

Health Risks Assessment of Heavy Metals in Vegetables Collected from Bagerhat District

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ABSTRACT

This study aimed to determine the concentration of heavy metals (Cu, Zn, Cd, and Pb) in popular vegetables cultivated in Bagerhat, Bangladesh, and to assess the associated health risks of local inhabitants. The decreasing sequence of the concentrations (mean and range) of studied heavy metals in all types of vegetables was as follows: Cu (12.251, 0.161-43.216) > Zn (10.311, 0.187-35.822) > Pb (1.036, 0.032-3.015) mg kg⁻¹ fresh weight. The concentrations of heavy metal were also compared with the reference value and it was found that the average concentrations of Pb in most of the studied vegetable samples exceeded the permissible limit, which was a matter of health issue. Significant positive correlations were found in Cu-Zn ($r=0.753$), Cu-Cd ($r=0.822$), Cu-Pb ($r=0.612$), Zn-Cd ($r=0.725$), and Cd-Pb ($r=0.729$) at $p < 0.05$ significant level; indicating a common source of pollution. Various health risks assessment parameter like target cancer risks (TCR) of Pb was below the standard guideline value (10^{-4}) but the non-carcinogenic health risks assessment parameter (HI) was above the standard limit (1.0), which revealed that the combined impact of all heavy metals was a matter of health concern for the local inhabitants. Therefore, to avoid various health problems, regular monitoring is strongly recommended.

Keywords: Heavy metals, Risk assessment, THQ, TCR, HI, Vegetables, Bangladesh

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I. Introduction

Plants play an essential role in human life [1]. Vegetables are a major constituent of the human diet owing to the high contents of vitamins, minerals, trace elements, cellulose, hemicellulose, pectin, and fiber [2, 3]. In addition, vegetables are playing a pivotal role in the prevention of cancer and reducing the risk of cardiovascular and other chronic diseases [1]. Unfortunately, these essential foods are being polluted by heavy metals, and intake of contaminated vegetables might exert deleterious health effects on humans such as like damage of DNA, change the genetic code and reduce the energy level of the human body. Besides, heavy metal inhibits the common functions of the liver, kidneys, lungs, and heart, etc. [4, 5-8]. Heavy metals generally combine with the thiol, amino, and imino group of protein, and form the metal complex. As a result, the proteins lose their biological activities and cause the breakdown of the cell [10/9]. Heavy metals also affect the physiological functions of plants. They retard the nitrogen fixation, chlorosis, metabolism, and growth of plants [9]. Heavy metals are non-biodegradable and could persist for a long time in the environment.

At present, heavy metal pollution has become one of the most serious environmental problems not only in Bangladesh but also all over the world [10]. Due to the rapid growth of industrialization, use of different fertilizers, pesticides, and herbicides in agricultural fields the cultivated vegetables can be contaminated with heavy metals. Besides, random disposal of household wastes, livestock manure, and unused metallic parts are the main causes of heavy metal deposition in vegetables. Moreover, heavy metals are also deposited on the different parts of vegetables in the air [11]. In this context, the risks associated with the consumption of contaminated vegetables may be a potential health concern.

Recently, many countries have launched regular monitoring and assessment of heavy metals in food and vegetables. But there is insufficient data available for the contamination level of heavy metals in Bagerhat, Bangladesh. This study, therefore, aimed to determine the concentration of heavy metals (Cu, Zn, Cd, and Pb) in commonly consumed vegetables and to assess the health risks of consumers through the analysis of carcinogenic and non-carcinogenic risk indices analysis. Health risks have been evaluated by numerous methods but most commonly, the risk to human health is computed in terms of the target hazard quotient (THQ) which is based on the concentration of trace metals in the edible parts of vegetables, in comparison with the reference dose of the metal intake and body weight of the consumers [12].

II. Materials And Methods

2.1 Description of the study area

Bagerhat District is located in between 21°49' and 22°59' north latitudes and in between 89°32' and 89°98' east longitudes. It has an area of area 3959.11 sq km, is bounded by Gopalganj and Narail districts on the north, Bay of Bengal on the south, Gopalganj, Pirojpur, and Barguna districts on the East, Khulna district on the west [13]

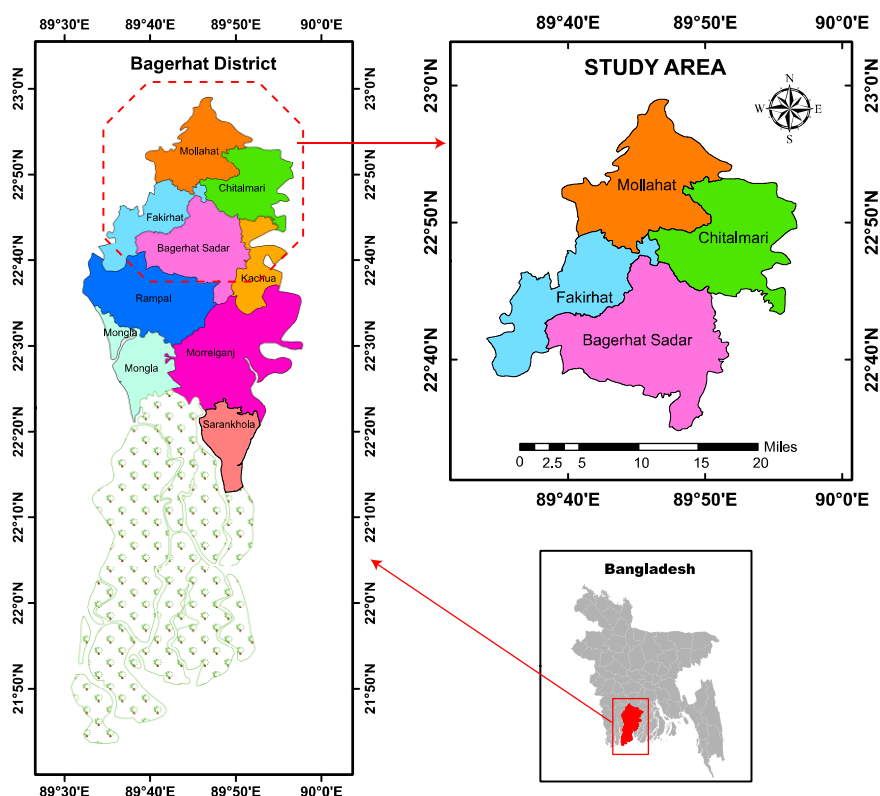


Figure 1: Map of the study area (Mollahat, Chitalmari, Fakirhat, and Bagerhat Sadar sub-districts)

2.2 Sample Collection

In this study, various types of leafy, fruit, and root vegetables were randomly collected from four sub-districts (Mollahat, Chitalmari, Bagerhat Sadar, and Fakirhat) of Bagerhat District, Bangladesh. **Table 1** shows the general description of the studied vegetables. Each vegetable sample was washed thoroughly with tap water followed by distilled water to remove dust. The edible part of the samples was cut into small pieces, air-dried for 2 days, and kept in a hot air oven at 100 ± 1 °C for 4 h to attain constant weight.

Table 1: General description of the studied vegetables cultivated in Sub-districts of Bagerhat

Sl. No.	Local Name	Common Name	Scientific Name	Sample ID	Vegetable Type
1	Lal Shak	Red Spinach	<i>Amaranthus gangeticus</i>	LS	Leafy Vegetable
2	Palong Shak	Spinach	<i>Spinacia oleracea</i>	PS	Leafy Vegetable
3	Fulkopi	Cauliflower	<i>Brassica oleracea botrytis</i>	FK	Fruit Vegetable
4	Misti Kumra	Sweet gourd	<i>Cukurbita pepo</i>	MK	Fruit Vegetable

5	Olkopi	Kholrabi	<i>Brassica oleracea</i>	OK	Root Vegetable
6	Mula	Radish	<i>Raphanus raphanistrum</i>	MU	Root Vegetable

2.3 Sample preparation and analysis

Analytical grade (Wako, Japan, and Merck, Germany) reagents were purchased from the local market and used for all purposes without any further purification. Each dry sample was digested by adopting the wet digestion method described by Allen et al., [14]. Briefly, 15 mL of 5:1:1 tri-acid mixture (70% HNO₃, 70% H₂SO₄, and 65% HClO₄) was added to each beaker containing 1 g dry sample [14]. The mixture was subjected to digest at 80 °C until the transparent solution appeared. After cooling, the digested samples were filtered using Whatman No. 42 filter paper and poured into a Teflon bottle and finally, the filtrate was diluted to 50 mL with deionized water for heavy metal analysis [14]. The concentrations of heavy metals (Cu, Zn, Pb, and Cd) in the digested vegetable samples were analyzed by using atomic absorption spectrophotometer (Shimadzu model AA-7000, made by Japan). The detected trace metals were converted to fresh weight from dry weight by using a conversion factor (0.085) for all the presented data. This conversion was carried out because people consume the vegetable on a fresh weight basis, not on a dry weight basis. Generally, fresh vegetables contain a lower quantity of heavy metals than dry vegetables because of moisture contents. So, the concentration of 1.0 mg/kg of heavy metal on a dry weight basis is equal to 0.085 mg/kg of heavy metals on a fresh weight basis. The determined values of heavy metals in vegetables were used to assess the health implications of the human.

2.3 Health risks assessment

USEPA Region III risk-based deterministic model was employed to assess the non-carcinogenic health risk and carcinogenic health risk of local consumers.

2.3.1 Non-carcinogenic health risks assessment

The degree of toxicity of hazardous metals depends on their daily intake value. In this study, the EDI of metals was determined based on the metal concentrations, the daily ingestion rate of vegetables, and the average body weight of the consumers. Daily intake of contaminated vegetables is a general pathway of heavy metal exposure to humans. EDI of heavy metals from these foods can be calculated by using the equation [4, 14]. Where MC is the concentration of heavy metal (mg kg⁻¹ fresh weight); RI denotes the rate of Ingestion that was considered as 0.126 kg/day for vegetables [15], and BW is the average body weight of the people. In this study, BW was considered 49.5 kg for Bangladeshi people [4, 15-16]. Target hazard quotient (THQ) was calculated by the following formula [4, 17]:

$$EDI = \frac{MC \times RI}{BW} \quad (1)$$

$$THQ = \frac{EDI}{RFD} \quad (2)$$

Where, THQ represents non-cancer risks, Reference dose (RFD) of Pb, Cu, Zn, and Cd, were considered 0.004, 0.04, 0.30, and 0.001 mg kg⁻¹day⁻¹, respectively [18]. Hazard Index (HI) is the sum of hazard quotients of all metals. It was calculated by the formula [4, 15].

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

2.3.2 Carcinogenic health risks assessment

Target Cancer Risk (TCR) is conceptualized as the possibility of developing cancer risk during one's lifetime due to overexposure to a specific carcinogenic substance. It can be estimated by using the equation (4) [4, 15]:

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

The reference values of carcinogenic potency slope (S_{cpo}) Pb is 0.0085 (mg kg⁻¹day⁻¹)⁻¹ [18].

2.3.3 Statistical analysis

All statistical analyses were performed by using Statistical Package for Social Sciences (SPSS) version 16.0 and Microsoft Excel 2010.

III. Results and Discussion

3.1. Heavy metal concentration in studied vegetables

The determined concentration of heavy metals in leafy (LS, PS), fruits (FK, MK), and root (OK, MU) vegetables are shown in **Table 2**. The decreasing sequence of the concentrations (mean and range) of studied heavy metals in all types of vegetables was as follows: Cu (12.251, 0.161-43.216) > Zn (10.311, 0.187-35.822)

> Pb (1.036, 0.032-3.015) mg kg⁻¹ fresh weight. The mean highest amount of Cu, Zn, Cd, and Pb was found in MK, FK, LS, and OK, respectively whereas the lowest concentration was observed in MU for all metals (except Pb in FK).

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

Sample types	Sample ID	Cu	Zn	Cd	Pb
Leafy vegetables	LS	7.093 (3.161-13.814)	7.785 (3.232-11.811)	0.087 (0.05-0.132)	1.628 (0.814-3.015)
	PS	11.602 (2.465-24.102)	11.928 (2.933-23.279)	0.060 (0.031-0.143)	1.022 (0.411-2.891)
	Mean	9.347	9.856	0.073	1.325
Fruit vegetables	FK	13.004 (6.224-25.712)	21.089 (7.612-35.822)	0.076 (0.035-0.182)	0.293 (0.091-0.782)
	MK	23.847 (9.174-43.216)	15.488 (5.528-27.327)	0.071 (0.042-0.136)	0.947 (0.236-1.887)
	Mean	14.425	18.288	0.074	0.62
Root vegetables	OK	10.859 (4.033-35.617)	4.862 (2.113-9.219)	0.083 (0.053-0.176)	1.815 (0.317-2.718)
	MU	0.685 (0.161-1.644)	1.998 (0.187-2.991)	0.033 (0.021-0.112)	0.088 (0.032-0.182)
	Mean	5.77	3.43	0.058	0.951
Mean concentration (Range)		12.251 (0.161-43.216)	10.311 (0.187-35.822)	0.072 (0.021-0.182)	1.036 (0.032-3.015)

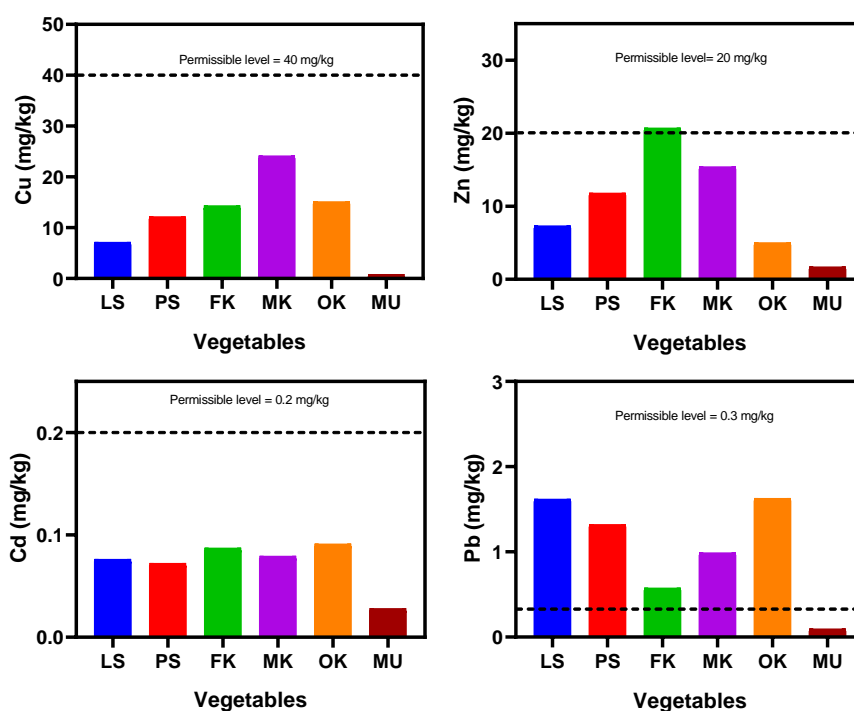


Figure 2: Comparison of heavy metals concentration in vegetables with the standard permissible value

According to **Table 2**, it could be seen that the concentration of Cu, Zn, and Cd was comparatively higher in fruits vegetables than leafy and root vegetables. On the other hand, the level of Pb was found to be higher in leafy vegetables than fruit and root vegetables. However, the mean concentrations of heavy metals in different types of vegetables had been compared with the reference value recognized by FAO/WHO to know the pollution level in this area and was shown in **Figure 2** [19-20]. The concentration of Zn in FK and Pb in LS, PS, FK, MK, and OK was higher than the permissible limit of Zn (20 mg kg⁻¹) and Pb (0.3 mg kg⁻¹). Higher concentration Pb was probably found due to deposition from vehicle emission and different metal factories [21].

Moreover, the accumulation of higher content of heavy may be due to the application of an excessive amount of inorganic fertilizer, different types of pesticides, and wastewater in agricultural fields [22, 23].

The obtained results were compared with similar studies and shown in **Table 3**. The mean concentration of Cu ($12.251 \text{ mg kg}^{-1}$) was similar to the obtained result in Dhaka (18.1 mg kg^{-1}) and comparatively lower than the other parts of Bangladesh. Similarly, the concentration of Zn ($10.311 \text{ mg kg}^{-1}$) was much lower than the reported value of Zn in Dhaka (51.2 mg kg^{-1}) but higher than that of other studies. The value of Cd was almost similar to other studies but the value of Pb (1.036 mg kg^{-1}) was slightly higher than the concentration of Pb in Jashore, Dhaka, Noakhali, Narayanganj, and Satkhira districts of Bangladesh.

Table 3: Comparison of the level (mg kg^{-1} fresh weight) of heavy metals in vegetables with other findings

Location	Cu	Zn	Cd	Pb	Ref.
Jashore	0.791 (0.095-2.618)	2.4 (1.064-4.059)	0.044 (0.02-0.065)	0.463 (0.051-1.257)	[4]
Dhaka	18.1 (8.30-34.3)	51.2 (16.3-119)	0.21 (0.009-1.05)	0.76 (0.06-3.45)	[24]
Noakhali	20.6 (2.1-86.3)	-	0.058 (0.006-0.265)	3.7 (0.67-16.5)	[25]
Narayanganj	0.796 (0.293-1.219)	1.679 (1.087-2.313)	0.0142 (0.008-0.024)	0.313 (0.184-0.467)	[26]
Satkhira	0.873 (0.075-4.448)	2.885 (0.787-8.113)	0.048 (0.00425-0.089)	0.83 (0.037-3.147)	[15]
Bagerhat	12.251 (0.161-43.216)	10.311 (0.187-35.822)	0.072 (0.021-0.182)	1.036 (0.032-3.015)	Present study

3.2. Pearson's correlation matrix analysis

To identify the possible sources of origin of heavy metals, Pearson's correlation matrix in vegetable samples was analyzed (**Figure 2**). Significant positive correlations were found in Cu–Zn ($r=0.753$), Cu–Cd ($r=0.822$), Cu–Pb ($r=0.612$), Zn–Cd ($r=0.725$), and Cd–Pb ($r=0.729$) at $p < 0.05$ significant level; indicating a common source of pollution. Subsequently, no correlation was observed in Pb–Zn ($r=0.348$), which suggests that the originating source was different for these metals.

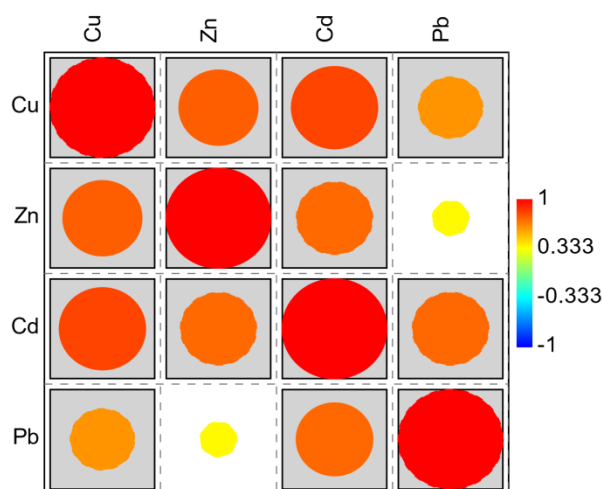


Figure 3: Pearson's correlation among the studied metals (at $p < 0.05$ significant level)

3.3. Health risks assessment

The carcinogenic (TCR) and non-carcinogenic health risks (EDI, THQ, HI) were analyzed based on USEPA deterministic method, and the obtained values were presented in **Table 4**. In this study, the mean EDI value of Cu, Zn, Cd, and Pb in LS, PS, FK, MK, OK, and MU were observed 0.0181, 0.0309, 0.0364, 0.0614, 0.0384, and 0.0019; 0.0186, 0.0300, 0.0527, 0.0392, 0.0127, and 0.0043; 0.0186, 0.0300, 0.0527, 0.0392, 0.0127, and 0.0043; 0.0041, 0.0034, 0.0015, 0.0025, 0.0041, and 0.0002 respectively. Besides, these values were compared with the respective reference dose D_f . 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 it 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 [27]. It was observed that the EDI values of all heavy metals were below than D_f for all studied vegetables therefore the study people are safe from potential health hazards. The THQ values

for individual heavy metals via consumption of leafy, fruit, and root vegetables were presented in **Table 4** and the acceptable guideline value for THQ is ≤ 1.0 [18, 14].

Table 4: EDI ($\text{mg kg}^{-1}\text{day}^{-1}$) of leafy, fruit and root vegetables

Sample types	Sample ID	Cu	Zn	Cd	Pb
Leafy vegetables	LS	0.0181	0.0186	0.0002	0.0041
	PS	0.0309	0.0300	0.0002	0.0034
	FK	0.0364	0.0527	0.0002	0.0015
Fruit vegetables	MK	0.0614	0.0392	0.0002	0.0025
	OK	0.0384	0.0127	0.0002	0.0041
Root vegetables	MU	0.0019	0.0043	0.0001	0.0002
	RFD	0.04	0.3	0.001	0.004

Table 5: THQ, HI, and TCR of leafy, fruit, and root vegetables

Sample types	Sample ID	THQ				HI	TCR
		Cu	Zn	Cd	Pb		
Leafy vegetables	LS	0.451	0.062	0.193	1.029	1.735	3.50E-05
	PS	0.771	0.100	0.183	0.839	1.894	2.85E-05
	FK	0.911	0.176	0.222	0.365	1.673	1.24E-05
Fruit vegetables	MK	1.534	0.131	0.201	0.628	2.494	2.13E-05
	OK	0.961	0.042	0.231	1.035	2.268	3.52E-05
Root vegetables	MU	0.049	0.014	0.071	0.060	0.194	2.03E-06

According to Ambedkar and Maniyan if the concentrations of heavy metal are above the tolerable level, recommended by regulatory agencies and depending on daily intake by consumers, might pose a health impact [35/28]. However, THQ values of Zn and Cu were less than 1.0 for all types of vegetables but THQ of Cu in MK (1.534) for fruit vegetables and Pb in LS (1.029) for leafy vegetables, and in OK (1.025) were slightly higher than the recommended value of 1.0. Besides this, the combined impact of almost all metals (HI) was higher than the acceptable limit of 1.0 for all types of vegetables (except MU in root vegetables). Therefore, intake of these leafy, fruit, and root vegetables on the regular basis is a matter of concern for non-carcinogenic health risks. Furthermore, the percentages of THQs of heavy metals due to studied vegetables in the Bagerhat, Bangladesh were displayed in **Figure 4**. The highest contributors of THQs of Cu, Zn, Cd, and Pb were MK (33%), FK (33%), FK (20%), and PS (21%) vegetables, respectively, and while the lowest contributors were MU for all heavy metals. However, the decreasing sequence of HI for all metals in studied vegetables was as follows: MK (24%) > OK (22%) > PS (19%) > LS (17%) > FK (16%) > MU (2%)

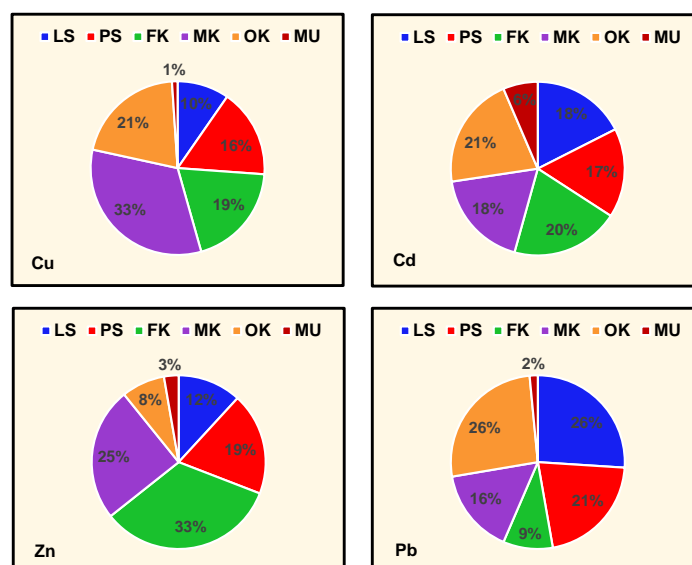


Figure 4: % of the non-carcinogenic risk of heavy metals in different types of vegetables

Prolonged exposure to a specific carcinogen may develop cancer and the probability increases with the contact time. The TCR value denotes not only an estimation of expected cancer but also represents the probability of developing carcinogenic risk to the human [34]. According to the USEPA, the TCR categories are

described as; if the TCR value is less than equal to 10^{-4} , there is no carcinogenic risk, when this value exceeds this threshold limit of 10^{-4} the risk is high [34]. Due to the absence of S_{cpo} the TCR of Cu, Zn and Cd were not calculated. In this study, the TCR of Pb was observed 10^{-5} to 10^{-6} , which was below the maximum risk limit of 10^{-4} (Figure 5). This finding revealed that the study people were safe from any carcinogenic health risks due to the consumption of leafy, fruits, and root vegetables from this region. However, the TCR of Fe, Zn, Mn, Cu, and Pb for study area people is a matter of concern. The decreasing sequence of carcinogenic risk of Pb in studied vegetables was as follows: OK (26.1%) > LS (26%) > PS (21%) > MK (16%) > FK (FK) > MU (2%).

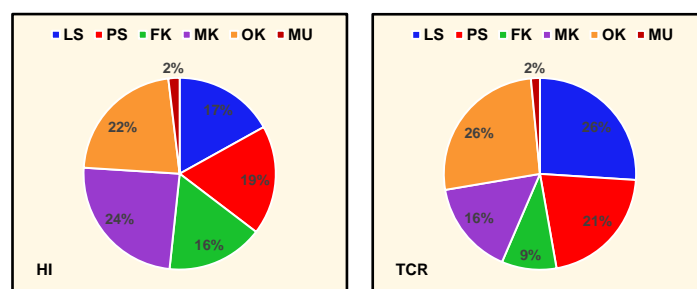


Figure 5: Contribution of different types of vegetables in HI and TCR

IV. Conclusions

In this study the concentration of heavy metals in commonly consumed vegetables in Bagerhat, Bangladesh was determined and their health risks were assessed. Although the concentration of heavy metals in most of the samples was within the permissible limit, the concentrations of Pb in all vegetables were higher than the safe limit recognized by joint FAO/WHO. Higher Pb content in vegetables might be due to deposition from vehicle emission, excessive use of wastewater, fertilizer, and pesticides in agricultural fields. Although the TCR of Pb was below the standard guideline value (10^{-4}) but the non-carcinogenic health risks assessment parameter (HI) was above the standard limit (1.0), which revealed that the combined impact of all heavy metals was a matter of health concern for the local inhabitants.

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