Open Access Journal of Biomedical Engineering and Biosciences
DOI: 10.32474/OAJBEB.2019.03.000159
ISSN: 2637-4579
Research Article

Toxic Elements Accumulation in Vegetables from Soil Collected from the Vicinity of a Fertilizer Factory and Possible Health Risk Assessment

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Received: March 06, 2019 **Published:** March 13, 2019

Abstract

Present study deals with the investigation of the magnitude of toxic elements in commonly consumed vegetables grown in the vicinity of a Fertilizer Factory and evaluate the degree of health risk burden due to dietary intake of those vegetables as well. The vegetable samples showed greatest probabilities of toxic elements (Cr, Mn, Fe, Ni, Cu, Zn, As and Pb) contamination depending on their species and locations. To find out the possible source of contaminants, soil samples from where the vegetable samples were collected were also analysed. In most cases concentration of all the elements (Cr, Mn, Fe, Ni, Cu, Zn, As and Pb) in soil samples are equal to or near the suggestive world average value. To find out the solubilisation of these contaminants, soil-plant Transfer Factor (TF) was also calculated and the value obtained are below 1 in each case with an exception of Cu for Brinjal (1.065)and Sponge Gourd (1.027).

Health risk assessment was also done on the basis of various health risk indices calculation. It was found that all the vegetables are highly contaminated with the toxic elements analysed. Estimated daily intake of metal revealed that all the elements are within the reference dose (suggested by WHO. USEPA) except arsenic (As) and lead (Pb) and therefore HRI value for As and Pb was also found significantly high to pose any health hazard. Calculated non-carcinogenic (THQ) value for Cr and Ni was below 1 but for As and Pb the THQ value was high enough for public health concern. Cr, Ni, As and Pb present in different vegetable samples posed significant levels of carcinogenic risk as their values exceed the safe limit (10^{-6} - 10^{-4}) suggested by USEPA.

Keywords: Toxicity; Hazard Quotient; Xrf Spectroscopy; Health Risk Index; Transfer Factors; Carcinogenic Risk

Introduction

Unplanned industrialization and its wastages pouring system are responsible for toxic element and heavy metal contamination in water bodies as well as agricultural field and its products. The natural geochemical cycling of the ecosystem is therefore influenced with the pollution. The soil properties i.e. organic matter, clay contents and pH have major influences on the extent of the effects of metals on biological and biochemical properties. Moreover there is always a tendency to transfer metal from soil to plant and ultimately to human body. As, Cd, Hg, Pb and Se are not essential for plants growth, since they do not perform any known physiological function in plants. But Co, Cu, Fe, Mn, Ni and Zn are essential elements required for normal growth and metabolism of plants but can easily lead to poisoning if the concentration be greater than optimal values. Uptake of heavy elements by plants and subsequent accumulation along the food chain is a potential threat to human health. Heavy metals become toxic when they are not metabolized by the body and accumulate in the soft tissues. Chronic level ingestion of toxic metals has undesirable impacts on humans and the associated harmful impacts become perceptible only after several years of exposure. Foodstuffs are the major intake source of toxic elements into the human body. Among food system, vegetables are the most exposed food to environmental pollution due to aerial burden. Vegetables take up heavy metal/toxic elements, accumulate them in their edible and non-edible parts. These accumulated toxic elements may be high enough to cause clinical problems to human beings. One of the most important aspects of food quality assurance is heavy metal contamination. As awareness of the risk of elemental (toxic) contamination in food chain increases, national and international regulations on food quality should bestrictly monitored. It has been shown that accumulation of toxic metals in the kidney and liver through foodstuffs is detrimental. Entrance of toxic elements through vegetables consumption may cause disruption of several biochemical processes, leading to cardiovascular, nervous, kidneys and bone diseases and moreover pregnant women and children are more vulnerable to toxicity.

As a developing country, Bangladesh heavily relies on agriculture for its economy. To feed the hungry mouth of 180 million people, its lands are being overly used for crop production. In doing so, fertility of the land-soil suffers. The lack nourishment in soil and also in order to maintain a good balance of production, artificial fertilizers are repeatedly or sometimes excessively used and therefore several Fertilizer Factories are grown all over the country. The Jamuna Fertilizer Company Ltd.,Jamalpur of Bangladesh contributing significantly to the production of urea and meet the demand for fertilizer of the country from years. The main raw materials used are the natural gas along with different catalysts like Fe_2O3, Fe_3O4, Al_2O3, K_2O, CaO, and SiO_2 etc. The liquid wastes generated from the factory are discharged to the environment which finally go to the Jamuna River and to the adjacent agricultural lands, and eventually the vegetables and crops grown on those lands and thus contaminate the environment and cause toxicity to the public there after. Therefore, this study aimed to investigate concentrations of Cr, Mn, Fe, Ni, Cu, Zn, As and Pb in the most frequently consumed vegetables collected from the lands adjacent to 'The Jamuna Fertilizer' area to get an idea about the toxicity level of the vegetables grown in the area.Also a value of intake of heavy metals in human diets was calculated to estimate the risk to human health. Since Cr, Ni, As and Pb are highly carcinogenic, therefore the possible carcinogenic and non-carcinogenic risk due to consumption of those vegetables to human was calculated as well.

Materials and Methods

Study Area

The North-East side of 'The Jamuna Fertilizer Co. Ltd.'Jamalpur of Bangladesh was selected as study area and different vegetables as well as soil samples were collected for the experiments. The geographical coordinates of 'The Jamuna Fertilizer Co. Lld.', Jamalpur of Bangladesh are at 24° 40' 0" North, 89° 50' 0" East, which is beside of the Jamuna River. The sampling site is a residential cum agricultural field area and the samples were collected randomly from ten different points around the Fertilizer factory. There are several car workshops near the Fertilizer factory site from where solid and different types of wastage are frequently dumped into the nearby land. The site map of sampling area is shown in Figure 1.

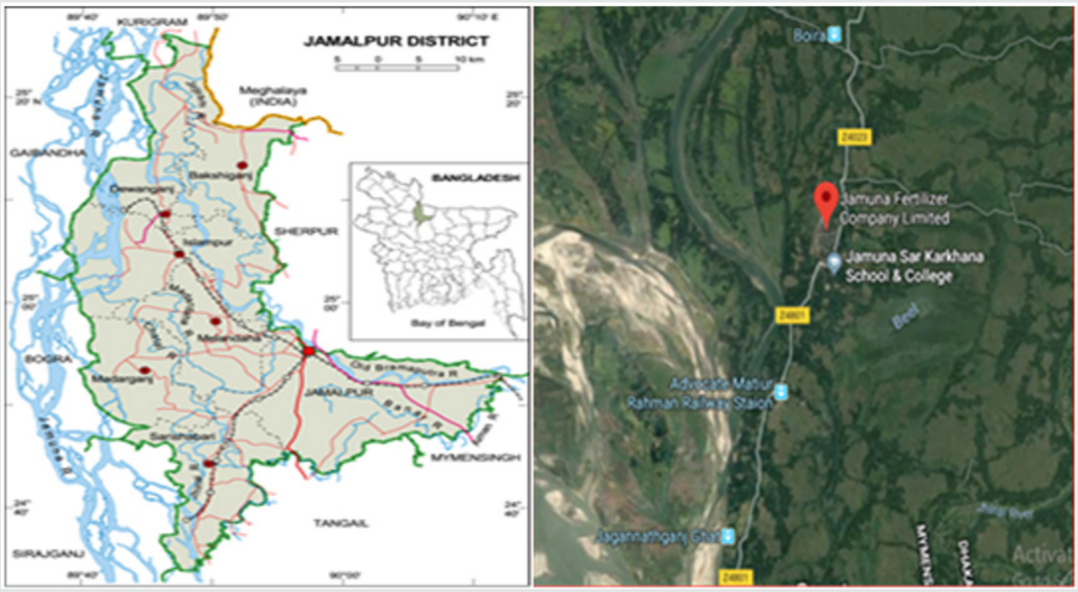

Figure 1: Site map of 'The Jamuna Fertilizer Company Limited', Bangladesh.

Sample Preparation

The vegetable samples collected for this research works are shown in the Table 1 with their scientific names and abbreviated identities. In rder to show the elemental correlation, soil samples were also collected with the vegetable samples from the different places of sampling site and labeled as SS1 - SS10.

Table 1: Description of vegetable samples analysed.

Local name	English name	Designation (sample ID)	Scientific name
Dhundul (L)	Sponge Gourd	SG1	*Luffacylindrica*
Begun	Brinjal	BR	*Solanummelongena*
Kachkola	Plantain	PL	*Musa paradisiaca*
MuchaAlu	Potato	PO	*Solanumtuberosum*
Morich	Chilli	CH	*Capsicum species*
Kakrol	Teasle Gourd	TG	*Momordicacochinchinensis*
Kachu	Giant Taro	GT	*Alocasiamacrorrhiza*
Dhundul (R)	Sponge Gourd	SG2	*Luffacylindrica*
Papaya	Green Papaya	GP	*Carica papaya*
Jhingga	Ribbed Gourd	RG	*Luffaacutagula*

Preparation of Soil Samples

The soil samples after collection were sieved with a stainless steel sieve to remove dirt and vegetable materials. All samples were then taken into porcelain dishes separately. Each dish with the particular sample was placed in an oven at around 70 ºC until a constant weight was obtained. The dried mass of each sample was then pulverized to fine powder using a mortar and pestle, and preserved in a plastic vial with the identification mark inside a desiccator. Finally, the homogeneous powder was used to prepare pellet (7 mm dia. and 1mm thick using 10 ton pressure by a pellet maker (Specac, UK) for elemental analysis by XRF.

Preparation of Vegetable Samples

The plant samples were cut into suitable pieces with a stainless steel knife, washed first with tap water, and then rinsed with deionized water three times. All plant samples were then taken into porcelain dishes separately. Each dish with the particular sample was marked by an identification number and placed in an oven at around 70°C for overnight drying which was continued until a constant weight was obtained. The dried mass of each sample was then transferred to a mortar and ground to fine powder using a pestle and preserved in a plastic vial with identification mark inside a desiccator.

Sample Irradiation and Method Validation

The experiments and sample irradiation have been done using EDXRF Spectroscopy System. The X-Ray beam of 22.4 keV from 109Cd point source hits the target sample and the characteristic X-rays are produced. The [Si (Li)] detector (Canberra) having the resolution of 175 eV at 5.9 keV has been applied for the detection of characteristic X-rays. These detected X-Rays are converted into voltage pulses and amplified by the spectroscopy amplifier and processed in MCA having16K+channel.The irradiation and spectrum data acquisition are operated and controlled by a software package provided with the system. The standard materials were also irradiated under similar experimental conditions for construction of the calibration curves for quantitative elemental determination in the respective samples. The commercial software AXIL has been applied for the qualitative and quantitative elemental analysis.

A direct comparison method based on EDXRF technique was used for elemental concentration measurement Islam, et al. [1] and Jolly, et al. [1] and Jolly, et al.[2] in different samples. As the analysis is based on direct comparison, the standards of similar matrices were used to construct the calibration curve in order to avoid any matrix effect. Three soil standards (Soil-7 /IAEA, Montana-1/2710a, Montana-2/2711a) and five plant standards (Apple Leaf/NIST 1516, Spinach/NIST 1570a, Orchard Leaf/NIST 1571, Tomato Leaf/NIST 1573a, and Peach Leaf/NIST 1574) were used for the construction of calibration curves for carrying out elemental analysis in soil and vegetables respectively. The curves were constructed by plotting the sensitivities of the elements as a function of their atomic number. The validation of the calibration curve constructed for elements present was checked through analysis of standard reference materials "Montana-1" for soil and "Orchard Leaf/NIST 1571" for vegetable. The results obtained for elements of interest and certified values for corresponding elements are shown in the Table 2. All results in respect to certified known values were found to vary within the acceptable range of error.

Table 2: Comparison between present results and the certified values of standard reference materials (mgkg^{-1}).

Element	Soil (Montana-1)			Plant (Orchard Leaves)		
	Results obtained	Certified values	% Error	Results obtained	Certified values	% Error
K	21113	21700	2.71	16655.29	14700	13.30
Ca	9136	9640	5.23	18590	20900	11.05
Mn	2128	2140	0.56	94.26	91	3.58
Fe	39685	43200	8.14	281.291	300	6.24
Ni	8.67	8.0	-8.38	1.321	1.3	-1.62
Cu	3409	3420	0.32	7.545	12	37.13

Zn	4179	4180	0.02	8.299	25	66.80
As	1441	1540	6.43	6.443	10	35.57
Se	1.2	1.0	-20.00	0.174	0.08	117.50
Pb	5382	5520	2.50	46.15	45	2.56

Statistical Analysis

Pearson's correlation matrix is used to identify the relationship among the pairs of parameters in soil sample. The correlation coefficient matrix measures how well the variance of each constituent can be explained by relationship with each other Liu, et al. [3] and soil data were subjected to statistical analysis using IBM SPSS software (version 20). PCA has been performed to extract principal components (PC) of soil from the sampling points and to evaluate spatial variations and possible sources of heavy metals in soil sample Chabukdhara, et al. [4] and Nema, et al. [4]; Sarbu, et al. [5] and Pop, et al. [5].

Data Analysis

Metal Pollution Index

Metal Pollution Index (MPI) was computed to determine overall metal concentration in each variety of vegetable sample analyzed. This index was obtained by calculating the geometrical mean of concentrations of all the elements consist in vegetable sample collected following Ureso, et al. [6].

Daily Intake of Metal (Dim)

The daily intake of heavy metals by people through consumption of vegetables was calculated following Chary, et al. [7]. The average body weight was considered to be 70 kg that each person consumes approximately 300 gm WHO, et al. [8] of vegetables per day.

Health Risk Index

Assessment of health risk due to contaminated vegetable intake was done using a Hazard Quotient (HQ) (U.S. Environmental Protection Agency US EPA 1989). HQ is the ratio between exposure and the reference oral dose (RfD). An estimate of the potential hazard of heavy metal to human health (HQ) through vegetables intake is calculated by

HQ=DIM/RfD

Where (Div) is the daily intake of vegetable (Kg/day),(Celement) is the concentration of element in the vegetable (mg kg^{-1}), RfD denotes the oral reference dose for the element (mg $kg^{-1}$of body weight/day).Although the HQ-based risk assessment method does not provide a quantitative estimate for the probability of an exposed population experiencing a reverse health effect, it indeed provides an indication of health risk level due to exposure to pollutants Chary, et al. [7].The HQ is a highly conservative and relative index. When HQ is < 1, there is no obvious risk from the substance over a lifetime of exposure, while HQ is > 1, the toxicant may produce an adverse effect. The probability of experiencing long-term carcinogenic effects increases with the HQ value. This risk assessment method has been used by researchers Jolly, et al. 2017 and proved to be valid and true.

Transfer Factor

The uptake of elements from soil to plants is measured by transfer factor (TF). The TF for any element can vary considerably depending on the kind of plant, as well as from one environment to another. The main parameters that modify the TF are the physical and chemical characteristics of soil, behaviour of trace metal present in soil and plant, change of environment Al-Hamarneh, et. al. [9]; A. Martinez et al. [10]. The metal transfer factor from soil to plants have been calculated on dry weight basis and the formula Jolly et al. [11] is

Transfer Factor (TF) =CPlant/ CSoil

Where,Cplant and Csoil represent the elemental concentration of plant and soil respectively.

Hazard Index (Hi)

To evaluate the potential risk to human health through more than one heavy metal, the HI has been developed (USEPA, 1989). The HI is the sum of the Hazard Quotients as described by the equation;

HI = ΣHQ = HQCr+ HQMn+ HQFe+HQCu+ HQZn++HQNi+ HQAs+ HQPb

It is assumed that the magnitude of adverse effect will be proportional to the sum of multiple metal exposures. It also assumes similar working mechanisms that linearly affect the target organ.

Non-Carcinogenic Risk

In this study, the non-carcinogenic health risk associated with the consumption of collected vegetables were assessed based on the target hazard quotients (THQs) and the calculations were made using the standard assumption for an integrated following USEPA 1989,

$$THQ= [(EFr\times ED\times FIR\times C)/(RfD\times BW\times AT)]\times 10^{-3}$$

Where THQ is the target hazard quotient (dimensionless), EFr is the exposure frequency (365 days year-1), ED is the exposure duration (70 years for adult) equivalent to the average human lifetime , FIR is the body ingestion rate for vegetables (166g preson-1day^{-1}),C is the element concentration in samples (mg kg^{-1} fresh $weight^{-1}$), BW is the average body weight (60 kg for adults), AT is the average time for non-carcinogens (365 day $year^{-1}$×number of exposure years), and RfD is the oral dose (mg kg^{-1} day^{-1});. The RfDs represent an estimate of the daily exposure over a lifetime without an appreciable risk of deleterious effects. If the THQ is less than 1, the exposed population is unlikely to experience obvious adverse effects. If the THQ is equal to or higher than1, there is a potential health risk Wang, et al. [12] and to which related interventions and protective measurements should be taken.

Carcinogens Risk

In this study, carcinogenic risks were estimated as the incremental probability of an individual to develop cancer over a lifetime exposure to that potential carcinogen (i.e. incremental or excess individual lifetime cancer risk) USEPA et al. [13]. Acceptable risk levels for carcinogenic range from 10^{-4} (risk of developing cancer over a human lifetime is 1 in 1,000,000). The equation used for estimating the target cancer risk (lifetime cancer risk) is as follows USEPA et al. [13]:

$$TCR= EFr\times ED\times FIR\times C\times CSFo/BW\times AT)\times 10^{-3}$$

Where TCR (dimensionless) represents the targets carcinogenic slope factor from the Integrated Risk Information System (USEPA 2010) database which was 1.5 mg kg^{-1} day-1 for arsenic(As) and 0.0085 mg kg^{-1} day-1 for the lead (Pb),0.5 mg kg^{-1} day-1 for the chromium(Cr) and 1.7mg kg^{-1} day^{-1} for the nickel (Ni).

Result And Discussion

Analysis Of Soil Samples

Level Of Toxic Metal In Soil Samples

Table 3: Concentration of heavy metals in different types of soil samples.

Element	Concentration (mg/kg)										
	(SS1)	(SS2)	(SS3)	(SS4)	(SS5)	(SS6)	(SS7)	(SS8)	(SS9)	(SS10)	WAV (PPM) **
Cr	<5.18	<5.18	<5.18	<5.18	<5.18	<5.18	<5.18	<5.18	<5.18	<5.18	47
Mn	261.52 ±4.76	431.86 ±5.54	237.53 ±4.65	261.52 ±5.29	187.14 ±2.72	355.09 ±3.67	242.32 ±3.57	191.94 ±2.29	297.51 ±3.04	398.27 ±4.37	270
Fe	55610 ±3214	36870 ±2351	54295 ±3361	52820 ±4319	41380 ±3218	62465 ±4327	29335 ±2741	42530 ±3265	47665 ±3809	56860 ±4213	40000
Ni	<0.19	17.57 ±0.72	31.45 ±1.32	<0.19	<0.19	<0.19	<0.19	<0.19	18.8 ±0.76	20.55 ±1.06	13
Cu	15.74 ±0.89	12.41 ±1.41	13.72 ±1.32	20.56 ±2.11	16.84 ±1.43	22.54 ±2.03	12.14 ±1.01	10.22 ±0.78	17.99 ±1.25	14.97 ±1.52	13
Zn	119.97 ±4.65	61.55 ±2.43	129.68 ±3.12	199.11 ±4.45	135.31 ±3.51	197.36 ±4.61	87.01 ±3.43	106.65 ±4.01	240.09 ±5.12	152.78 ±4.38	45
As	8.69 ±1.23	7.5 ±0.73	9.34 ±0.74	8.1 ±0.65	5.63 ±0.42	7.47 ±0.61	8.08 ±0.78	7.42 ±0.43	8.28 ±1.05	9.22 ±1.01	4.4
Pb	89.3 ±3.65	73.76 ±2.62	91.72 ±3.76	86.48 ±4.23	71.9 ±3.09	127.25 ±4.87	66.77 ±3.26	81.08 ±3.45	99.33 ±4.12	80.62 ±3.71	22

** World Average Value (Kabata, A, Pendias, H.1984)

Concentration of the elements (Cr, Mn, Fe, Ni, Cu, Zn, As and Pb) in soil samples are presented in Table 3 .Highest concentration of Mn was found in the sampling siteSS2 (431.86 mg kg^{-1}) and lowest concentration was found in the sampling site SS5 (187.14 mg kg^{-1}), whereas concentration of Mn according to world average value is 270 mgkg-1.Range of other elements viz: Fe, Ni, Cu, Zn, As and Pb was found 29335-56860, 17.57-31.45, 10.23-22.54, 61.55-240.09, 5.63-9.34 and $66.77^{-1}27.14$ mg kg^{-1} respectively. Concentration of Cr was too low to detect by the system. Average concentration of most of the elements are more or less identical to the World average value Pendias, et al. [14] with an exception of Zn, As and Pb. Higher value of Zn and As can be attributed due to use of crop fertilizer or additives, on the other hand higher value of As and Pb can be attributed due to the contamination of solid waste discharged from the industry.

Result from Pearson Correlation Matrix Analysis

Table 4: Pearson correlation for soil samples.

Elements	Mn	Fe	Ni	Cu	Zn	As	Pb
Mn	1						
Fe	0.225	1					
Ni	0.391	0.162	1				
Cu	0.162	0.636*	-0.223	1			
Zn	0.013	0.596	0.030	0.801**	1		
As	0.287	0.404	0.571	-0.112	0.110	1	
Pb	0.226	0.764*	0.017	0.709*	0.666*	0.159	1

*Correlation is significant at the 0.05 level (2-tailed)

**Correlation is significant at the 0.01 level (2-tailed)

Pearson's correlation matrix is generated in order to identify the correlation among the heavy metals found in the soil samples collected from adjacent area of The Jamuna Fertilizer Factory Ltd. The Pearson correlation matrix of different elements found in the soil sample is given in the Table 4.

The p value of correlation matrix indicates the strength of associations in between different elements. Such as, p value containing 0.01 and 0.05 indicates strong and significant correlations respectively. Pearson correlation matrix reveals that the correlation between Cu and Fe (r = 0.636) is significant and Zn possesses positive correlation with Fe (r = 0.596) and strong correlation with Cu (r = 0.801). Pb and Fe (r = 0.764) also exhibit significant correlation. Pb also possess significant correlation with Cu (r = 0.709) and Zn (r = 0.666). The strong positive correlation could indicate similar source origin of heavy metal in different soil sample.

Result from Principal Component Analysis

In order to obtain reliable estimates of the different sources of heavy metals contributing to the soil analysed, Principal Component Analysis (PCA) was used to identify major elements associated with sources. Varimax rotation is used to maximize the sum of variances of the factor coefficients which better explains the possible groups/factor that influence fine air particulate matter. The total variance in each factor was calculated as the sum of the squared loadings for the given factor. A plot of this eigenvectors as a function of factor number is given in the Figure 2. There are two factors which represents 72% of the total variance. The scree plot is used to identify the number of PCs to be retained to understand the underlying element structure. The calculated factor loadings together with cumulative percentage and percentage of variance are explained by the each factor as listed in Table 5.

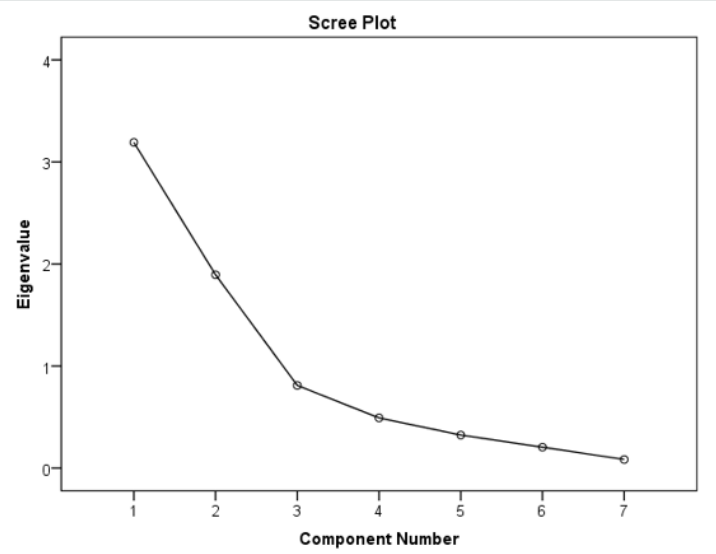

Figure 2: Scree plot of the characteristic roots of principal component analysis.

Table 5: Principal Component Analysis (PCA) with varimax rotation for all heavy metals found in studied.

Heavy metal	Component	
	PC1	PC2
Cu	.924	-.167
Pb	.884	.144
Zn	.869	.005
Fe	.817	.352
Ni	-.103	.859
As	.079	.835
Mn	.152	.616
Eigen values	3.191	1.894

In this study, two principal components PC1 and PC2 explain more than 45% and 27% of variance respectively. The first principal component PC1 loaded with Cu (0.924), Pb (0.884), Zn (0.869), Fe (0.817) . Iron is the fourth most abundant element and second most abundant metal in the Earth's crust. It is a major element in soil with a median value of 2.1% A. W. Rose et al. [15]. It is present mostly as Fe^{2+} in ferro-magnesian silicates, such as olivine, pyroxene, amphibole and biotite, and as Fe^{3+} in iron oxides and hydroxides, as the result of weathering. The anthropogenic sources of iron have been reported to include the iron and steel industry, sewage (C. Reimann et al., 1998). Iron sulphate is also used as a fertilizer and herbicide C. Reimann, et al. [16]. Our sampling site, Jamuna Urea fertilizer factory uses natural gas as starting material along with catalyst Fe_2O_3, Fe_3O_4 M.A Samad, et al. [17]. Thus the wastes generated from fertilizer factory may contaminate our environment. Zinc occurs naturally in soil (about 70mg kg^{-1} in crustal rocks) B. E. Davies, et al. [18], but Zn concentrations are rising unnaturally, due to anthropogenic additions.

Most Zn is added during industrial activities, such as mining, coal, and waste combustion and steel processing.. Vehicle brakes and tire wear as possible sources of Zn. The natural Pb content in soil is, of course, related to the composition of the parent rock. Although the species of Pb vary considerably with soil type, it is mainly associated with clay minerals, Mn oxides, Fe and Al hydroxides and organic matter. A baseline Pb value for surface soil on the global scale has been estimated to be 25 mg kg^{-1}; levels above this suggest an anthropogenic influence A. Kabata-Pendias, et al. [14]. Along with fugitive Pb, Pb-bearing glass and pottery glazes, batteries, old lead-based paints, the corrosion of lead pipes in areas of soft water and sewage sludge are all potential sources of Pb. Copper (Cu) in the Earth's crust is the most abundant in intermediate rocks and has a tendency to be excluded from carbonate rocks.

It forms several minerals of which the common primary minerals are simple and complex sulfides. These minerals are quite easily soluble in weathering processes and release Cu ions, especially in acid environments. Copper is therefore, considered among the more mobile of the heavy metals in hypergenic processes. However, Cu is a very versatile trace cation and in soils or depositional material exhibits a great ability to chemically interact with mineral and organic components of soil A. Kabata-Pendias, et al. [14]. Cu is also released from burning of fuel, wearing out of tires, leakage of oils, and corrosion of batteries and metallic parts. So we can assume that the first principal component PC1 containing Cu, Pb, Zn, Fe reveals the both natural and anthropogenic sources for measured heavy metal in different soil sample.

The second principal component PC2 is loaded with Ni (0.859), As (0.835), Mn (0.616). Natural and anthropogenic sources (e.g. mining and smelting, coal fly ash, bottom ash, metal manufacturing waste, commercial waste, atmospheric fall-out and deposition, urban refuse, and sewage sludge) contribute to the levels of nickel found in soil. Nickel catalyst also used in different steps of Urea production in Urea fertilizer factory. Manganese constitutes approximately 0.1% of the earth's crust, and is a naturally occurring component of nearly all soils. Accumulation of manganese occurs in the subsoil rather than on soil surface. An estimated 60-90% of soil manganese is associated with the sand fraction ATSDR, et al. [19]. Mn is likely to occur in soils as oxides and hydroxides in the form of coatings on other soil particles. The Earth's crust is an abundant natural source of arsenic. Arsenic is mobilized in sediments with Fe, Mn and organic matter from upper stream to lower stream of the river Jamuna. Arsenic may remain sorbed or co- precipitated with Mn species. Mn-oxyhydroxide minerals that strongly sorb arsenic Mohammad Arifur Rahman, et al. [20] fortifies the fact. So, we can assume that there is a probable relation between Mn and As in our studied samples.

Level of Toxic Metal in Vegetable Samples

The experimental results obtained from vegetable samples analyses have been projected in Table 6. It was found that elements and their concentration are varied from sample to sample and also from their locations. The relative abundance of different elements obtained in the samples collected from the Industrial area is shown in Table 7. Mn, Fe, Cu, Zn and Pb are abundant in all types of vegetables comparing to other elements analysed. Cr is found in Sponge Gourd (L), Plantain and Ribbed Gourd which are within the world average value and in case of other vegetables the concentration was too low to detect by the system. Highest concentration of Mn was found in Giant Taro (100.00 mg kg^{-1}) and lowest

Table 6: Concentration (mg/kg) of heavy metals in different types of vegetable samples.

Element	Concentration, mg/kg										
	Sponge Gourd L	Brinjal	Plantain	Potato	Chilli	Teasle Gourd	Giant Taro	Sponge gourd R	Green Papaya-	Ribbed Gourd	W.A.V
Cr	0.82± 0.12	<0.05	0.52± 0.09	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.71± 0.13	0.10-0.50
Mn	56.10± 4.83	58.60± 6.62	59.91± 10.62	58.37± 7.12	64.06± 8.07	55.26± 3.49	100± 10.45	53.21± 2.36	58.48± 11.67	67.32± 3.97	30-300
Fe	74.84± 15.26	69.5± 5.26	68.37± 7.44	52.59± 0.78	118.81± 6.26	60.52± 0.89	139.43± 0.61	87.01± 6.16	151.26± 1.21	93.50± 9.09	
Ni	0.92± 0.17	<0.65	0.81± 0.11	<0.65	<0.65	<0.65	0.89± 0.14	<0.65	0.83± 0.12	0.67± 0.19	0.10-5.0
Cu	10.09± 1.02	13.2± 0.96	10.38± 0.1	6.85± 0.11	6.65± 2.22	8.63± 0.26	6.72± 0.67	10.49± 0.14	6.86± 0.86	11.75± 2.68	30-May
Zn	7.85± 0.1	7.90± 0.05	8.67± 0.49	9.47± 0.22	9.01± 0.49	6.97± 0.46	9.84± 0.1	9.71± 0.33	9.05± 0.64	12.20± 0.3	27-150
As	<0.01	<0.01	<0.01	6.31± 0.21	4.39± 2.65	<0.01	5.13± 0.48	4.34± 0.21	1.64± 2.65	3.39± 0.32	1-1.7
Pb	40.81± 7.22	32.80± 1.6	35.37± 3.76	30.39± 2.95	36.36± 5.63	13.89± 0.32	23.15± 4.35	10.05± 0.81	5.77± 2.36	13.51± 1.83	10-May

Table 7: Relative abundance of different elements in the vegetables.

Cr	SG(L)>RG>PL >Remaining in BDL (<0.05mg/kg^{-1})
Fe	GP>GT>CH>RG>SG2>SG1>BR>PL>TG>PO
Mn	GT>RG>CH>PL>BR>GP>PO>SG1>TG>SG2
Ni	SG1>GT>GP>PL>RG> Remaining in BDL (<0.65mg/kg^{-1})
Cu	BR>RG>SG2)>PL>SG1>TG>GP>PO>GT>CH
Zn	RG>GT>SG2>PO>GP>CH>PL>BR>SG1>TG
As	PO>GT>CH>SG2)>RG>GP> remaining BDL(0.01 mg kg^{-1})
Pb	SG1>CH>PL>BR>PO>GT>TG>RG>SG2>GP

Was found in Sponge Gourd-R (53.21 mg kg^{-1}) and all the values are within the world average value. Highest concentration of Fe was found in Green Papaya (151.26 mg kg $^{-1}$) and lowest in potato (52.59 mg kg^{-1}) but Fe is considered as an essential element for human health. Concentration of Ni was also found within the world average value and in most cases it was too low to detect by the system. All varieties of vegetables were found to contain Cu and Zn in a reasonable amount and were within the world average value. Concentration of As was found within the world average value for Sponge Gourd (L), Brinjal, Plantain, Teasle Gourd and Green Papaya but it was found in higher concentration in potato (6.31 mg/kg), Chilli (4.39 mg/kg), Giant Taro (5.13 mg/kg), Sponge Gourd-R (4.34 mg/kg) and Ribbed Gourd (3.39 mg/kg). Concentration of Pb was found much higher than the world average value in all types of vegetables analysed which may be due to the contamination of industrial discharge associated with the disposal of damaged battery from nearby workshop. In a study Jolly, et al. [11] reported to have found As, Cu, Mn, Pb, Ni, and Zn below the world average value in the vegetables collected from Rooppur Area, Bangladesh Lokeshappa, et al. [2] studied the elemental concentration in different agricultural products and the results obtained are comparable with the present study.

Transfer Factor

The Transfer Factor (TF) for Cr, Mn, Fe, Ni, Cu, Zn, As, and Pb for different vegetables varied greatly between plant species and locations Table 8. Transfer factor for As, Pb and Cu are relatively high compared to other elements analysed. pH, electrochemical properties of soil, electrolyte concentration are the major factors that influenced the migration transformation ability of toxic elements indirectly. The TF of Cr in vegetables ranges from 0.137-0.158, which is identical with the value reported by Avci, H and (Deveci T, et al. (2013)). Cr does not easily translocate in the plant and mostly concentrated in the root and hence the lower TF value for Cr was observed. For elements Mn, Fe, Znand Ni, the TF value is <<1, which may be due to pH value of the soil that influenced the sorption of elements in the soil and its bioavailability. A similar observation was reported by Jolly, et al. [11] where the TF value for the vegetables Spinach, Amaranth, Brinjal, Tomato, Radish, Bean, Cauliflower and Carrot collected from Rooppur area of Bangladesh was too low.

Table 8: Transfer factor from vegetables to soil samples.

Element	Sponge gourd	Brinjal	Plantain	Potato	Chilli	Teasle Gourd	Giant Taro	Sponge Gourd	Green Papaya	Ribbed Gourd
Cr	0.158	-	-	-	-	-	-	-	-	0.137
Mn	0.215	0.136	0.252	0.223	0.342	0.156	0.413	0.277	0.197	0.169
Fe	0.001	0.002	0.001	0.001	0.003	0.001	0.005	0.002	0.003	0.002
Ni	-	-	0.026	-	-	-	-	-	0.044	0.033
Cu	0.641	1.065	0.757	0.333	0.395	0.383	0.554	1.027	0.381	0.785
Zn	0.065	0.129	0.067	0.048	0.067	0.035	0.113	0.091	0.038	0.080
As	-	-	-	0.779	0.779	-	0.710	0.850	0.530	0.710
Pb	0.457	0.445	0.386	0.351	0.506	0.109	0.347	0.124	0.058	0.168

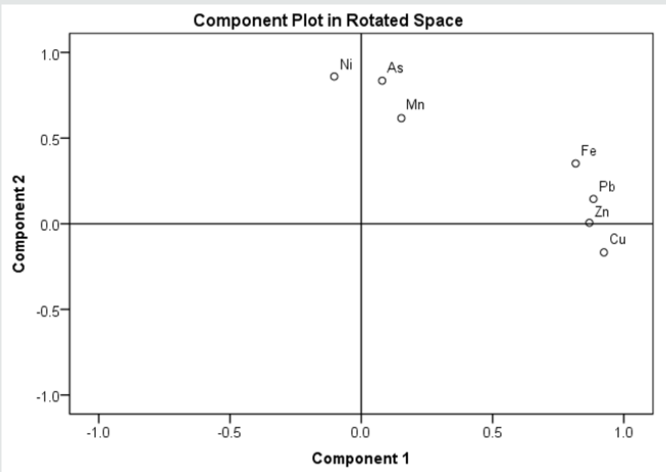

Figure 3: Component plot in rotated space of principal component analysis.

Citation: S Akter, S.M. A Islam, M. O Rahman, K.M. Mamun, Y.N. Jolly, et al., Toxic Elements Accumulation in Vegetables from Soil Collected from the Vicinity of a Fertilizer Factory and Possible Health Risk Assessment. Op Acc J Bio Eng & Bio Sci 3(2)- 2019. OAJBEB.MS.ID.000159. DOI: 10.32474/OAJBEB.2019.03.000159.

Toxic element Arsenic is found in almost all of the vegetable samples and the TF of as (0.530 - 0.877) is quite high comparing to Pb (0.058 - 0.506). This study showed that the transfer factor of As and Pb are quite higher in almost all the vegetable samples analysed. The present result agrees with the investigation made by Zhuang, et al. [21] in the food crops in the vicinity of Dabaoshan mine, South China where the transfer factors for heavy metals were significantly higher in vegetables (Figure 3).

Metal Pollution Index

Metal Pollution Index (MPI) is suggested to be a reliable and precise method for elemental pollution monitoring of wastewater irrigated areas Usero, et al. [6]. Metal (MPI) pollution Index of different vegetables are projected graphically in Figure 4 and followed the sequence of RG>GT>CH>PO>SG2>GP>SG1>PL>BR>TG. In a study Jolly et al 2013b reported that the calculated MPI values in different vegetable samples collected from Rooppur , Pabna area of Bangladesh and found the sequence as SG>BR>PL>GP. In the present study, Ribbed Gourd (RG), Giant Taro (GT), Chilli (CH) Potato (PO) found to show higher MPI value and hence are the most popular and frequently used vegetables in Bangladesh. However higher MPI are calculated for different vegetables grown around 'The Jamuna Fertilizer co. Ltd.' areas and suggested that these may cause more health risks for the workers and the residents around.

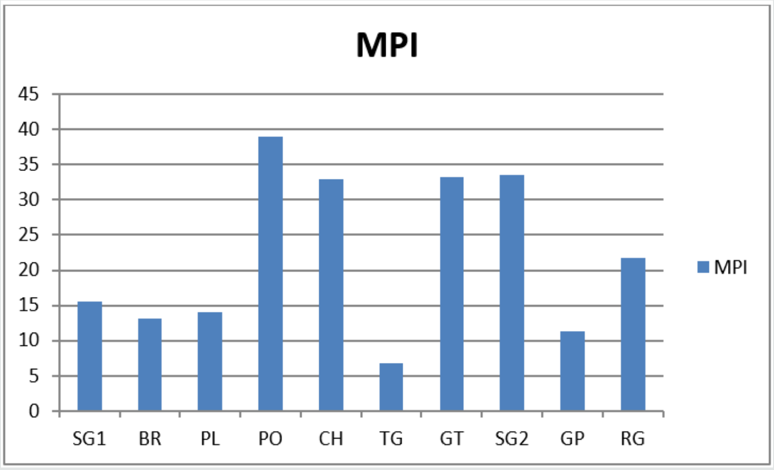

Figure 4: Metal Pollution Index in the Vegetable samples.

Daily Intake Of Metal (Dim) Through Vegetables

The degree of toxicity of heavy metal to human depends upon their daily intake of heavy /toxic metal through different food stuffs. Daily intake of metals from mixed vegetables by human being are projected in the table considering the intake values by taking the average values of metals in all ten varieties of vegetables analyzed in this research work. It may be a realistic estimation for the average intake of metals from vegetables as different vegetables are consumed by different segment of population variably at different time throughout the year. In the present study, DIM was calculated by considering average body weight 70 kg according to WHO and daily consumption of vegetables was 300 g WHO et al. [8]. However the table revealed that intake of heavy metals except Mn, As and Pb are within the permissible values referred by different agencies Table 9. Mn is considered as an essential element for human health thus high value of HRI can be ignored but DIM value for As and Pb are too high compared to suggestive value and hence are really alarming.

Table 9: Estimated Daily Intake of Metal (DIM) through vegetables.

Trace elements	Average conc. Of 10 vegetables (µg/g)	Intake by human being (mg/kg)	RfDa (mg/day)	References
Fe	91.57	27.50	10.0-60.0	WHO 1994
Cu	9.16	2.75	2.0-3.0	WHO 1994
Mn	63.14	18.96	0.5-5.0	WHO 1994
Zn	9.07	2.72	15.00	WHO 1994
As	2.52	0.76	0.0003	USEPA 2002
Cr	0.24	0.07	105	US EPA 2010
Ni	0.74	0.22	1.400	US EPA 2010
Pb	24.21	7.27	0.188	WHO 1994

Health Risk Index (Hri)/Hazard Quotient (Hq)

The calculated value of Health Risk Index (HRI) associated with the elements Cr, Mn, Fe, Ni, Cu, Zn, As and Pb through consumption of the vegetables collected near Jamuna Fertilizer Factory area are presented in the Table 10. The result revealed that HRI for Cr, Fe, Ni, Cu and Zn are below 1 indicating safe for the consumer. In a study Jolly, et al.[22], 2013b reported that HRI value for Cr, Mn, Fe, Co, Ni and Cu were within the value 1 for the vegetables collected from Rooppur, Pabna area of Bangladesh. In another study Singh, et al. [23] it was observed that Cu, Zn and Cr were not found to cause any risk to the people by consuming vegetables and cereals grown in an area around Dinapur Sewage treatment plant, India. khan et al. 2014 reported, HRI associated with the heavy metals Cu, Co, Fe, Zn, Mn in the vegetables collected from the embankment of Buriganga river area were also found below 1 (one). However Arsenic (As) is found to show HRI value greater than 1 for some vegetables like potato, chili, Giant Taro, Sponge Gourd 2, Green Papaya and Ribbed Gourd and hence have greatest potential to pose health risk to human. Mn also found to show HRI value greater than 1 but as it is an essential element, the effect could be negligible. HRI value for Pb are greater

Table 10: Estimated Daily Intake of Metal (DIM) through vegetables.

Elements	Sponge gourd	Brinjal	Plantain	Potato	Chilli	Teasle Gourd	Giant Taro	Sponge gourd	Green Papaya	Ribbed Gourd
Cr	0.0007	-	0.0004	-	-	-	-	-	-	0.001
Mn	2.162	2.256	2.308	2.249	2.468	2.129	3.858	2.050	2.253	2.593
Fe	0.002	0.002	0.002	0.002	0.004	0.002	0.004	0.003	0.005	0.003
Ni	0.058	-	0.052	-	-	-	0.056	-	0.053	0.043
Cu	0.321	0.421	0.331	0.218	0.211	0.274	0.214	0.333	0.218	0.373
Zn	0.033	0.034	0.037	0.040	0.038	0.029	0.042	0.041	0.038	0.052
As	-	-	-	26.753	18.599	-	21.741	18.393	6.950	14.367
Pb	12.971	10.422	11.241	9.660	11.557	4.413	7.357	3.196	1.833	4.294

Than 1 for all the vegetables analysed, so there are huge concern for potential health effect. In a study Cui, et al. [24] reported to have been exposed by Cd and Pb through consumption of vegetables collected from an area near a smelter in Nanning, China. Jolly, et al. [11] b also reported to have found HRI value greater than 1 for Pb in spinach, amaranth, bottle gourd collected from Rooppur, Pabna area of Bangladesh. However high HRI value for As and Pb are representative indicate a health risk due to their high concentration in the respective vegetables and their toxicity. Furthermore As and Pb are associated with cancer risk, therefore it is important to monitor regularly for the prolonged ingestion of those elements that pose a health risk.

Hazard Index(Hi)

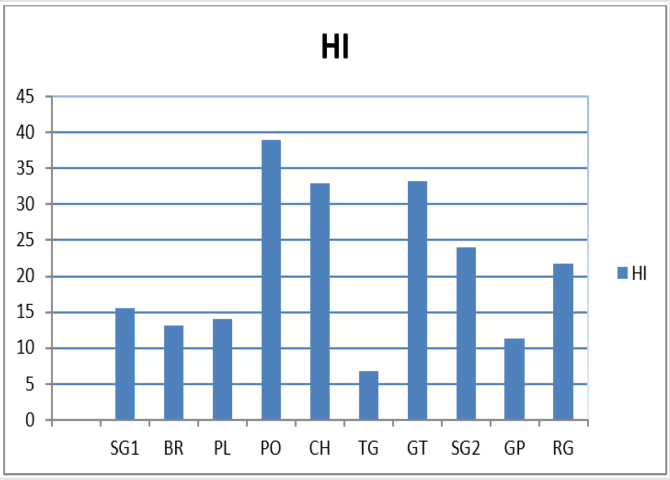

Figure 5: Hazard Index.

The hazard index (HI) value expresses the cumulative non-carcinogenic effects of multiple elements exposed to consumption of one or more foodstuffs.The calculated HI value followed the decreasing order of Potato>Sponge Gourd1>Giant Taro >Chilli>Ribbed Gourd>Sponge Gourd2> Plantain>Brinjal> Green Papaya>Teasle Gourd. When the hazard index exceed 1.0, there is concern for health hazard and hence HI values of more than 1.0 Figure 5. for all the vegetables are suggested not to consume.

Non-Carcinogenic Risk

The non-carcinogenic risks from consumption of vegetables by the adults were assessed based on the Target Hazard Quotients (THQs). The THQ is the ratio of the determined dose of a pollutant to a reference dose level. If the ratio is greater than 1, the exposed population is likely to experience obvious adverse effects Wang, et al. [12]. The methodology for estimation of THQs does not provide a quantitative estimate on the probability of an exposed population experiencing adverse health effect, but it offers an indication of the risk level due to contaminant exposure. The estimated THQs of the toxic element Cr, Ni, As and Pb are shown in Table 11. In case of Cr and Ni, calculated THQ for all the vegetables studied were

Found less than 1 (Table 11). Sponge Gourd (SG1), Brinjal, Plantain, Teasle Gourd have shown THQ value 0 for Arsenic (As) as concentration of as in those vegetables was too low to detect by the system. But in case of Potato, Chilli, Giant Taro, Sponge Gourd (SG2), Green Papaya and Ribbed Gourd, the calculated THQ value were 4.0751, 2.8330, 3.3117, 2.8017, 1.0587 and 2.1884 respectively and hence are in the unacceptable range (unity). WHO suggested a non-carcinogenic value for Arsenic (As) in rice is 1.9. Potential health risks from exposure to Arsenic (As) through the consumption of those vegetables are therefore of great concern and can cause skin lesions, dark spots on hand and feet, swollen limbs and loss of feeling from hands and legs. In case of Lead (Pb), all the vegetables are found to have shown higher THQ value with the exception of Sponge Gourd (SG2).Green Papaya and Ribbed Gourd. Lead (Pb) directly affects the hematopoietic system, high lead levels are reasons for human carcinogens, causes weakness in finger, wrists or ankles and neurological problems. Shaheen, et al. [25], 2016 reported a calculated THQ value 0.19 and 0.19 for As and Pb respectively in non-piscine protein source in Bangladesh.

Table 11: Non-Carcinogenic Risk.

Element	Sponge Gourd	Brinjal	Plantain	Potato	Chilli	Teasle Gourd	Giant Taro	Sponge gourd	Green Papaya	Ribbed gourd	*FAO/WHO Suggestive non-carcinogenic value
Cr	0.0001	-	0.0001	-	-	0.0016	-	-	-	0.0001	0.017a
Ni	0.0089	-	0.0078	-	-	1.5167	0.0086	-	0.008	0.0065	-
As	-	-	-	4.0751	2.833	-	3.3117	2.8017	1.0587	2.1884	1.9b
Pb	1.9758	1.5876	1.7123	1.4715	1.7603	1.6201	1.1206	0.4868	0.2792	0.654	0.097c

aFishsample; bRice; c leafy vegetables; d fruity vegetables

*FAO/WHO (2011)

Total Carcinogenic Risk:

The target carcinogenic risks (TCRs) derived from the intake of Cr, Ni, As and Pb were calculated since these elements may promote both non-carcinogenic and carcinogenic effects depending on the exposure dose (Table 12). Inorganic As is classified as a known carcinogen (USEPA group A) and Pb as the probable carcinogen (USEPA group B 2).The TCR values from exposure of Cr, Ni, As and Pb were found in the range of 12.13×10^{-3} to16.57×10^{-3} , 53.15 $\times10^{-3}$ to 72.99 $\times10^{-3}$, 114.80 $\times10^{-3}$ to 441.87 $\times10^{-3}$ and 2.29 $\times10^{-3}$ to 16.19 $\times10^{-3}$ respectively. In general, the excess cancer risk lower than 10^{-6} is considered to be negligible, cancer risk above 10^{-4} is considered unacceptable, and cancer risk lying between 10^{-6} and 10^{-4} is generally considered an acceptable range USEPA et al. [13], USEPA et al. [26]. From the table it is clear that all the contaminants in the studied vegetable samples posed significant carcinogenic risk level. (M.I. Hossen Real et al. 2017) reported that Cr, Ni, As and Pb posed carcinogenic risk in some staple foodstuff collected from Kawran Bazar wholesale market, Dhaka, Bangladesh, which agrees the present study [27,28].

Table 12: Carcinogenic Risk.

Element	Sponge Gourd	Brinjal	Plantain	Potato	Chilli	Teasle Gourd	Giant Taro	Sponge Gourd	Green Papaya	Ribbed gourd
Cr	0.079×10^{-3}	-	0.050×10^{-3}	-	-	-	-	-	-	0.069×10^{-3}
Ni	0.303×10^{-3}	-	0.267×10^{-3}	-	-	-	0.293×10^{-3}	-	0.273×10^{-3}	0.221×10^{-3}
As	-	-	-	1.834×10^{-3}	1.275×10^{-3}	-	1.490×10^{-3}	1.261×10^{-3}	0.476×10^{-3}	0.985×10^{-3}
Pb	0.067×10^{-3}	0.054×10^{-3}	0.058×10^{-3}	0.050×10^{-3}	0.060×10^{-3}	0.023×10^{-3}	0.038×10^{-3}	0.017×10^{-3}	0.009×10^{-3}	0.022×10^{-3}

Conclusion

The deterioration of environment due to various industrial activities have adverse effect on human health has emerged as a major problem all over the world, specially in a developing country like Bangladesh. The wastages containing different heavy and toxic elements are being mixed with the crops field and accumulated in vegetables and other plants and hence the agricultural products as well as human health are largely affected due to these toxic element contaminations. Present study was sketched to determine toxic element (Fe, Cu, Mn, Zn, As Cr, Ni,Pb) concentration in different vegetables available in Bangladesh and their possible health risk effect to the consumers. Study of elemental toxicity transfer process from soil to vegetables also carried out to find out possible source of elemental contamination in the analysed vegetables. The experimental results have been compared with the set value by FAO and WHO, Food & Nutritional Board and US EPA[29,30].

Mean concentration of Cr, Mn, Fe, Ni, Cu, Zn, As and Pb in soil sample are within the World Average Value. Results from Pearson correlation value indicate that Cu, Fe, Zn and Pb have significant co-relation and are originated from natural and somewhat anthropogenic sources. In case of vegetable samples, concentration of Cr, Mn, Fe, Ni, Cu and Zn are found within the World Average value but concentration of As and Pb are much higher than the World Average Value. For all types of vegetables TF value showed a value lower than 1. Highest MPI value was found in Ribbed Gourd and lowest was found in Teasle Gourd and all case MPI value was too high, therefore suggested to avoid those vegetables by the nearby residents. Calculation of Health Risk Index (HRI) was also done and found that arsenic (As) and lead (Pb) have HRI much higher than the safe value 1. The calculated Hazard Index(HI) followed a decreasing sequence of PO>GS1>GT>CH>RG>SG2>PL>BR>GP>TG. The non-carcinogenic risk from the consumption of vegetables for the adult were calculated for the toxic elements Cr, Ni, As and Pb and observed a value lower than 1. However the THQ value for As and Pb were very high for particular types of vegetables and can pose serious health effect. TCRs level for all the contaminants (Cr, Ni, As, Pb) were above 10-4 and hence are unacceptable suggested by USEPA et al. [13], USEPA et al. [26]. The study as a whole revealed that consumers might experience adverse health effect due to directly intake of these vegetables. From the point of food safety and health care it is suggested to do careful monitoring and necessary enforcement of legislation and laws to avoid any kind of adverse effect[31].

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DOI: 10.32474/OAJBEB.2019.03.000159