content in dried vegetable, mg/kg.											
S. No.	Vegetable	As	Fe	Cr	Mn	Cu	Zn	Cd	Pb	Hg	
V1	SL	0.56 ± 0.06	500 ± 23	2.9 ± 0.6	71 ± 11	25 ± 8	29 ± 10	0.42 ± 0.11	1.1 ± 0.4	0.07 ± 0.03	
V2	SML	0.78 ± 0.09	192 ± 27	1.0 ± 0.4	17 ± 9	29 ± 11	24 ± 7	0.25 ± 0.07	0.7 ± 0.03	0.08 ± 0.04	
V3	ATL	2.08 ± 0.22	2255 ± 116	10 ± 3	200 ± 16	36 ± 4	106 ± 15	0.67 ± 0.11	0.7 ± 0.04	0.14 ± 0.05	
V4	CAL	1.86 ± 0.24	1985 ± 97	17 ± 5	213 ± 18	46 ± 6	118 ± 13	1.57 ± 0.23	2.3 ± 0.2	0.18 ± 0.05	
V5	SO	1.99 ± 0.22	1406 ± 65	8.0 ± 2.1	676 ± 34	62 ± 8	98 ± 10	1.46 ± 0.19	4.7 ± 0.4	0.12 ± 0.04	
V6	CS	2.05 ± 0.31	1618 ± 71	7.6 ± 1.8	59 ± 7	71 ± 9	121 ± 11	0.75 ± 0.09	1.8 ± 0.2	0.14 ± 0.05	

SL = Solanum lycopersicum, SML = Solanum melongena L., ATL = Amaranthus tricolor L., CAL = Chenopodium album L., SO = Spinacia oleracea, CS = Coriandrum sativum.

respectively. The Zn, Cd and Pb content in the Amaranthus leaves of the studied area was found to be comparable to the values reported in African and Bangladesh Amaranthus leaves [6] [7]. The high content of Pb (5.75 mg/kg) in the Amaranthus leaves grown in Lagos was reported [11]. Chenopodium album L. is cultivated as a food crop in Asia and parts of Africa. Especially in India, it has major cultivations and is eaten as a leafy vegetable like many others. The mean concentration value (n = 5) of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg was found to be 1.86 ± 0.24 , 1985 ± 97 , 17 ± 5 , 213 ± 18 , 46 ± 6 , 118 ± 13 , 1.57 ± 0.23 , 2.3 ± 0.2 and 0.18 ± 0.05 mg/kg. The content of all metals except Mn was found higher than reported safe limits. The Chenopodium album L. was reported as phytoextractants for metals i.e. Cr, Mn, Fe, Cu, Zn, Cd and Pb from the contaminated soils [4] [23]. In the present work, the *Chenopodium album* L. was found to be as good phytoextracting plant for the metals i.e. As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg. Spinach (Spinacia oleracea) is one of widely used as leafy vegetable rich in nutrients, antioxidants and anticancer constituents. The high loading of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg was marked with mean value of 1.99 ± 0.22 , 1406 ± 65 , 8.0 ± 2.1 , 676 ± 34 , 62 ± 8 , 98 ± 2.1 10, 1.46 ± 0.19 , 4.7 ± 0.4 and 0.12 ± 0.04 mg/kg, respectively. All metals (except Zn) were strongly accumulated in the Spinacia oleracea by crossing the safe limits. The Cu, Cd and Pb contents observed in the Spinacia leaves of the studied area was found to be comparable the values reported in the leaves of the Bangalore, Delhi and Bangladesh regions [6] [10] [24].

The Coriandrum sativum has a very strong aroma with rich in nutrients, vitamins and antibacterial chemicals. The fresh leaves of *Coriandrum sativum* are widely used an ingredient in Indian foods. The mean concentration value of As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg (n = 5) was 2.05 ± 0.31 , 1618 ± 71 , 7.6 ± 1.8 , 59 ± 7 , 71 ± 9 , $121 \pm 11, 0.75 \pm 0.09, 1.8 \pm 0.2$ and 0.14 ± 0.05 mg/kg, respectively. The As content in the *Coriandrum sativum* leaves of the studied area was found to be higher than the value reported in the coriander leaves of the West Bengal [5]. The content of other metals *i.e.* Cu, Zn, Cd and Pb in the leave of studied area was found to be comparable to the values reported in the leaves of Bangalore city and Bangladesh [6] [24].

3.3. Bioaccumulation Index of Metals

The bioaccumulation index (BI) is a ratio of vegetable to soil content of the metal. Significant BI values (>0.8) of the Mn and Cd with Spinacia oleracea, Zn and Hg with Coriandrum sativum, Hg with Amaranthus tricolor L. and Cd and Hg with Chenopodium album L. were recorded, may be due to coordination of the metals with the biomass oxalates [25].

3.4. Metal Pollution Index

The metal pollution index (MPI) for As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg in Solanum lycopersicum, Solanum melongena, Amaranthus tricolor L., Chenopodium album L., Spinacia oleracea and Coriandrum sativum was found to be 4.8, 3.1, 11.0, 15.0, 16.2 and 11.0 mg/kg, respectively. The several folds higher MPI value for the leafy vegetables was marked, may be due to their higher biomass production as shown in Figure 2.

3.5. Health Risk Index

The daily intake of metals (DIM) is shown in Table 3. The DIM value As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg

for six vegetables was ranged from 0.24 - 0.88, 82 - 958, 0.4 - 7.2, 7 - 287, 11 - 26, 10 - 50, 0.11 - 0.67, 0.26 - 2.00 and 0.030 - 0.077 mg/g/day/person, respectively. The remarkably high DIM value of four metals *i.e.* Fe, Mn, Cu and Zn for all vegetables was observed. Among them, the highest DIM value for metals *i.e.* As, Fe, Cr, Zn, Pb and Hg with the *Amaranthus tricolor* L. was marked as shown in **Figure 3**. The HRI values of metals in the vegetables are summarized in **Table 4**. The extremely high HRI value of As (>500) with all vegetables was observed. Among the use of the vegetables are summarized in **Table 4**. The extremely high HRI value of As (>500) with all vegetables was recorded. The highest HRI value of metals *i.e.* As, Mn, Cu, Cd, Pb and Hg with the *Spinacia oleracea* was observed due to higher Cd and Pb contents, **Figure 4**.

3.6. Factor Analysis

The results of FA gave two factors accounted for 92.86% of the total variance (**Table 5**). Factor-I accounted for 71.51% of the total variance. It explained more than the half of the total variance, and is positively correlated with Cr, Fe, Zn, Hg and As (**Table 6**). Factor-1 could characterize the behavior of the above-mentioned HMs in the relation to oxides/oxyhydroxides of Fe. Factor-II was positively correlated with Cd, Mn and Pb, (**Table 6**). This factor may be termed as the association of Mn oxides with Cd and Pb.



Figure 2. Metal pollution index in vegetables. SL = Solanum lycopersicum, SM = Solanum melongena, AT = Amaranthus tricolor, SO = Spinacia oleracea, CS = Coriandrum sativum.



Figure 3. The DIM value for various vegetables.



Figure 4. HRI value of vegetables for nine metals: As, Fe, Cr, Mn, Cu, Zn, Cd, Pb and Hg.

rane of Dairy make of metals, mg/g/day/person.									
Vegetable	As	Fe	Cr	Mn	Cu	Zn	Cd	Pb	Hg
S. lycopersicum	0.24	213	1.2	30	11	12	0.18	0.47	0.030
S. melongena	0.33	82	0.4	7	12	10	0.11	0.30	0.034
Amaranthus tricolor L.	0.88	958	4.3	85	15	45	0.28	0.30	0.060
Chenopodium album L.	0.79	844	7.2	91	20	50	0.67	0.98	0.077
Spinacia oleracea	0.85	598	3.4	287	26	42	0.62	2.00	0.051
C. sativum	0.29	229	1.1	8	10	17	0.11	0.26	0.020
Table 4. Health risk index of metals.									
Vegetable	As	Fe	Cr	Mn	Cu	Zn	Cd	Pb	Hg
S. lycopersicum	793	14	0.8	216	266	41	179	117	298
S. melongena	1105	5	0.3	52	308	34	106	74	340
Amaranthus tricolor L.	2947	64	2.8	607	383	150	285	74	595
Chenopodium album L.	2635	56	4.8	647	489	167	667	244	765
Spinacia oleracea	2819	40	2.3	2052	659	139	621	499	510
C. sativum	968	15	0.7	60	251	57	106	64	198

Table 3. Daily intake of metals, mg/g/day/person

4. Conclusion

The soil in the studied area was extremely contaminated with toxic metals *i.e.* As, Pb and Hg. They were accumulated in the vegetables, and their prolonged consumption may disturb the biological and biochemical processes in the humans. The significant BI value of metals *i.e.* Mn, Zn, Cd and Hg with the leafy vegetables was observed. Among them, the highest MPI value for the metals with *Spinacia oleracea* was recorded which may be due to the higher biomass production. The extremely high HRI value for As in all tested vegetables was observed. The detailed investigation of the MPI value for various types of leafy vegetables grown in the studied area is urgently required for mapping of the human health hazards in local people.

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Table 5. Results of factor analysis of heavy metals in the vegetables.								
Variable	Factor-I	Factor-II						
Cd	0.41	0.91						
Cr	0.92	0.28						
Cu	0.61	0.55						
Fe	0.96	0.10						
Mn	0.18	0.93						
Pb	0.10	0.99						
Zn	0.95	0.28						
Hg	0.96	0.21						
As	0.96	0.26						
Eigenvalue	6.44	1.92						
% Variance	71.51	21.35						
%Cumulative variance	71.51	92.86						

Loading value > 0.70, significant at p < 0.05.

Table 6. Correlation matrix of heavy metals.												
	As	Fe	Cr	Mn	Cu	Zn	Cd	Pb	Hg			
As	1											
Fe	0.91	1										
Cr	0.72	0.86	1									
Mn	0.49	0.32	0.31	1								
Cu	0.75	0.47	0.37	0.44	1							
Zn	0.96	0.93	0.83	0.37	0.76	1						
Cd	0.66	0.61	0.82	0.72	0.56	0.71	1					
Pb	0.45	0.22	0.32	0.91	0.65	0.41	0.80	1				
Hg	0.84	0.89	0.95	0.23	0.54	0.92	0.74	0.28	1			

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