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Contamination Status of Heavy Metals in Vegetables and Soil in Satkhira, Bangladesh

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Received 13 Feb 2019, Revised 04 July 2019,	Abstract Vegetables are the principal source of nutrients and play a crucial role to maintain sound
Accepted 05 July 2019	health. But, vegetables are being vitiated by various types of unwanted contaminants and becoming an alarming issue powedays. Inteke of contaminated vagetables may cause several
	diseases and hamper normal physiological functions. Therefore, the main purpose of this study is to establish a database about the contamination status of heavy metals in popular vegetables
Keywords ✓ Heavy metals	and their growing soil in Satkhira, Bangladesh; to assess the associated health risks of consumers through target health quotient (THQ) and target cancer risk (TCR) analyses. The
 ✓ Health risk, ✓ Carcinogenic, 	average concentration of Mn, Fe, Cu, Zn, Cd and Pb is 33.91, 356.71, 10.27, 33.59, 0.57 and 9.76 mg kg ⁻¹ in vegetables and 239.34, 3399.38, 22.48, 65.63, 0.68, 11.53 mg kg ⁻¹ in growing
✓ Non-carcinogenic,	soil. The concentration of heavy metals has been compared with the standard value
 ✓ Hazard index ✓ Satkhira. 	recommended by WHO/FAO and it is found that the average concentrations of Fe, Pb, and Cd in the leafy, fruit and root vegetables exceeded the permissible limit. Moreover, the value of
	THQ, non-carcinogenic parameter, is greater than 1.0 for Fe and Pb in leafy, fruits and root vegetables. Therefore, the THQ of Fe and Pb may pose a potential health risk to human.
	Besides, the probability of developing cancer is greater than USEPA risk limit (>10 ⁻⁶) and the TCR of Pb shows high cancer risk whereas Cd pases a very high cancer risk. Therefore, the
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1. Introduction

At present, heavy metal pollution is one of the most alarming environmental problems not only in Bangladesh, a developing country, but also all over the world [1]. Soil, water, and even air are being contaminated with heavy metals due to the rapid growth of industrialization, use of different fertilizers, pesticides and herbicides in agricultural fields. Besides, randomly disposal of household wastes, livestock manure, and unused metallic parts are the main causes of the pollution of soil and water [2, 3]. The entrance of heavy metal in the food chain is the major route of heavy metal exposure to human [1, 4]. Vegetables are an important part of human diets and the prime source of minerals, fiber, and vitamins. Plants, mainly leafy vegetables, absorb these heavy metals from contaminated soil. Besides, heavy metals are also deposited on the different parts of vegetables from the air [5]. Heavy metals are the major contaminants of the food supply that affect the nutritive values of vegetables eventually pose deleterious effects to human health. Intake of contaminated vegetables supply the heavy metals in the human body that cause a detrimental effect on the human body like damage of DNA, change the genetic code and reduce the energy level of the human body. Exposure to certain metals like mercury and lead may also cause autoimmune disorders such as rheumatoid arthritis, kidney diseases, etc. Besides, they inhibit the common functions of liver, kidneys, lungs, and heart, etc [6-9]. Heavy metals generally combine with the thiol, amino and imino group of protein and form a metal complex. As a result, the proteins lose their biological activities and cause the breakdown of the cell [10]. In addition, heavy metals also affect the philological functions of plants and retard the nitrogen fixation, chlorosis, metabolism and the growth of plants [10]. Recently, many countries have launched regular monitoring and assessment of heavy metal in food and vegetables. But there is insufficient data available about the contamination level of heavy metals in Bangladesh. Due to unrest growth of industrialization, geographical position, household activities and the excess use of fertilizer, vegetables in this study area might be

contaminated. This study, therefore, aims to determine the concentration of heavy metals in commonly consumed vegetables and to assess the carcinogenic and non-carcinogenic health risks of consumers through THQ and TCR analyses.

Geographical Location of the Study Area

Satkhira is a district of Khulna division having an area of 3858.33 sq km. It located in between 21°36' and 22°54' north latitudes and in between 88°54' and 89°20' east longitudes (Figure 1). It is bounded by Jashore district on the north, the Bay of Bengal on the south, Khulna district on the east, the West Bengal state of India on the west.



Figure 1: Map of the study area (Satkhira, Bangladesh)

2. Materials and Methods

2.1 Sample Collection and Preparation

Different species of leafy, fruit and root vegetables and their growing soils were randomly collected from ten relevant sites of Satkhira, Bangladesh. Table 1 presents a general description of the studied vegetables cultivated in this region. The vegetable samples were washed with distilled water and the edible parts of the vegetables were chopped into small piece. To obtain constant weight, the samples were air dried and kept in an oven at 100±1°C. The soil samples were also air dried at room temperature and ground into finely powdered form. To determine the concentration of heavy metals (Mn, Fe, Cu, Zn, Pb, and Cd), the vegetable and soil samples were digested [11] and the concentration of heavy metals in digested solution was quantified by using an atomic absorption spectrophotometer (Shimadzu AA-7000). The overall analyses were followed in accordance with a general method described by Ara et al and Nazah et al [12, 13]. All data were calculated by using the Microsoft Excel 2010 and Statistical Package for Social Sciences (SPSS) version 16.0.

Sl. No.	Local Name	Common Name	Scientific Name	Sample ID	Vegetable Type
1	Lal Shak	Red Spinach	Amaranthus gangeticus	LS	Leafy Vegetable
2	Palong Shak	Spinach	Spinacia oleracea	PS	Leafy Vegetable
3	Kalmi shak	Water spinach	Ipomoea aquatica	KS	Leafy Vegetable
4	Fulkopi	Cauliflower	Brassica oleracea botrytis	FK	Fruit Vegetable
5	Tomato	Tomato	Solanum lycopersicum	ТО	Fruit Vegetable
6	Misti Kumra	Sweet gourd	Cukurbita pepo	MK	Fruit Vegetable
7	Papa	Рарауа	Carica papaya	PP	Fruit Vegetable
8	Begoon	Egg Plant	Solanum melongena	BG	Fruit Vegetable
9	Olkopi	Kholrabi	Brassica oleracea	OK	Root Vegetable
10	Mula	Radish	Raphanus raphanistrum	MU	Root Vegetable

Table 1: Description of the studied vegetables cultivated in Satkhira area

2.2 Estimated Daily Intake (EDI)

Daily intake of contaminated vegetables is a general pathway of heavy metal exposure to the human. EDI of heavy metals from these foods can be calculated by using the equation [12, 14]:

$$EDI = \frac{H_{mc} \times R_i}{B_{m}} \tag{1}$$

Where H_{mc} is the concentration of heavy metal (mg kg⁻¹ dry weight); R_i denotes the rate of ingestion that was considered as 0.126 kg/day for vegetables [15, 16] and B_w is the average body weight of the people. In this study, B_w was considered 49.5 kg for Bangladeshi people [12, 17].

2.3 Target Hazard Quotient (THQ)

THQ was calculated by the following formula [12, 14]:

$$THQ = \frac{EDI \times E_f \times D_e}{D_f \times T_{avncar}}$$
(2)

Where, THQ represents non-cancer risks, E_f denotes the exposure frequency (365 days year⁻¹); D_e denotes exposure duration (71.8 years); Reference dose (D_f) of Fe, Mn, Cu, Zn, Pb and Cd are 0.7, 0.14, 0.04, 0.30, 0.0035 and 0.003 (mg kg⁻¹day⁻¹) respectively [18–20]; and T_{avncar} implies average time for non-carcinogens (365 days year⁻¹× D_e) [19].

2.4 Hazard Index (HI) and Target Cancer Risk (TCR)

Hazard Index is the sum of Hazard quotients of all metals. It was calculated by the formula [18, 21].

$$HI = \sum THQ = THQ_{(Mn)} + THQ_{(Fe)} + THQ_{(Cu)} + THQ_{(Zn)} + THQ_{(Pb)} + THQ_{(Cd)}$$
(3)

TCR was estimated by using the equation [19]:

$$TCR = THQ \times S_{cpo}$$
(4)

The reference values of carcinogenic potency slope (S_{cpo}) of Pb and Cd are 0.0085 and 6.1 mg kg⁻¹ body weight days⁻¹ respectively [18].

3. Results and Discussion

Vegetables are a major constituent of the human diet, playing a significant role in the prevention of fatal diseases. Unfortunately, these essential foods are polluted by heavy metals that have become an alarming issue across the globe. Heavy metals are found in the earth crust and they can exist in the environment for a long time without any biodegradation. Some heavy metals, for example, Mn, Fe, Cu, and Zn are essential micronutrient for biological functions of the human body. On the other hand, Cd and Pb are not essential for a living being, therefore, they are considered as toxic elements in nature. To assess the health risks of people in Satkhira, Bangladesh, the concentration of heavy metals in commonly consumed vegetables and cultivated soil was estimated and displayed in Table 2. The average concentration of Mn, Fe, Cu, Zn, Cd, and Pb was 33.91, 356.71, 10.27, 33.59, 0.57 and

9.76 mg kg⁻¹ in vegetables. In this study, the decreasing order of heavy metals in leafy, fruit and root vegetables were Fe>Mn>Zn>Cu>Pb>Cd, Fe>Zn>Mn>Cu>Pb>Cd and Fe>Mn>Zn>Pb >Cu>Cd respectively. The concentration of Mn, Fe, and Cu (except Zn, Cd, and Pb) was higher in leafy vegetables than fruit and root vegetables. Iron is an essential element for the physiological functions in the human body like hemoglobin formation. The tolerable limit is beneficial for the human but excessive amount (above 450 mg kg⁻¹) may cause gastrointestinal side effects. In addition, the highest amount of Fe was found in all of the vegetables and varied from 99.25 to 1661.30 mg kg⁻¹ depending on the vegetable species. Usually, Fe takes part in photosynthesis and chlorophyll synthesis. As a result, leafy vegetables contain higher Fe than fruit and root vegetables [11, 17].

Sample ID	Mn	Fe	Cu	Zn	Cd	Pb
LS	22.073	867.150	14.452	27.491	0.724	13.151
	9.10-44.14	219.7-1126.5	4.61-52.34	11.82-52.68	0.20-0.99	6.20-18.11
PS	32.109	685.166	13.493	40.33	0.708	12.02
	10.19-99.02	105.24-1211.8	0.89-22.28	19.72-95.54	0.44-0.85	5.13-18.17
KS	87.501	575.219	15.424	17.110	0.481	7.91
	22.20-232.1	109.11-1017.6	6.21-21.82	9.27-28.36	0.09-0.59	4.51-14.25
FK	8.570	322.041	2.989	38.108	0.390	3.452
	4.18-15.17	99.53-521.21	1.71-6.73	10.65-49.89	0.05-0.56	0.44-5.20
ТО	24.501	455.135	8.758	30.701	0.625	11.363
	11.42-42.23	109.6-1306.14	1.43-19.66	16.24-45.26	0.22-0.97	8.13-37.03
MK	28.041	875.3926	10.551	42.212	0.341	13.136
	9.67-44.26	321.93-1034.5	2.63-19.16	30.12-49.72	0.13-0.73	6.04-15.12
PP	9.013	267.132	8.102	40.976	0.532	5.204
	2.01-16.43	91.67-317.05	2.34-11.42	12.51-74.35	0.23-1.05	0.75-5.69
BG	18.512	499.734	11.7885	33.531	0.231	1.622
	4.04-49.12	99.5-1144.27	3.62-18.43	13.27-44.35	0.10-0.38	0.49-4.25
OK	38.323	309.201	7.751	37.198	0.579	13.350
	9.45-101.21	100.26-477.87	1.18-14.22	25.19-49.32	0.32-0.96	6.22-16.29
MU	35.244	244.913	8.056	33.509	0.713	9.231
	11.45-69.96	132.27-644.54	1.12-14.81	23.46-59.46	0.22-0.84	0.53-14.05
Av. conc. in LV	47.23	709.18	14.46	28.31	0.64	11.03
Av. conc. in FV	17.73	483.89	8.44	37.11	0.42	6.96
Av. conc. in RV	36.78	277.06	7.90	35.35	0.65	11.29
Average	33.91	356.71	10.27	33.59	0.57	9.76
Range	2.01-232.1	91.67-1306.14	0.89-52.34	9.27-95.45	0.05-1.05	0.44-37.03

Table 2: Average concentration of heavy metals (mg kg⁻¹) in vegetables

Where, Av= Average, Conc.= Concentration, LV= Leafy Vegetables, FV= Fruit Vegetables, RV= Root Vegetables

Pb and Cd can easily accumulate in soil due to its low solubility and can persist in the environment for a long time. The higher concentration of Pb (37.03 mg kg⁻¹) and Cd (1.05 mg kg⁻¹) was observed in leafy and root vegetables. Exposure to lead may cause autoimmune disorders such as rheumatoid arthritis, kidney diseases, etc [8]. To assess the pollution level, the concentrations of heavy metal were compared with the reference value recognized by FAO/WHO [22–24] and other similar studies (Table 3). According to Figure 2, it can be seen that the concentration of Fe, Cd, and Pd in vegetables was higher than the permissible limit. The average concentration of Cd was comparatively higher than other similar studies except for Varanasi, India and Rajshahi, Bangladesh. In addition, the average concentration of Pb was observed higher than almost all other studies. This can be explained due to different sources like fumes from automobile exhausted, dry cell batteries, sewage effluents, run off of wastes and atmospheric depositions, high vehicular traffic. Besides, the application of organic and inorganic fertilizers, fungicides, pesticides, manure and bio-solids in relevant fields may contribute the level of these heavy metals (26, 27).

The carcinogenic and non-carcinogenic health risks were calculated based on equation (1-4) and the results of EDI, HI, THQ and TCR are presented in Table 4 [22]. The comparison of EDI values of heavy metals with the respective references dose (D_f) revealed that the EDI values of Pb and Cd were greater than D_f for all vegetables, while the EDI values of Fe was observed to be greater than D_f only for leafy and fruit vegetables. In this regard, the New York State Department of Health (NYSDOH) suggested if the ratio of EDI/ D_f is less than or equal to the D_f , the risk will be minimal. But if this ratio is >1–5 times than the D_f then risk will be low if it is >5–10 times than the D_f , the risk would be moderate and if >10 times than the D_f , the risk will be high [34]. In this study, the ratio (EDI/ D_f) of Pd for all types of vegetables was several hundred times higher than corresponding D_f , indicates a potential health risk.



Fig. 2: Comparison of contamination status of heavy metals in vegetables with the standard value

THQ is the measure of the possibility of developing non-carcinogenic health problems and the acceptable guideline value for THQ is ≤ 1.0 [19]. According to Ambedkar and Maniyan, if the value of THQ for individual heavy metal exceeds its tolerable limit, it might pose non-carcinogenic health effects to the human [35]. In this study, THQ values of the most of heavy metals were lower than the permissible limit 1.0, except Fe (2.5788) and Pb (8.0196) for leafy vegetables; Fe (1.7596) and Pb (5.0581) for fruits vegetables; Fe (1.0074) and Pb (8.2116) for root vegetables. Therefore, the THQ analysis suggested that Fe and Pb might pose a non-carcinogenic health risk to the inhabitants in this region. Furthermore, the combined impact of all studied metals (HI) was higher than the acceptable limit (1.0) for all types of vegetables. Therefore, intake of leafy, fruit and root vegetables on a

regular basis is a matter of concern for non-carcinogenic health risks. The percentages of non-carcinogenic risk of different heavy metals were calculated and displayed in Figure 3 (a-c). In this study, the percentages of Fe, Mn, Cu, Zn, Cd and Pb was observed 20%, 6%, 7%, 4%, 61% in leafy vegetables, 21%; 3%, 6%, 3%, 4% and 61% in fruit vegetables; 9%, 6%, 4%, 3%, 5% and 73% in root vegetables.

Location	Fe	Mn	Cu	Zn	Cd	Pb	Ref.
Jashore	190.32	33.29	9.31	28.23	0.514	5.45	[12]
	69.52-446.68	11.33-130.18	1.12-30.80	12.52-47.76	0.24-0.77	0.61-14.79	
Dhaka	1160.0		17.63		0.32	11.48	[28]
	1126-1209		10.27-29.1		0.216-0.40	7.32-17.0	
Rajshahi		4.54			1.05	5.1	[16]
		1.36-8.11			0.15-1.75	0.43-1.37	
Patuakhali			42.35		2.0	8.35	[29]
			4.12-529.42		0.011-25.88	0.47-103.53	
Narayanganj			9.37	19.76	0.168	3.69	[30]
			3.45-14.35	12.79-27.22	0.095-0.283	2.16-5.50	[]
Misurata			3.36	8.15	0.14	0.25	[31]
			0.75-5.75	0.042-16.83	0.02-0.27	0.02-0.511	
Changxing					0.035	0.039	[32]
					0.003-0.230	0.003-0.178	L- J
Varanasi			22.38	48.62	1.51	1.15	[22]
			9.50-56.30	25.20-94.30	0.10-4.30	0.20-2.56	[33]
Satkhira	356.71	33.91	10.27	33.59	0.57	9.76	Present
	99.5-1211.8	2.01-232.1	0.89-52.34	9.27-95.45	0.05-1.05	0.44-37.03	Study

Table 3: Comparison of the level (mg kg⁻¹ dry weight) of heavy metals in vegetables with similar findings

Table 4: EDI, THQ, HI and TCR of leafy, fruit and root vegetables

Parameters	Sample types	Fe	Mn	Cu	Zn	Cd	Pb
EDI	Leafy Vegetable	1.8052	0.1202	0.0368	0.0721	0.0281	0.0016
	Fruit Vegetable	1.2317	0.0451	0.0214	0.0944	0.0011	0.0177
	Root Vegetable	0.7052	0.0936	0.0201	0.0899	0.0016	0.0287
THQ	Leafy Vegetable	2.5788	0.8587	0.9199	0.2402	0.5413	8.0196
	Fruit Vegetable	1.7596	0.3223	0.5369	0.3148	0.3597	5.0581
	Root Vegetable	1.0074	0.6688	0.5029	0.2999	0.5481	8.2116
HI	Leafy Vegetable			13.	1586		
	Fruit Vegetable			8.3	516		
	Root Vegetable			11.	2389		
TCR	Leafy Vegetable	-	-	-	-	3.3E ¹	6.8E ⁻²
	Fruit Vegetable	-	-	-	-	$2.1E^{1}$	4.2E ⁻²
	Root Vegetable	-	-	-	-	3.3E ¹	6.9E ⁻²

The TCR value denotes not only an estimation of expected cancer but also it represents the probability of developing carcinogenic risk to the human [34]. In this study, the possibility of developing cancer was calculated based on the USEPA deterministic approach. Prolonged exposure of a specific carcinogen may develop cancer and the probability increases with the contact time. According to NYSDOH the TCR categories are described as; if the TCR value is less than equal to 10^{-6} the carcinogenic risk is low; when this value is 10^{-5} to 10^{-3} the risk is moderate; but when this value is 10^{-3} to 10^{-1} and $\ge 10^{-1}$ the risk is high and very high [34]. In this study, the TCR of Pb ($6.8E^{-2}$, $4.2E^{-2}$ and $6.9E^{-2}$ for leafy, fruit and root vegetables respectively) showed high cancer risk whereas Cd ($3.3E^{1}$, $2.1E^{1}$, and $3.3E^{1}$ for leafy, fruit and root vegetables respectively) posed a very high cancer risk to the population. Thus, the TCR of Pb and Cd for study area people is a matter of concern.



Figure 3: % of the non-carcinogenic risk of heavy metals in (a) leafy (b) fruits and (c) root vegetables

Sampling Sites	Mn	Fe	Cu	Zn	Cd	Pb
L-01	192.62	3750.78	31.14	49.50	0.71	10.90
L-02	178.67	3775.67	36.62	53.78	0.83	9.71
L-03	214.13	3968.30	16.81	82.38	0.61	13.45
L-04	272.40	3978.97	18.56	62.85	0.75	12.13
L-05	282.89	3863.20	23.20	73.73	0.83	10.43
L-06	266.92	3982.62	19.72	65.77	0.42	12.32
L-07	251.17	3946.98	20.53	62.81	0.69	9.03
L-08	243.50	3859.15	25.65	71.86	0.54	13.71
L-09	223.21	3988.84	15.43	89.77	0.70	11.45
L-10	267.90	2879.35	17.19	43.81	0.75	12.16
Mean	239.34	3799.38	22.48	65.63	0.68	11.53
Reference value ^[17]	270	40000	30	100	1	50

Table 5: Amount of heavy metals (mg kg⁻¹) in soil sample of Satkhira, Bangladesh

Table 6: Comparison of the average concentration of heavy metals (mg kg⁻¹) in growing soil with similar studies [12]

Study Area	Mn	Fe	Cu	Zn	Cd	Pb	Ref.
Entire Bangladesh	669.56	37247.15	54.29	202.81	1.26	9.4	[17]
Jashore, Bangladesh	199.38	3773.29	11.85	49.58	0.68	12.61	[12]
Iswardi, Bangladesh	283.50	15684.70	21.43	123.283	0.538	68.84	[28]
Chittagong, Bangladesh	160.79		32.63	139.30	2.43	7.33	[36]
Mymensingh, Bangladesh	182.33	24683.33	49.10	123.19		59.39	[37]
Bogra, Bangladesh			131.87	28.46	6.95	9.6	[38]
Gazipur, Bangladesh			36.18	176.66	0.2	75	[39]
Dhaka, Bangladesh			75.04	103.34	0.52	3.84	[40]
Dhaka, Bangladesh		1715.80	39.14	115.43	11.42	49.71	[1]
Rosetta, Egypt		65387.8	92.8	156.3	13.5	45.7	[41]
West Africa		6224.92		275.57	1.42	28.76	[42]
Nador, Morocco	263.00	76429.00	37.50	73.00			[43]
Palermo, Italy	519.00		63.00	138.00	0.68	202	[44]
Sialkot, Pakistan		17991.62	26.85	94.2	36.8	121.4	[45]
Fuyang, China			40.77	159.85	0.37	40.59	[46]
Satkhira, Bangladesh	239.34	3799.38	22.48	65.63	0.68	11.53	Present study

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To identify the possible sources of contamination the concentration of heavy metals in soil was investigated and estimated as Fe (3799.385), Mn (239.341), Cu (22.483), Zn (65.629), Cd (0.681) and Pb (11.529) mg kg⁻¹ respectively (Table 5). The decreasing order of heavy metal content was Fe > Mn > Zn > Cu > Pb > Cd. Besides, the level of individual heavy metals in soil samples was compared to the standard value summarized by Mahfuza et al [17] (Table 5). According to the pollution level, the concentration of Fe, Cu, Zn, Cd, and Pb were observed within the permissible limit except for Mn in L-04 and L-05. Moreover, the average concentration of heavy metals in studied soils has been compared with other observations in our country as well as other countries (Table 6). This study showed that the average concentration of Fe, Cu, and Zn in soil samples were comparatively lower than various locations in Bangladesh as well as other countries whereas Mn, Cd, and Pb showed few exceptions.

4. Conclusion

From this study, it was possible to establish a database about the contamination status of heavy metals in popular vegetables and their growing soil in Satkhira, Bangladesh. Although the concentration of heavy metals in soil was within the permissible limit but the concentration of Fe, Cd, and Pb in vegetable were higher than the safe limit recognized by joint FAO/WHO. The higher concentration of heavy metal in vegetables might be due to anthropogenic activities like deposition from vehicle emission, excessive use of wastewater, fertilizer, and pesticides in agricultural fields. To assess the health risk of consumers, different health risk indices (EDI, THQ, HI, and TCR) were calculated which revealed moderate to high health risk among the population. Intake of contaminated vegetables is the biggest concern for carcinogenic and non-carcinogenic health effects. Therefore, regular monitoring and effective steps should be taken to prevent the entrance of heavy metals in the food chain.

5. Acknowledgments

None

6. Conflict of Interests

The authors declare no conflict of interest

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