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Heavy metal contamination of soil and vegetation in ambient locality of ship breaking yards in Chittagong, Bangladesh

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Abstract : *Chittagong is known for the ship scrap processing region of Bangladesh. The heavy metals released from this dirty work are responsible for the increasing stress on the agriculture of this area. The objective of this study was to determine the accumulation of heavy metals in soil and translocation in rice and edible parts of vegetables commonly grown in this area. Results revealed that the mean levels of soil Cd, Cr, Cu, Mn, Ni, Pb and Zn in soils were above the background levels and maximum concentration exceeded the guidelines for metals in soil. Soil pH and organic carbon content had no significant effect on metal accumulation in soil of this area. Mustard as tender leafy vegetable growing near the scrap processing area had accumulated heavy metals 1.47 mg Cd kg^{-1}, 1.97 mg Pb kg^{-1} and 32.85 mg Zn kg^{-1} respectively which were significantly ($p<0.05$) higher than the recommended maximum tolerable levels proposed by the joint FAO/WHO expert committee on food additives. Results showed that rice and leafy vegetables had the highest capacity for heavy metal enrichment among grain, legume, solanaceous, root and leafy vegetables. Among the four heavy metals: Cd, Cu, Pb and Zn, Cd had the highest capacity for transferring from soil into grain, with transfer factor of rice were more than 250-fold than those of Cu and Pb. Concentration of metals in soil and vegetation with their respective transfer factor will provide baseline data of this area.*
Keywords: *Accumulation, contamination, heavy metals, ship scraps, transfer factor.*

I. Introduction

The ship breaking industry extends over 14 km along Fauzdarhat to Kumira coast of Chittagong, Bangladesh. Approximately 27,000 workers are working round the year in these ship yards [1]. Large percentages of them live with their families in the localities close to the ship breaking yard. Ship breaking industry decomposes end-of-life ships into steel and other engineered products for domestic consumers, recycling every part of the hull and machinery [2]. Heavy metals are found in many parts of ships such as in paints, coatings, anodes and electrical equipment (TABLE 1). Of the metals, mercury, lead and cadmium are of the greatest concern because of their ability to travel long distances in the environment [3].

Ship scraps processing sites are usually located in fields adjacent to land used for agricultural purposes in this area. The operations commonly used in processing ship body are strong acid leaching and the open burning of dismantled components. Metal released from the processing of ship scrap may enter the surrounding paddy fields, vegetable fields and vegetable gardens through air deposition, irrigation and rain water. The sediments and soil at the ship breaking yards are highly contaminated with mineral oil and other pollutants from present and historic ship breaking activities [4] and are exposed to dynamic tidal and wave action over the long term. Sea water at high tides submerges beach and near-shore ship breaking areas; bring the pollutants to the villages which inundate the ground including agricultural fields and ponds. Such redistribution of metals to the ambient locality as well as on site heavy metal pollution may pose direct human health hazards through the transfer of pollutants to the food chain. Plants can take up these metals from soil by their roots, transport them upwards to their shoots, and finally accumulate them inside their tissues.

The impact of hazardous substances, including heavy metals on beach soil and marine environment had been studied extensively [5, 6, 7, 8, 9, 10, 11]. Similarly, higher concentrations of cadmium, copper, lead, nickel and zinc were recorded in the urban areas of Chittagong, Bangladesh [11, 12], but there is no work on soil pollution in the nearby locality where huge population are staying in relation to ship breaking activities. The present study was carried out in the adjacent village of ship recycling area especially along the Dhaka-Chittagong highway where local economy is mainly dependent on the ship breaking industry, agriculture and metal recycling from obsolete ship products. There are thousands of small unauthorized workshops, where operations are going on all the year round. Most uncontrolled ship scrap processing sites are located in or close to agricultural land where rice and vegetables are regularly grown. The potential contamination of these local food items may pose a significant threat to local residents. The objective of the present research was to investigate the effect of uncontrolled ship recycling activities on the soils and plants of ambient locality, especially by the important metal contaminants. It was hypothesized that these soils may contain huge amounts of different heavy metals and that may also affected the food cycle. The results can be useful for selecting appropriate clean-up measures for the deteriorated environment.

Table1 Metallic substances that inherited in the ship's structure

Metallic substances	Location on the ship
Arsenic, As	Ballast water
Cadmium, Cd	Bearings, batteries, anodes, bolts and nuts, paints and coatings, metal fumes from cadmium-coated steel
Chromium, Cr	Chromium in paints (lead chromate), ship's coatings and structure
Copper, Cu	Electric wire, cathode protector, anodes
Iron, Fe	Body of the ships, anodes, metal fumes of iron oxides
Lead, Pb	Connectors, couplings, bearings, paints and coatings, cable insulation, lead ballast generators, motor components, piping Batteries: emergency, radio, fire alarm, start up, lifeboats
Mercury, Hg	Thermometer, electrical level switches, luminescent lamps, fluorescent light tubes, fire detectors and tank-level, anodes
Nickel, Ni	Stainless steel, copper-nickel alloys, aluminum alloys, and nickel-based alloy, used in electro-plating and welding products
Zinc, Zn	Anodes, paints and coatings, metal fumes of zinc oxides

II. Materials and Methods

A segment of area covering the villages beside the ship yards to the stalls selling the goods and scraps from ships along Dhaka- Chittagong highway in Sitakund, Chittagong was selected for the study. Sitakund has a tropical monsoon climate [14]. The average maximum temperature is 32.3ºC during May, and the minimum, 13.9ºC in January. The annual average rainfall is 2877 mm. The active uncontrolled processing of ship scraps has left open incineration sites scattered among agricultural fields, and ship wastes dumped beside the ponds. Amid the ship scrap recycling activities, agricultural operations, such as planting rice and vegetables were still taking place in the affected area. Spots for soil and plant sampling were selected on the basis of visual activities of ship scarping (Fig. 1). Soil and plant samples were collected from agricultural fields adjacent to ship scrapping operational sites. These areas are continuously receiving discharge from working area. There were thirty sampling sites in different distances from the ship breaking yard. All of the sampling locations in the present study can be classified into three different groups: paddy field, vegetable field and vegetable garden near homestead area.

2.1 Soil and vegetable sampling

The study was carried out during the dry seasons of 2014. Eight rice and twenty two vegetable samples and their corresponding soils (top soil, 0–15 cm) were collected randomly. Soil sampling was done with a stainless steel spade. All of the samples were put in polythene bags and transported to the laboratory on the day of sampling. The soil samples were air-dried and gently crushed to pass through 2 mm mesh sieve. Edible parts of the plant samples were thoroughly washed to remove all adhered soil particles. Plant samples were cut into small pieces, air-dried for 2 days and finally oven-dried at 65°C in a hot-air oven for 24 hours. Dry plant samples were ground in warm condition and passed through 1 mm sieve.

Figure 1 location of the study area [19].

2.2 Analysis

The total concentrations of metals were determined by Atomic Absorption Spectrophotometer (Aligent 240) after strong acid digestion (1:1 mixture of concentrated nitric and perchloric acids ($v\ v^{-1}$)) of about 200 mg of ground plant and soil samples. The digested samples were filtered and collected in 5 ml of 2.0 M HCL as in [15]. Particle size distribution of soil was determined by the Hydrometer method after treatment with 300 g kg^{-1} H_2O [16]. Textural classes were determined using a triangular co-ordinate diagram. The pH of the soil samples was determined in the laboratory (dry soil and distilled water ratio of 1: 5) and measured by using a Corning glass electrode pH meter [17]. Organic carbon content of the soil samples was determined by wet oxidation with 1 N $K_2Cr_2O_7$ solution and concentrated H_2SO_4 mixture, followed by rapid titration with 1N $FeSO_4$ solution, as recommended by Nelson and Sommers [18]. Total nitrogen content in soil was determined by the Micro-Kjeldahl method following H_2SO_4 acid digestion and alkali distillation [17].

2.3 Contamination assessment

The assessment of soil contamination can be carried out in many ways. In this work, the index of geo-accumulation (Igeo) and transfer factor (TF) have been applied to assess heavy metals distribution and contamination. The Igeo, was proposed to assess the pollution levels of bottom sediments by Müller in 1969 [20]. Igeo can also be applied to the assessment of surface soil contamination by heavy metals [21, 12]. This index (Igeo) of heavy metal is calculated by computing the base 2 logarithm of the measured total concentration of the metal over its background concentration using the following mathematical relation:

$$Igeo = \log_2 (Cn/1.5Bn) \qquad (1)$$

where, Cn is the measured total concentration of the element in the mud grain size fraction of sediment, Bn is the average concentration of element in shale (background) and 1.5 is the factor compensating the background data (correction factor) due to lithogenic effects. The background concentrations of heavy metals are used from [22]. The calculated Igeo are interpreted as $Igeo<0$ = practically unpolluted, $0<Igeo<1$ = unpolluted to moderately polluted, $1<Igeo<2$ = moderately polluted, $2<Igeo<3$ = moderately to strongly polluted, $3<Igeo<4$=strongly polluted, $4< Igeo<5$= strongly to extremely polluted and $Igeo >5$ = extremely polluted [23]. TF of metals from soil to the edible parts of a vegetable was calculated as the ratio of the metal concentration (mg kg^{-1} DW) in the plant's tissues to the total metal concentration in soil (mg kg^{1} DW).

2.4 Statistical analysis

The data were statistically analyzed using the statistical pack- SPSS 20.0 (SPSS, USA). A variance analysis ($p < 0.05$) of total metal concentrations among different sampling sites was performed using a one-way ANOVA test. t –test was used to examine the statistical significance of the differences in the mean concentrations of heavy metals among different vegetable samples. The correlation analysis was done by a Pearson correlation, and the level of significance was set at $p < 0.05$.

III. Results and Discussion

3.1 Heavy metals in soils

Heavy metal concentrations in the agricultural soil samples collected from the ambient locality of ship breaking yards in Chittagong ranged from 0.80 to 6.20 mg kg^{-1} Cd, 54.40 to 94.50 mg kg^{-1} Cr, 120.32 to 426.52 mg kg^{-1} Cu, 61.72 to 2103.00 mg kg^{-1} Fe, 75.70 to 89.50 mg kg^{-1} Ni, 316.98 to 1388.25 mg kg^{-1} Pb and 91.44 to 154.03 mg kg^{-1} Zn. The mean concentrations of heavy metals in control (uncontaminated) site were 0.04, 24.00, 6.00, 3.30, 56.10, 28.00 and 24.00 mg kg^{-1} respectively (TABLE 2). Higher concentrations of metals were observed in all sites as compared to the concentration of heavy metals found at the control site.

The vegetable gardens situated in homestead areas near incineration sites, electroplating industries and shops of ship scraps, had the highest concentrations of metals (TABLE 3). The values greatly exceeded the maximum permissible agricultural soil concentration [24], and highlighted the significant impact of ship breaking activities on these spots. In paddy fields maximum concentration of Pb, Cr and Ni exceeded the maximum permissible limit and in vegetable fields maximum concentration of Cu and Pb exceeded the maximum permissible limit; other metals fall within the range of permissible limit. Only the average values of Cr and Ni were below the recommended maximum levels in vegetable garden [24]. The Cd content was highest (6.20 mg kg^{-1}) in the soils of vegetable garden followed by the soils of paddy field (1.16 mg kg^{-1}). The highest value of Cr was 94.50 mg kg^{-1} recorded at vegetable field.

Pearson's correlation matrix for analyzed soil heavy metals was calculated to see if some of the metals were interrelated with each other and the results are presented in TABLE 3. All of the metal pairs showed significant correlation with each other, other than between Cr with Cu and Ni, Cu with Ni and Zn, Fe with Pb. High correlations between specific heavy metals in soil may reflect similar levels of contamination and/or release from the same sources of pollution, mutual dependence and identical behavior during their transport in the system [5, 25, 26]. In agricultural soils, the major input of heavy metals is the application of agrochemicals and other soil amendments [27]. According to the correlation study, ship scrap recycling activities in the present study and neighboring coastal communities were the dominant source of metal pollution in the vicinity.

Table 2 Heavy metal concentration (mg kg^{-1} dry weight) in soils of three major cultivation areas in ambient locality of ship breaking yards in Chittagong, Bangladesh.

Area		Cd	Cr	Cu	Fe	Ni	Pb	Zn
		mg kg^{-1} dry weight						
Paddy field (n= 8)	Mean ±SD†	1.16±0.34	54.40±8.30	39.13±47.55	61.72±25.00	75.70±24.40	316.98±47.28	91.44±11.40
	Range	0.73-1.86	15.65-190.00	76.00-88.20	30.32-80.50	9.36-108.00	226.00-368.00	78.00-116.00
Vegetable field (n= 10)	Mean ±SD	0.80±0.33	94.50±6.00	120.32±38.76	1712.00±56.00	77.78±4.20	528.81±182.42	127.72±47.44
	Range	0.21-1.12	10.70-122.00	28.81-190.17	495.00-1846.00	12.18-112.70	148.70-748.67	41.73-214.66
Vegetable garden (n= 12)	Mean ±SD	6.20±4.05	80.20±56.00	426.52±315.79	2103.00±436.00	89.50±60.00	1388.25±980.00	154.03±64.41
	Range	1.63-14.60	36.50-160.00	101.68-919.02	43.00-2199.00	83.00-120.00	941.67-1694.33	93.53-331.56
Control site‡		0.04	24.00	6.00	3.30	56.10	26.00	24.00
Permissible limits§		5	100	100	-	100	100	300

†SD= Standard deviation of the replications
‡Metal background value in the soil of uncontaminated site.
§Maximum permissible agricultural soil concentration in some European countries [24].

Table 3 Pearson's correlation coefficients of soil heavy metals, collected from the ambient locality of ship breaking yards in Chittagong, Bangladesh.

	pH	OC	TN	Clay	Cd	Cr	Cu	Fe	Ni	Pb	Zn
pH	1	0.330***	0.323***	-0.994	-0.011	0.078	-0.055	-0.611	-0.051	-0.242	-0.271
OC		1	0.935***	-0.261	-0.569	-0.286	-0.548	-0.612	-0.537	-0.852	-0.496
TN			1	-0.243	-0.644	-0.074	-0.612	-0.612	-0.380	-0.887	-0.483
Clay				1	-0.051	-0.057	-0.001	0.574*	0.044	0.165	0.226
Cd					1	0.490**	0.994*	0.613**	0.508***	0.883**	0.881***
Cr						1	0.542	0.412*	0.835	0.394**	0.629**
Cu							1	0.663*	0.549	0.877*	0.912
Fe								1	0.516*	0.755	0.757**
Ni									1	0.607**	0.655*
Pb										1	0.814**
Zn											1

***Correlation is significant at the 0.001 level, **Correlation is significant at the 0.01 level, *Correlation is significant at the 0.05 level
OC= Organic carbon TN= Total nitrogen

3.2 Index of geo-accumulation (Igeo)

Average Igeo of different metals in paddy, vegetable field and vegetable garden soil are given in Fig. 2. Igeo is distinctly variable, and suggests that soil around the ship breaking yard ranged from uncontaminated to strongly/extremely contaminate with respect to the analyzed metals. Among the metals, high Igeo values which denotes strongly to extremely polluted conditions are found for Pb, Cd and Cu that are mainly in association with ship breaking industries. According to Igeo, it can be seen that the extent of soil pollution is the least significant for Zn, Cr and Ni despite the influence of ship yard activities.

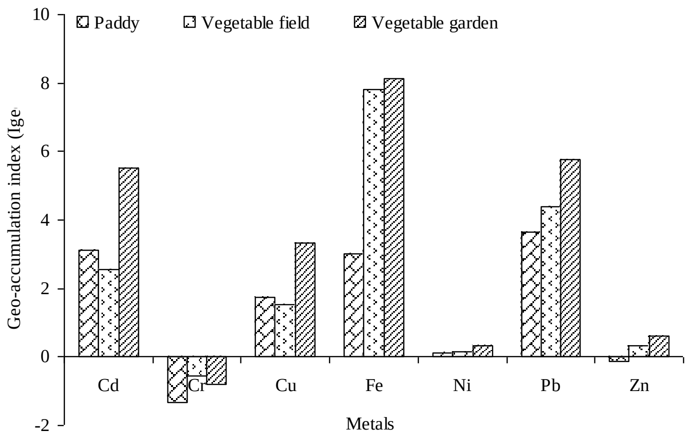

Figure 2 Geo-accumulation index (Igeo) of heavy metals in agricultural soils from the ambient locality of ship breaking yards in Chittagong, Bangladesh.

3.3 Soil properties

Absorption and accumulation of heavy metals in plant tissue depend upon many factors, including temperature, moisture, organic matter, pH, and nutrient availability. In the present study, the pH of most soils ranged between 4.95 and 5.90, with only several samples from the paddy fields having a pH of more than 6.00. The concentrations of organic carbon and nitrogen were the highest for samples from control, lower in samples collected on the paddy and vegetable field, and lowest in samples collected from the vegetable garden (TABLE 4). Organic complexing molecules serve as carriers of micronutrients which have been shown to increase Cd uptake [28], whereas the presence of organic matter has been reported to increase the uptake of Zn in the wheat plant [29]. Soil factors- pH, organic carbon and total nitrogen (TABLE 3) were significantly correlated to each other ($P < 0.001$) but no significant ($p < 0.05$) correlation of organic matter and pH was found with metal content in soil.

Table 4 Basic properties of the soil collected from ambient locality of ship breaking yards in Chittagong, Bangladesh

	Paddy	Vegetable field	Vegetable garden	Control
pH	6.00±0.03d	4.95±0.02a	5.40±0.01b	5.90±0.06c
OC%	0.57±0.02b	0.54±0.03b	0.47±0.01a	0.58±0.03b
N%	0.87±0.01a	0.81±0.03b	0.63±0.03c	1.29±0.03d
Clay%	22.00±0.05a	24.50±0.01d	23.25±0.01c	22.50±0.01b
Texture	Loam	Loam	Loam	Loam

The value shown is mean±S.D.
The different small letters stand for the mean difference is significant at the 0.05 level among the sampling locations.

3.4 Concentration of metallic compounds in paddy and different vegetables

The concentrations of heavy metals (mg kg^{-1}DW) in the edible plant samples are given in TABLE 5. The concentrations of metals varied greatly among plant species and sampling locations (Fig. 3). These results demonstrate that Cd, Pb and Zn were the dominant metal pollutants accumulated in edible part of rice and vegetables grown in agricultural soils in the ship breaking region as referred to the Codex Alimentarius Commission (CAC) standard values [30] (TABLE 5). In contrast, the Cu concentrations of all vegetable samples were below the CAC standard values. The concentrations of Pb and Zn in rice exceeded the food safety limit by FAO/WHO with the average levels of Pb and Zn being 47 and 5 times that of the maximum permissible level, respectively (TABLE 5). Concentrations of Cd and Cu were the highest in rice grain which were similar to the baseline of permissible level. No Cu contamination was detected in any vegetables. Regarding metal concentrations in vegetables, Cd, Pb and Zn occurred at high levels in leaf vegetables while the Cu content of legume was higher than other metal contents.

Table 5 The concentrations (mg kg^{-1} DW) of metals in the edible parts of different vegetables.

Sampling areas	Vegetable type	Edible part	Vegetable species	Cd	Cu	Pb	Zn
Paddy	Cereal grain	Rice grain	*Oryza sativa L.*	0.39±0.05	43.04±3.41	14.23±0.54	102.67±8.99
Vegetable field	Legume	Bean	*Lablab purpureus L.*	0.01±0.00a	1.00±0.16c	0.03±0.01a	2.63±1.24b
		Pea	*Pisum sativum L.*	0.03±0.02b	1.50±0.35d	0.04±0.00a	5.17±0.14c
	Solanaceous	Tomato	*Solanum lycopersicum L.*	0.01±0.00a	0.37±0.09ab	0.04±0.01a	0.99±0.03a
	Root	Radish	*Raphanus sativus L.*	0.01±0.00a	0.65±0.05b	0.17±0.09b	3.18±0.52b
	Leafy	Black Gram	*Vigna mungo L.*	0.00±0.00a	0.96±0.04c	0.02±0.01a	2.09±0.55b
		Hearts pea	*Cardiospermum halicacabum L.*	0.04±0.01b	0.11±0.04a	0.29±0.07c	14.66±0.78d
Vegetable garden	Legume	Bean	*Lablab purpureus L.*	0.03±0.03a	1.37±0.64cd	0.05±0.02a	4.17±1.03bc
		Pea	*Pisum sativum L.*	0.04±0.03a	2.02±0.02e	0.06±0.01a	6.37±0.50c
	Solanaceous	Tomato	*Solanum lycopersicum L.*	0.06±0.03a	0.60±0.19ab	0.09±0.01a	1.39±0.11a
		Hot pepper	*Capsicum frutescens L.*	0.05±0.04a	0.74±0.22b	0.06±0.01a	2.98±0.73ab
	Root	Radish	*Raphanus sativus L.*	0.02±0.01a	0.16±0.03a	0.33±0.07b	4.29±0.36bc
		Sweet potato	*Ipomoea batatas L.*	0.15±0.06a	0.04±0.03a	0.72±0.13c	4.77±0.21bc
	Leafy	Cabbage	*Brassica oleracea var. Capitata L.*	0.04±0.02a	0.78±0.07bc	0.61±0.22c	8.90±0.97d
		Mustard	*Brassica sinapis* L.	1.47±0.81c	1.54±0.40de	1.97±0.01f	32.85±3.36g
		Vine spinach	*Basella alba L.*	0.47±0.07ab	1.03±0.18bc d	0.94±0.04d	20.74±0.31f
		Water Cress	*Enhydra fluctuans L*	0.63±0.07b	0.91±0.07bc	1.20±0.15e	13.58±1.97e
			Permissible levels§	0.4	40.00	0.3	20.00

The value shown is mean±S.D.
§The different small letters stand for the mean difference is significant at the 0.05 level within the sampling location.The tolerance limit of contaminants in vegetables [30, 34]

The concentrations of all the heavy metals in the edible part of leafy vegetables were significantly higher than in the edible portion of legume, solanaceous and root vegetables ($p < 0.05$) , which are in agreement with previous reports [31, 32, 33]. Leafy vegetables usually grow quickly and have high transpiration rates. This favors the uptake of metals by roots and the resulting translocation of metals from roots to above- ground tissues. In addition, their broad leaves make these plants more susceptible to physical contamination by dust from soil and the splashing of rainwater. But this relation was not true for vegetable field.

3.5 Transfer factor

The transfer factor (TF) is one of the key components of human exposure to metals through the food chain. Plant species have a variety of capacities to remove and accumulate heavy metals [35]. Metals with high TF are more easily transferred from soil to the edible parts of plants than ones with low TF. The extent of metal enrichment was higher in cereal (0.101) than in vegetables. Among the vegetables, extent of metal was the highest in leaf vegetables (0.073), followed by legume (0.014), root (0.014) and solanaceous (0.008) vegetables (Fig. 4) [36]. The TF value for Cd was the highest among the selected four metals, and was more than 250 times the TFs of Cu and Pb in cereal. The TF of Zn was 6 times less than Cd.

The mobility of metals from soil to plant is a function of the physical and chemical properties of the soil and of plant species, and is altered by innumerable environmental and human factors [37]. The highest TF values are found for Cd and Zn because these metals are more mobile in nature. Cd occurs with Zn in nature and Cd (II) is retained less strongly by the soil than other toxic cations [36, 38]. The result also supports the findings that accumulation of Pb was comparatively less than that of Cd in plants [39].

The long-term activity of ship breaking yards has resulted in a significant buildup of heavy metals in the soil and their subsequent transfer to the food chain. The extremely high concentrations of metals were probably due to the burning of metal parts in the recycling process. When ship scrap was burned, heavy smoke containing various kinds of heavy metals, metalloids, and organic pollutants would have been discharged into the air, and the plants growing on the sites could be the first recipients of these substances. The elevated levels of Pb in vegetables near ship breaking yard may be attributed to long term use of contaminated waters, high levels of Pb in these soils, nearness of the fields to highways and atmospheric deposition. In the ship scraps recycling sites, ship waste combustion locations are usually close to ponds and streams because these provide a convenient supply of water for metal extraction processes. Metals leached out from the sites. In addition, it is very common for ship scraps to be dumped beside ponds, and metals in these scraps could enter with rainwater into soil and water systems. Therefore the elevated concentrations of metals may be due to long term use of contaminated waters for irrigation and the drainage of ship wastes and waste water to agricultural lands where crops were grown.

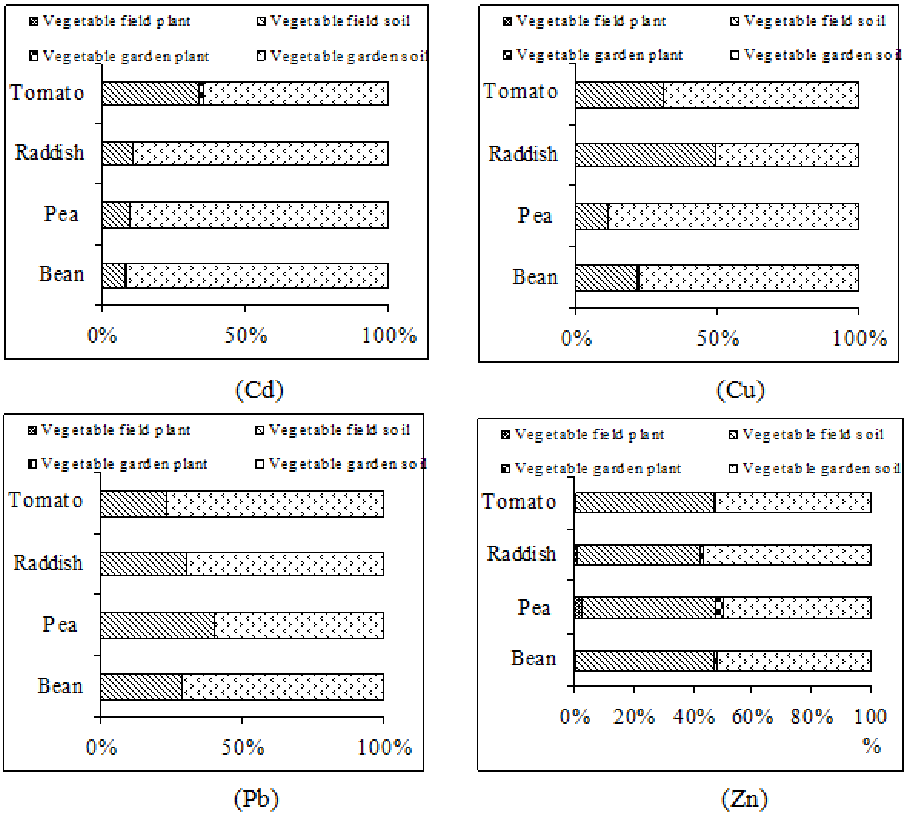

Figure 3 Distribution of heavy metal enrichment in agricultural soils from the ambient locality of ship breaking yards in Chittagong, Bangladesh.

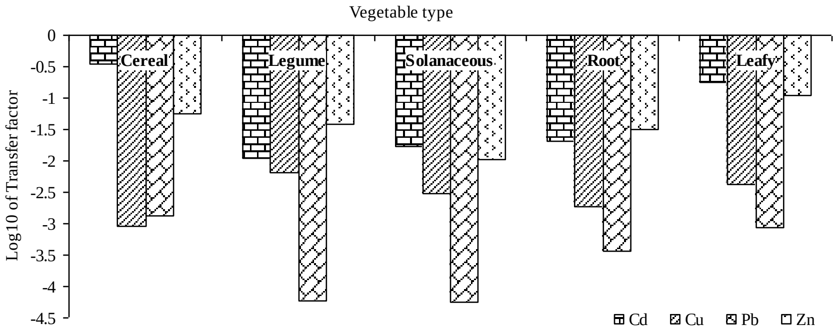

Figure 4 The transfer factor (TF) values of heavy metals in vegetables collected from the ambient locality of ship breaking yards in Chittagong, Bangladesh.

IV. Conclusion

The concentration of metal contaminants in vegetables is a great concern to local residents because vegetables produced from gardens are mostly consumed locally. The prolonged human consumption of unsafe concentrations of heavy metals in foodstuffs may lead to the disruption of numerous biological and biochemical processes in the human body. The concentrations of Cd, Cu, Pb and Zn in the rice grains were extremely high. Therefore, it may be speculated that the rice grains produced around the studied ship breaking area could be a significant source of toxic metals to local residents. The exposure to two or more pollutants from consuming rice may result in additive and/or interactive effects [40, 41, 42]. If the whole intake of metals through dietary means (vegetables and rice) is taken into account, the potential health risks involved in the consumption of local food is alarming. This has important implications for policy in the programmes that aimed at monitoring and controlling heavy metal concentrations in soil and plant, which is a prerequisite in order to prevent potential health hazards of the people of the ambient localities of ship breaking yard.

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