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Assessment of Bioaccessibility and Health Risks of Toxic Metals in Roadside Dust of Dhaka City, Bangladesh

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Abstract: Spatial variations in the bioaccessibility and health risks induced by chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As) and lead (Pb) in roadside dust from different land-use areas, i.e., commercial areas (CA), planned residential areas (PRA), spontaneous residential areas (SRA) and urban green areas (UGA) in Dhaka city, Bangladesh, were investigated. An in vitro simple bioaccessibility extraction test (SBET) method, which allows the simulation of the gastric (GP) and intestinal phases (IP) of human digestion, was applied to evaluate bioaccessibility and human health risk, assessed using United States Environmental Protection Agency (U.S. EPA) modelling. The average bioaccessible concentration of Zn was the highest in both the gastric (74.4–244.5 µg/g) and intestinal phases (74.4–244.5 µg/g) in all the land-use areas except UGA. The bioaccessibility percentages of Co and Cu in the IP phase and As in the GP phase were >40% for all the land-use categories. Carcinogenic (Cr, Ni, As and Pb) and non-carcinogenic human health risks were evaluated for the ingestion pathway, in both children and adults. The results suggest that there were no non-carcinogenic risks for adults and children exposed to roadside dust toxic metals, but the risk levels of roadside dust toxic metals in some sampling areas were high. The carcinogenic risks of Cr in SRA (for children) and Ni in CA (for both adults and children), PRA (for children) and UGA (for children) were found to be within a tolerable range of 10^{-6} to 10^{-4}.

Keywords: roadside dust; toxic metals; bioaccessibility; health risk assessment; land use area

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1. Introduction

Road dust is considered a host media for potentially toxic elements in emissions into the atmospheric environment. The accumulation of dust on the surface of the road, which contains excessive amounts of potentially toxic elements compared to the background value (upper continental crust), is said to cause adverse health and environmental risks [1,2]. Toxic metals that exist in road dust affect the urban environmental quality as well as human health due to their bioaccumulation, toxicity, and non-biodegradable properties [3]. Toxic metals generated by anthropogenic activities can be accumulated directly or individually in roadside dust through atmospheric deposition and absorption. Among other toxic metals, Cr, Cd, Cu, Zn, Ni, As, and Pb are considered priority control contaminants by the United States Environmental Protection Agency (U.S. EPA) due to their toxic and persistent nature [4]. The natural and anthropogenic sources of the toxic metals in road dust are diverse, and the level of toxic metal pollutants may be influenced by several factors such as land use types, location, vehicular and road roughness types, seasons, and the antecedent dry period [5–8].

Dhaka city is one of the largest metropolitan areas in South Asia, with many anthropogenic pollution sources (emissions from traffic and brick kilns, domestic waste, industries, and dust from constructions activities) [9,10]. Apart from the population explosion and traffic congestion, air pollution has emerged as a major problem, and has created concern among the citizens of Dhaka city. Construction and roadside dust are both major causes of air pollution, according to Bangladesh's Department of the Environment (DoE). The contamination by toxic metals (Pb, As, Cu, Zn, Cr, Hg, Cd, Co and Ni) in the road dust of Dhaka city has been highlighted in previous studies [10–12].

In recent years, road dust pollution studies in Dhaka city have mainly focused on metal content and contamination assessment [13,14], spatial distribution [12], source identification [11], and assessment of health risks [15]. In order to assess human health risks, most of the studies in Dhaka city have considered the concentration of total toxic metals, which underestimate or overvalue the risk as the total amount of toxic metals is not available for absorption in the human body [16] and does not cause harm [17]. As a result, the health hazards caused by metal(loid)s are determined by their solubility and absorption availability in the gastrointestinal environment, which is referred to as metal bioaccessibility [18–20]. This bioaccessibility test mimics the pH of the stomach, which provides an idea of the real toxicity risk of metals in the human body. Additionally, the bioaccessibility of toxic metals has been taken into consideration in calculating the average daily exposure dose through ingestion, and thereafter, the assessment of health risk [21,22].

For the determination of bioaccessible concentrations of toxic metals in road dust, several in vitro methods such as the simplified bioaccessibility extraction test (SBET) [23,24], the solubility bioaccessibility research consortium (SBRC), the physiologically-based extraction test (PBET) [25,26], the in vitro gastrointestinal method (IVG), and the unified bioaccessible assay (UMB) [27,28] have been proposed by several studies. Thus, the in vitro simulation tests provide relatively more reliable results and are easier to maintain in the human body environment, and are also faster and more affordable [29,30]. Among them, the SBET can easily determine the bioaccessible concentration of toxic metals in road dust [31–33] as well as the soil matrix [34–36], and this method is validated by the U.S. EPA [37,38] for investigations regarding in vitro lead bioaccessibility. The SBET is appropriate for large batches of samples since it uses simple reagents in a single extraction test for a short length of time. Typically, the SBET method is used to mimic gastric phase bioaccessibility, but recently the SBET has been used to determine bioaccessibility in both the gastric and intestinal phases [39,40].

In this study, bio-accessible simulations of roadside dust were performed in two sequential phases: the gastric phase (GP) with a low-pH solution (pH 1.5), and the intestinal phase (IP) with a high-pH solution (pH 7). The main objectives of this study were to determine the bioaccessible concentrations of toxic metals in the roadside dust using the SBET model in different land-use areas of Dhaka city and to estimate the potential non-carcinogenic and carcinogenic human health risks of toxic metals through road dust ingestion based on bioaccessible concentrations.

2. Materials and Methods

2.1. Study Site

This study was implemented in Dhaka, a densely populated metropolis and the capital of Bangladesh. The city of Dhaka is located on the banks of the river Buriganga at the heart of the Bengal Delta. With a 4.25% annual population growth (World Population Review Report 2022), (available online: https://worldpopulationreview.com/, accessed on 15 February 2022), Dhaka is the seventh most populated city in the world (World City Population Data 2021) (available online: https://www.jagranjosh.com/general-knowledge/list-of-most-populous-cities-of-the-world-1634730858-1, accessed on 15 February 2022). Over 18 million residents live in Dhaka city and its built-up zone is growing and its green area and environmental conditions are worsening rapidly. In terms of air pollution, Dhaka has remained at the top of the list of recent world cities with the poorest air quality. Moreover, Dhaka is the most polluted city in the world and has been plagued with air pollution for a long time. In winter, its air quality usually becomes unhealthy because of massive emissions of pollutants from construction work, roads, brickyards, and other sources. Due to the long dry periods in Dhaka city in the winter season, a dusty environment is created, which increases the risk of dust ingestion as well as health risks.

On the basis of the Dhaka Urban Transport Network Development Study report, land use in Dhaka city is dominated by residential areas (44.35%), which are a combination of planned residential areas and spontaneous residential areas. Planned residential areas are designed to provide a healthy environment with the provision of all necessary facilities, community services, and infrastructure, whereas spontaneous residential areas are unplanned areas consisting of housing, construction, narrow roads, and mixed uses of the land. The major planned residential areas are Banani, Uttara, Gulshan, Baridhara, and Nikunja, which are occupied by mainly middle- and high-income citizens. There are many spontaneous residential areas in Dhaka city, especially in Kafrul, Badda, Mirpur, Mohammadpur, and Pallabi, which are dominated by local and low-income people. A commercial area of a city is an area, district, or surrounding area predominantly made up of commercial structures, including a city center, central business district, financial district, main street, commercial strip, or shopping center that is primarily used for the exhibition and sale of merchandise. The commercial area occupies 4.29% of the total land in the Dhaka city area. All commercial activities such as trading, buying, and selling points are situated in these areas. The commercial area of Dhaka city is represented by Motijheel, Gulistan, Nawabpur, Khilgaon, and Mugda. Urban green space refers to any land that is covered by vegetation of any kind. Parks/urban green areas make up 1.2% of the total area of Dhaka city and are mainly used for recreational purposes.

2.2. Sample Collection

Roadside dust was sampled from twenty sites representing different types of land-use areas around the city: planned residential areas (PRA) (n = 5), spontaneous residential areas (SRA) (n = 5), commercial areas (CA) (n = 5) and urban green areas (UGA) (n = 5) (Figure 1). The sample collection time was February, 2020 (winter season), just after the driest month of the year so that the rain had not washed away the road dust. Dust samples were taken from the targeted locations on the main roads. Dust was collected by sweeping with a polyethylene brush and pan from both hardened ground shoulders of the road. The selected sites had been dry for at least one week. At each sampling location, at least 6 sub-samples were collected at a distance of 1 m apart from each other. Then, the dust samples were packaged, each sample contained about 300 g of dust. All dust samples were collected in appropriately coded polyethylene bags, sealed on the same day of collection, and sent to the Department of Agronomy, Bangladesh Agricultural University. All the samples were collected on the same day. In the laboratory, the samples were air-dried until their constant mass was obtained and then coarse debris, cigarette butts, leaves, and small stones were removed with a 2.0 mm nylon mesh sieve. After being moved to the Pollution Control Laboratory of Saitama University, Japan, two-thirds of all the samples were sieved with a selected diameter of sieves using the Vibratory Sieve Shaker AS 200-digit Retsch AS200 (the amplitude was 60 and the shaking time was 10 min). The remaining quarter portion was left as backup. We took extra care to limit fine particle excitation, which was quickly lost due to re-suspension during the sampling and screening. Finally, particle size fractions of <32 μm were performed to analyze the bioaccessible concentration of toxic metals (Cr, Mn, Co, Ni, Cu, Zn, As, and Pb) because fine dust particles play a significant role in assessing human health risks as well as toxic metal concentrations [23,24]. Unfortunately, a sample from the UGA was tainted in the laboratory, and we thus ignored it. As a result, we investigated and reported on nineteen samples in this study.

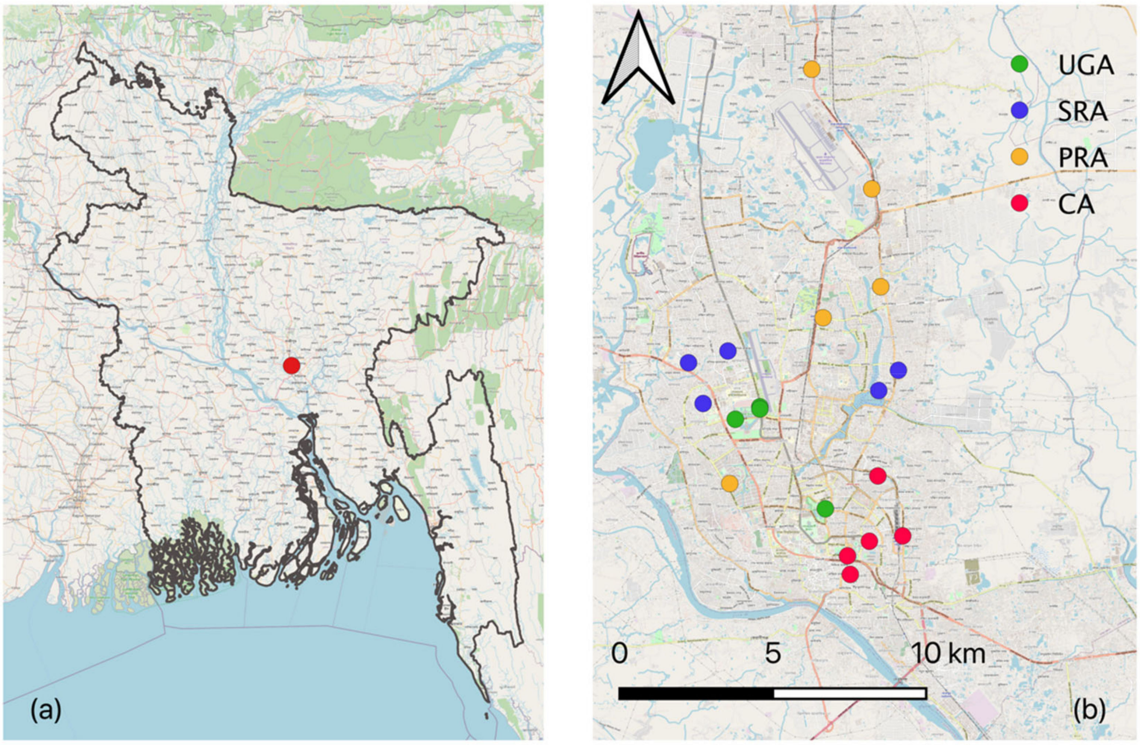

Figure 1. (**a**) Location of the study sites (Dhaka city center; red marked circle) on a map of Bangladesh, and (**b**) map of study area with different land-use categories. CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area.

2.3. Simple Bioaccessibility Extraction Test (SBET)

2.3.1. Preparation of Extraction Fluid

The extraction solution was made by mixing 60.06 g of glycine (free base, reagent grade) to 1.9 L of miliQ (Type I) water. After this, the volume of the solution was increased to 2 L (containing 0.4 M glycine). Afterwards, about 60 mL of concentrated (12.1 N) hydrochloric acid of trace-metal grade was added. This buffered solution was then placed in a water bath at a temperature of 37 °C until the extraction solution reached this temperature. The pH of the solution was then set to 1.50 ± 0.05 by adding HCl drop by drop.

2.3.2. Determination of Bioaccessible Concentrations of Toxic Metals in Roadside Dust

The in vitro bioaccessibility test for toxic metals in the roadside dust samples was conducted by the SBET and executed on particle sizes of <32 μm, which is suspensible and easily ingested into the human body. Two subsamples were examined for each sample: one in the gastric phase (GP) and the other in the gastric phase followed by the intestinal phase (IP). A total of 0.25 g of the roadside dust samples was weighed and added into 25 mL glycine (0.4 M; pH = 1.5 ± 0.05, pre-adjusted with concentrated hydrochloric acid) and maintained at 1:100 in the solid-to-liquid ratio. The samples were extracted by shaking the samples at 70 rpm end-over-end at 37 °C for 1 h in a water bath. The 10 mL aliquot mixture was taken from the mixture and then centrifuged at 3000 rpm for 10 min. Finally, the supernatant was filtered through a syringe filter (PTFE 0.20 μm), which represents the gastric phase (GP) of the extractant. The remaining aliquot mixture was then neutralized with $NaHCO_3$ (pH 7), and the initial volume of the extractant was restored by 0.4 M glycine. The mixture was shaken for 3 h at 37 °C, then centrifuged and filtered in the same manner as described above, which represented the intestine phase (IP) of the extractant. We confirmed that the change in the initial and final pH maximum was 0.5. The filtrates were kept at 4 °C in a refrigerator until they were examined by ICP-MS. The whole digestion process was completed in dark conditions in a 50 mL capped polythene centrifuge tube. The bioaccessible concentrations of the toxic metals were determined by ICP-MS (7700 series, Agilent Technologies, Santa Clara, California, USA) in the Center for Environmental Science in Saitama (CESS), Japan. Calibration curves were prepared by a multi-elemental standard XSTC-662 (Spex Certi Prep Metuchen, NJ, USA), and calibration curves with $R^2 > 0.999$ were permitted for the concentration calculation. The limits of detection (LOD) for Cr, Mn, Ni, Cu, Zn, As, Pb, and Co were 0.018 ng/mL, 0.041 ng/mL, 0.088 ng/mL, 0.048 ng/mL, 0.067 ng/mL, 0.085 ng/mL, 0.085 ng/mL, and 0.009 ng/mL, respectively.

2.4. Toxic Metals Bioaccessibility

Metal bioaccessibility in the roadside dust samples was calculated by dividing the extractable metal from the gastric phase (GP) or intestinal phase (IP) by the total metal content in the dust according to the equation below:

$$\%\text{Bioaccessibility} = (C_{\text{Bioaccessibile}}/C_{\text{Total}}) \times 100$$

where $C_{\text{Bioaccessible}}$ is the concentration of toxic metals extracted from the road-side dust via GP or IP, and C_{Total} is the concentration of toxic metals in the roadside dust obtained via aqua regia digestion [12].

2.5. Human Health Risk Assessment

The health risk of toxic metal uptake from contaminated urban roadside dust was calculated according to the model proposed by the U.S. EPA [41]. The average daily intake (DI) (mg kg^{-1} day^{-1}) of toxic metals through the incidental ingestion pathway was calculated according to following equation:

$$\text{DI} = \text{C} \times \frac{\text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times 10^{-6}$$

where C is the total metal concentration (mg kg^{-1}) in the roadside dust samples.

For an accurate assessment of health risk, bioaccessibility determined by the SBET extraction method was applied to adjust the DI according to the following equation:

$$\text{DI}_{\text{adjusted}} = \text{DI} \times \text{RBA}$$

where RBA is the relative bioavailability adjustment, which was the ratio of metal concentrations (sum of gastric and intestinal), extracted by the SBET method to their total concentration.

The non-carcinogenic risks posed by toxic metals to humans were evaluated by calculating the hazard quotients (HQs). The HQs for individual metal were calculated by dividing $\text{DI}_{\text{adjusted}}$ by the reference dose (R*f*D) (mg kg^{-1} day^{-1}):

$$\text{HQ} = \text{DI}_{\text{adjusted}}/\text{RfD}$$

Since interaction with two or more toxic metal pollutants could have a cumulative or synergistic effect, the sum of the HQs for all the studied toxic metal pollutants can be characterized as the total hazard index risk (HI) for a specific exposure pathway adjustment using the following equation:

$$\text{HI} = \text{HQ}_1 + \text{HQ}_2 + \text{HQ}_3 \ldots + \text{HQ}_n$$

In the present study, HI was calculated to describe the potential risk of noncarcinogenic consequences from a combination of Cr, Mn, Co, Ni, Cu, Zn, As, and Pb. HI or HQ values of less than 1 mean that there is no indication of adverse health effects or noncarcinogenic risks for toxic metals, individually or collectively. If the value of the HI or HQ is greater than 1, it is defined as having the potential for noncarcinogenic effects on human health [41].

The potential carcinogenic risk (CR) for toxic metals was calculated using the following equation:

$$CR = DI_{adjusted} \times CSF$$

where CSF is the cancer slope factor (mg kg^{-1} day^{-1}). In this study, we estimated the carcinogenic risk for Cr, Ni, As, and Pb. The toxic elements Cu and Zn are not carcinogenic to humans, and currently, there are data on CSF regarding the ingestion pathway for Mn and Co. When the estimated values of CR are greater than 10^{-4}, this shows that human tolerance is exceeded, when the CR value is lower than 10^{-6}, it means no cancer risk occurs. When the CR value is between 10^{-6} and 10^{-4}, this indicates that the cancer risk is within a tolerable range. The parameters and values used for the estimation in the human health risk models are presented in Table 1.

Table 1. Parameter definitions and values used in the human health risk model.

Factor	Definition	Unit	Permissible Limit for Children	Permissible Limit for Adult	Reference
ED	Exposure duration	year	6	25	[41]
EF	Exposure frequency	days/year	250	250	[41]
BW	Average body weight	kg	15	61.8	[41]
$AT_{non\text{-}cancer}$	Average time of exposure	days	ED × 365	ED × 365	[41]
AT_{cancer}	Average time of exposure	days	70 × 365	70 × 365	[41]
IngR	Ingestion rate	mg/day	200	100	[41]
RfD	Reference doses	-	3×10^{-3} (Cr); 2.4×10^{-2} (Mn); 2×10^{-2} (Co); 2×10^{-2} (Ni); 4×10^{-2} (Cu); 3×10^{-1} (Zn); 3×10^{-4} (As); 3.5×10^{-3} (Pb)		[42]
CSF	Cancer slope factor	-	5.0×10^{-1} (Cr); 8.5×10^{-3} (Pb); 1.5×10^{0} (As); 1.7×10^{0} (Ni)		[41,43,44]

2.6. Statistical Analyses

The data analyses were carried out using the statistical package 'R' [45]. The data were tested for normality using the Shapiro–Wilk method before the statistical analyses. Non-parametric Kruskal–Wallis tests were performed to compare the variation in metal bioaccessibility between the gastric and intestinal phases and among land-use categories followed by Dunn's post hoc test. Spearman's rank-order correlation method was used to evaluate the correlations between the total and bioaccessible concentrations of toxic metals in the roadside dust.

3. Results and Discussion

3.1. Spatial Variability of Bioaccessible Concentrations of Toxic Metals in Roadside Dust in Dhaka City

The mean (±standard deviation) of the bioaccessible extent of toxic metal (Cr, Mn, Co, Ni, Cu, Zn, As, and Pb) extraction by the SBET method from various land-use areas in Dhaka city are presented in Table 2. The total concentrations of the toxic metals of different land categories in Dhaka city were adopted from those in our previous study [12]. In all the land-use categories, when comparing the GP and IP phases, Ni, As, and Pb were found in higher concentrations in the GP phase, whereas Cr, Mn, Co, Cu, and Zn showed higher concentrations in the IP phase, suggesting that there were significant differences in toxic metal bioaccessibility between the two phases. In this study, considering both the bioaccessible phase and the different land types (except in UGA), Zn existed in higher concentrations (74.4–244.5 µg/g in the GP phase and 85.1–277.7 µg/g in the IP phase) compared to the other studied toxic metals. Thereafter, Mn (76.5–95.9 µg/g in the GP phase and 86.9–104.5 µg/g in the IP phase) and Cu (17.1–68.4 µg/g in the GP phase and

61.2–135.5 μg/g in the IP phase) showed higher concentrations. The average minimum bioaccessible concentrations were observed for Co (0.75–1.2 μg/g) in GP, and Ni showed an average minimum concentration (1.3–6.8 μg/g) in IP for all the land-use areas. However, it is notable that the most noticeable bioaccessibility concentration was found for Co, which was 10.46–15.89 times higher in the IP phase compared with the GP phase. It is worth mentioning that the bioaccessible concentration of As was below detection limit in the IP. The obtained GP bioaccessible concentration in this study was consistent with the results of [31]. The high concentrations (the total and bioaccessible concentrations) of toxic metals (Zn, Cu, and Mn) in the roadside dust indicated that these toxic metals are serious pollutants in Dhaka city. The bioaccessible concentration was correlated with the total concentration of the toxic metals in the roadside dust of Dhaka city, as shown in Figure 2. In the gastric phase, a strong correlation was observed for Co (0.97), Cr (0.95), Cu (0.97), Ni (0.8), Pb (0.95), and Zn (0.98), whereas a moderate correlation for Mn (0.65) and no correlation for As (0.008) were found. The coefficients of determination (r) varied between 0.8 and 0.98 for all the toxic metals evaluated (except Mn and As). As a result, the gastric phase bioaccessible concentrations of Cr, Co, Ni, Cu, Zn and Pb increased in tandem with the total concentrations of these metals. A comparable result was shown in Tehran street dust and surface soil for Cu, Pb, and Zn [24]. A similar, strong correlation was shown in the intestinal phase for Cr (0.87), Cu (0.88), Pb (0.9), and Zn (0.94), and a moderate correlation was observed for Mn (0.52) and Ni (0.70), whereas no correlation for Co (0.13) was obtained in the intestinal phase. Toxic metals (Cr, Cu, Zn, and Pb) from anthropogenic sources were more frequently linked to the intestinal stages.

Table 2. Concentrations of toxic metals and their bioaccessible amounts in different land categories (μg/g) in Dhaka city. (CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area).

Toxic Metals	Content	CA	PRA	SRA	UGA
Cr	Total Concentration	82.45 ± 12.25	95.61 ± 23.09	77.75 ± 6.63	65.08 ± 12.48
	Bioaccessible GP	7.95 ± 0.4	10.21 ± 3.73	7.38 ± 1.55	5.7 ± 1.67
	Bioaccessible IP	18.18 ± 0.87	21.88 ± 3.94	19.78 ± 2.72	17.37 ± 2.88
Mn	Total Concentration	569.98 ± 113.83	507.97 ± 59.03	456.04 ± 47.75	538.28 ± 96.63
	Bioaccessible GP	92.32 ± 10.69	94.41 ± 5.69	76.45 ± 5.29	95.88 ± 25.85
	Bioaccessible IP	96.83 ± 11.11	104.49 ± 7.29	86.86 ± 6.99	100.49 ± 16.44
Co	Total Concentration	9.84 ± 3.06	8.19 ± 1.37	7.46 ± 0.79	8.17 ± 0.73
	Bioaccessible GP	1.2 ± 0.5	1.04 ± 0.24	0.76 ± 0.1	0.75 ± 0.16
	Bioaccessible IP	12.56 ± 0.89	12.39 ± 0.46	11.96 ± 0.21	11.92 ± 0.31
Ni	Total Concentration	57.53 ± 36.89	36.26 ± 4.29	34.11 ± 8.88	44.54 ± 15.80
	Bioaccessible GP	8.19 ± 3.18	5.38 ± 0.35	4.08 ± 0.73	6.79 ± 4.16
	Bioaccessible IP	6.85 ± 4.71	3.76 ± 1.0	1.33 ± 1.44	5.71 ± 7.21
Cu	Total Concentration	227.68 ± 129.11	161.79 ± 66.55	92.19 ± 15.59	75.6 ± 21.65
	Bioaccessible GP	68.35 ± 49.27	38.12 ± 17.0	22.04 ± 3.35	17.12 ± 5.55
	Bioaccessible IP	135.52 ± 82.32	95.62 ± 44.92	61.16 ± 17.91	65.44 ± 18.58
Zn	Total Concentration	686.22 ± 153.92	506.03 ± 65.55	429.23 ± 98.54	295.89 ± 61.11
	Bioaccessible GP	244.45 ± 67.01	151.41 ± 25.77	124.78 ± 34.09	74.36 ± 18.50
	Bioaccessible IP	277.65 ± 76.69	196.55 ± 33.95	143.33 ± 29.48	85.1 ± 27.07

Table 2. *Cont.*

Toxic Metals	Content	CA	PRA	SRA	UGA
As	Total Concentration	5.15 ± 0.99	4.71 ± 0.43	4.68 ± 0.57	4.93 ± 0.63
	Bioaccessible GP	2.99 ± 0.3	2.86 ± 0.29	2.51 ± 0.13	2.42 ± 0.16
	Bioaccessible IP	nd	nd	nd	nd
Pb	Total Concentration	72.11 ± 12.68	73.72 ± 36.11	52.75 ± 10.54	36.72 ± 15.98
	Bioaccessible GP	28.18 ± 5.42	26.4 ± 11.72	17.55 ± 3.98	11.87 ± 7.79
	Bioaccessible IP	8.48 ± 0.24	8.48 ± 0.51	8.01 ± 0.11	7.71 ± 0.18

('nd' represents below the detection limit).

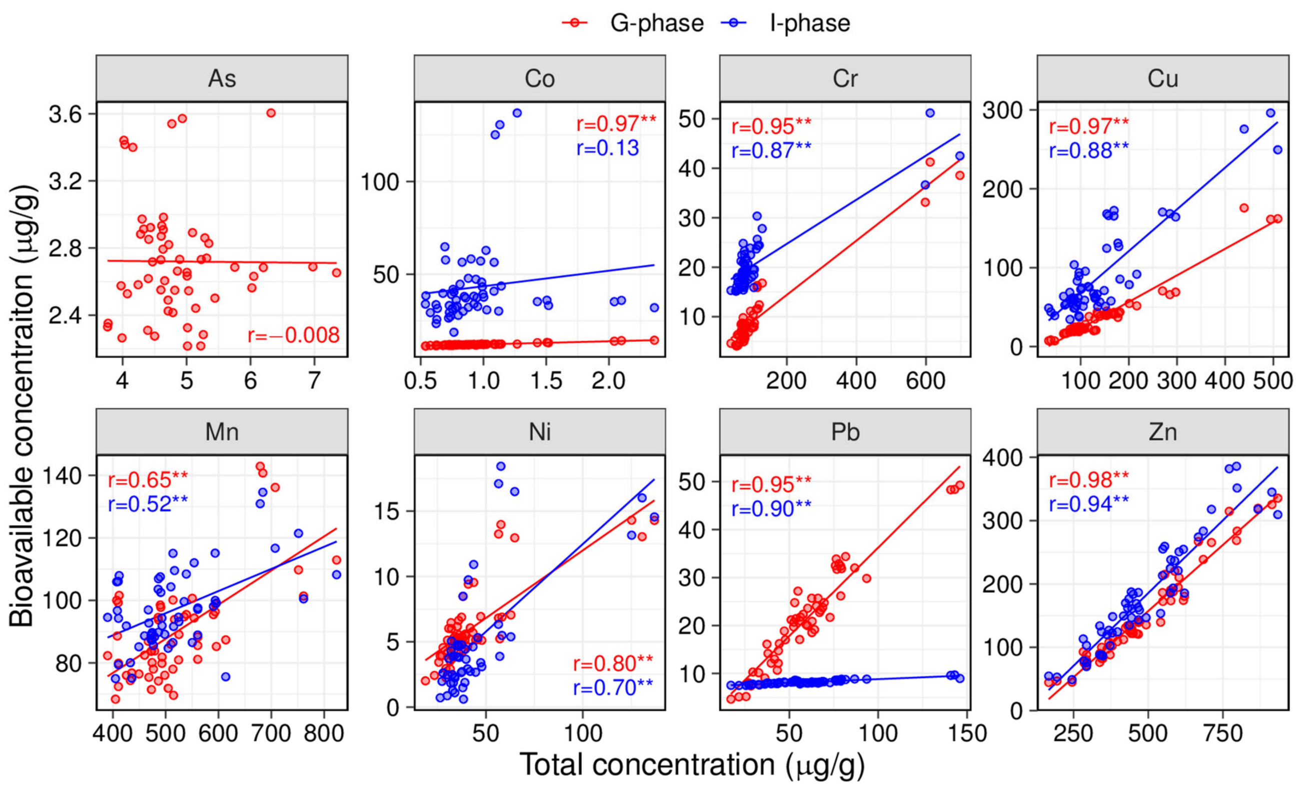

Figure 2. Scatter plot correlation for total concentration (µg/g) versus bioaccessible concentrations (µg/g) of toxic metals of roadside dust in Dhaka city. Double asterisk (**) designates statistically significant at $p = 0.01$.

3.2. Bioaccessibility (%) of Toxic Metals in the Gastric (GP) and Intestinal (IP) Phases

Figure 3 shows the bioaccessibility percentages for Cr, Mn, Ni, Co, Cu, Zn, As, and Pb in the gastric (GP) and intestinal (IP) phases for each land-use category in Dhaka city. The average bioaccessibility percentage of the toxic metals in the roadside dust from four land-use areas were ordered according to the GP as follows: As (56.5%) > Pb (34.8%) > Zn (29.8%) > Cu (24.7%) > Mn (17.5%) > Ni (14.4%) > Co (11.1%) > Cr (9.7%), and in IP as follows: Co (149.6%) > Cu (69.2%) > Zn (35.3%) > Cr (24.8%) > Mn (19.1%) > Pb (16.5%) > Ni (8.8%). Similarly, higher bioaccessibility in the intestinal phase for Cr, Cu, and Mn was reported in other studies [46,47]. These results indicate that the bioaccessibility of toxic metals in the GP and IP phases were significantly different, which could be attributed to the various forms of these metals that may occur in the gastrointestinal tract [48].

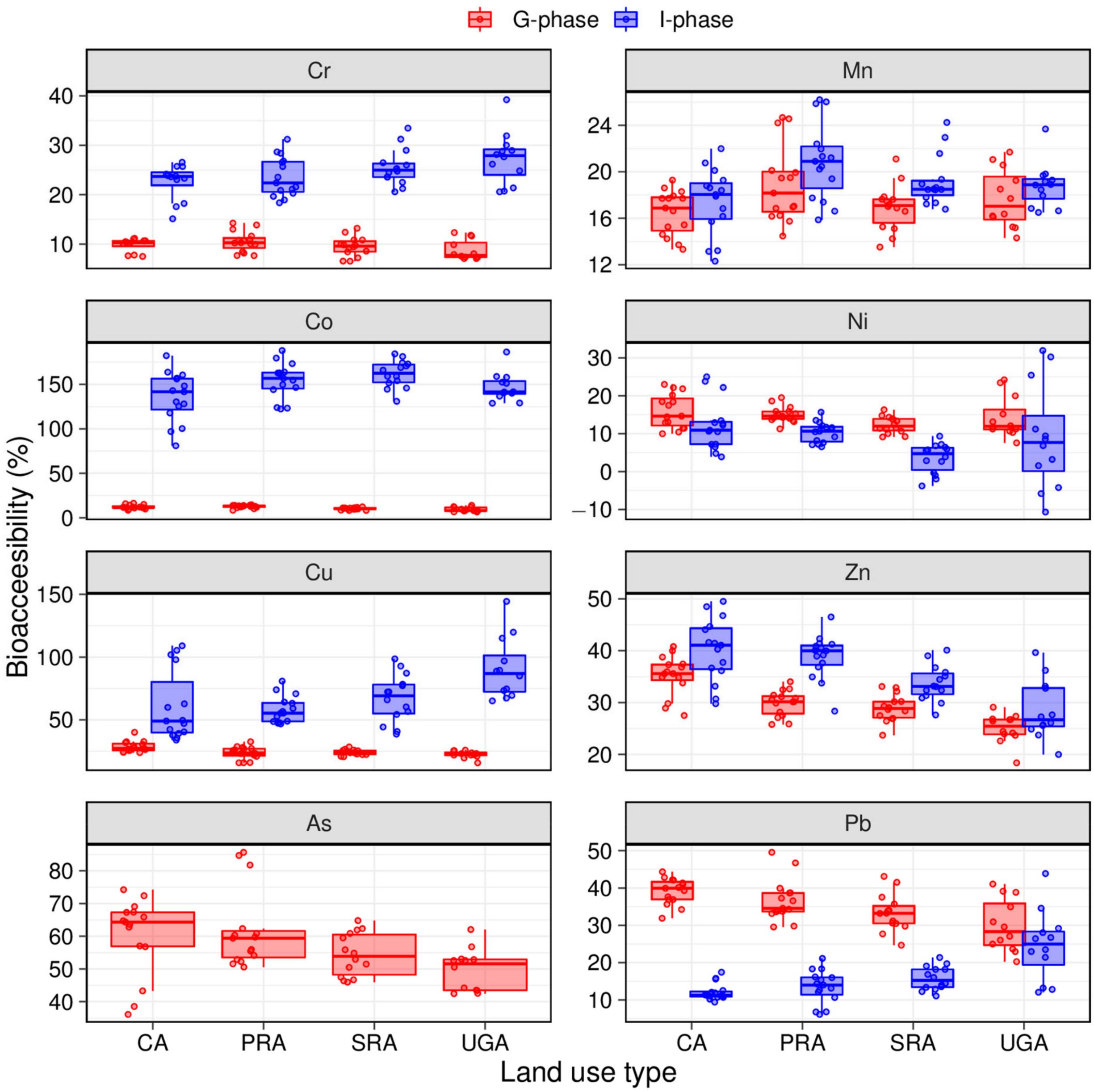

Figure 3. Bioaccessibility of toxic metals in roadside dust in Dhaka city. The circles represent the values of individual sampling points. CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area.

In this study, the average bioaccessibility percentage in the intestinal phase of the toxic metals was higher than the average for gastric phase bioaccessibility in all the land-use categories (Figure 4). Specifically, as mentioned in Figure 3, the bioaccessibility percentages of the toxic metals in the gastric phase had a similar sequence (As > Pb > Zn > Cu > Mn > Ni > Co > Cr) for CA, PRA, SRA and UGA. Among these, the bioaccessibility of As (for all the studied areas) and Pb (for CA) in the gastric phase was more than 40% of their total concentrations, indicating a relatively high health risk [49]. In addition, maximum As bioaccessibility found in the gastric phase suggested that As was more soluble in the gastric phase due to more acidic conditions (pH 1.5) when compared with the intestinal phase (below detection limit) [27]. According to Du et al. [50], Pb's bioaccessibility was reduced considerably from the GP phase to the IP phase, which supported our results. This is because an increase in $NaHCO_3$ in the IP phase may aid in the absorption of Pb into clay edges and the formation of Pb carbonate minerals [51]. Furthermore, pH has an effect on Pb bioaccessibility in the GP [24]. Cr showed the lowest bioaccessibility in the GP, which was similar to the findings in [25] where it was mentioned that Cr developed a residue form and was not easily absorbed by human gastrointestinal fluid [39]. On the other hand, the bioaccessibility percentages of the toxic metals in the intestinal phase were in a similar order to those in PRA and SRA (Co > Cu > Zn > Cr > Mn > Pb > Ni), but had a dissimilar order to those in CA (Co > Cu > Zn > Cr > Mn > Ni > Pb) and UGA (Co > Cu > Zn > Cr > Pb > Mn > Ni). Among these studied toxic metals, Co obtained the maximum percentage of bioaccessibility in the IP phase. However, in the IP, the percent bioaccessibility of Co and Cu was close to or greater than 100%, implying that the simulated solution was at least as effective in solubilizing these two metals. Moreover, the percent bioaccessibility of Cu increased from the GP phase to the IP phase which is consistent with the findings in [25,52] and suggests that a near-neutral pH of IP enhances the solubility of Cu. Moreover, as the pH of the solution increases from 1.5 to 7.0, the Cu in the solution forms an uncharged complex, $Cu(Gly)_2$, increasing Cu bioaccessibility in the IP phase [40]. Besides that, the physiochemical properties of dust and the interactions between toxic metals with road dust constituents can affect metal bioaccessibility [24,53]. For example, Zn bioaccessibility (GP) is influenced positively by organic matter (OM) and adversely by cation exchange capacity (CEC) [24]. Comparing the mean percent bioaccessibility with the GP and IP phases in different land-use areas, As, Co, and Cu showed relatively high levels, suggesting a relatively high health risk after ingestion although As and Co presented low total concentrations in the roadside dust of Dhaka city (Table 2). Cr, Zn and Pb, Ni, and Mn also had considerable levels of bioaccessibility in the human gastrointestinal tract, and their health risk could not be omitted. The mean bioaccessibility of Cu, Zn, and Pb in the GP and IP phases were significantly varied among the different land categories, indicating that the bioaccessibility of toxic metals (Cu, Zn, and Pb) has spatial variability, while Cai et al. [23] found no significant variation in toxic metal bioaccessibility in different functional areas. The bioaccessibility of Pb displayed functional area distribution characteristics in the urban dust of Hangzhou, China, according to Zhang et al. [26]. The bioaccessibility of Pb was mainly dominated by the total Pb concentration, which was similar to our findings for the GP and IP phase. However, Pb bioaccessibility in the IP phase might be reduced by calcium concentration [54], and Han et al. [55] suggested that Pb bioaccessibility depends on pollution sources and the sample matrix.

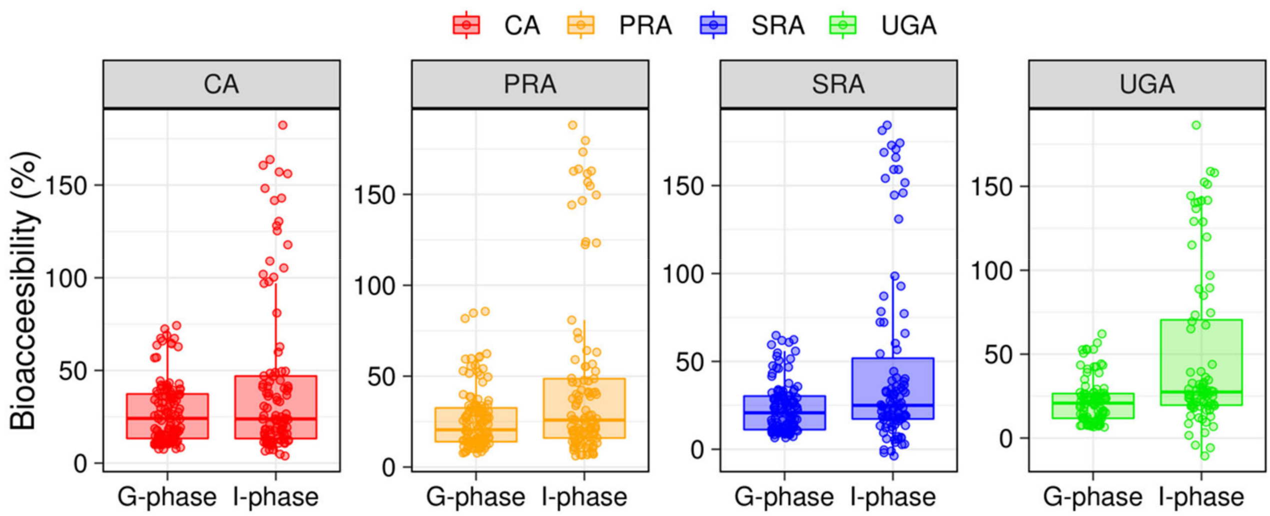

Figure 4. Bioaccessibility of gastric (GP) and intestinal (IP) phases in different land-use areas. CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area.

3.3. Health Risk Assessment of Toxic Metals in Road Dust

The bioaccessible concentrations of each toxic metal were used to assess non-carcinogenic (for Cr, Mn, Co, Ni, Cu, Zn, As, and Pb) and carcinogenic (for Cr, Ni, As, and Pb) risks in roadside dust so that a more accurate risk assessment could be conducted [56,57]. In this study, the health risks of toxic metals in the human body were assessed across the ingestion pathway for adults and children due to high exposure routes [58,59]. Dust containing toxic metals, when inhaled, can reach the gastrointestinal tract, where they are partially dissolved and then carried via the circulatory system and finally accumulate inside the body [60]. The non-carcinogenic health risks from exposure to toxic metals in roadside dust in different land-use areas in Dhaka city are presented in Figure 5.

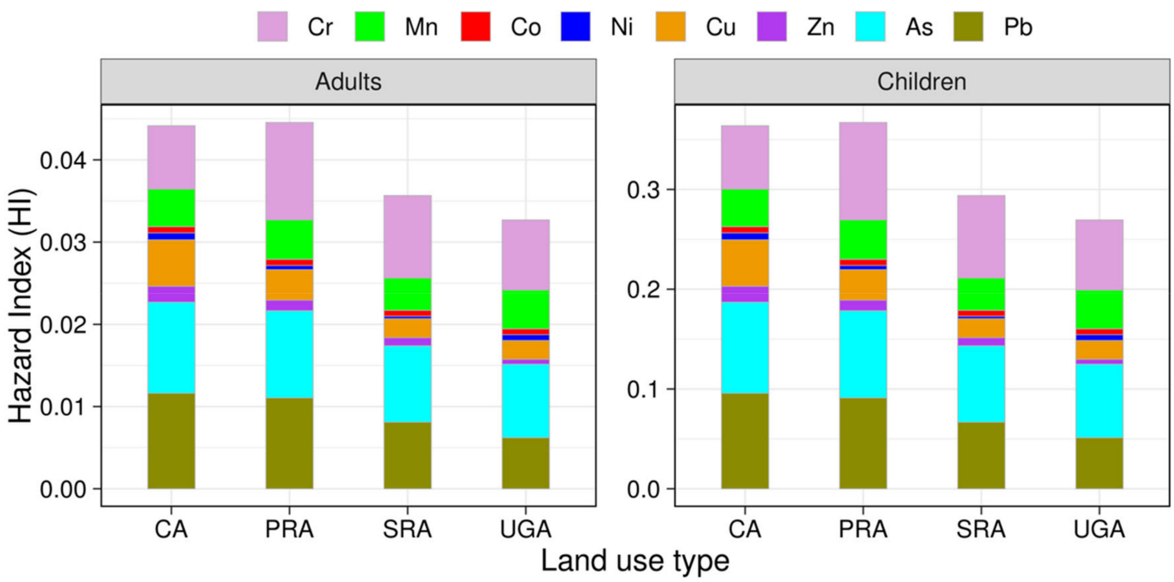

Figure 5. Hazard quotient (HQ) and hazard index (HI) of toxic metals for adults and children in different land-use areas in Dhaka city. CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area.

The results indicate that the HQ and HI values used for the assessment of non-carcinogenic risk were different for each toxic metal; nevertheless, the HQ values for all the studied toxic metals were less than 1, showing no adverse health effects for children and adults. Generally, the HQ values of the toxic metals for children were twelve times higher than for adults in all land-use areas as corroborated in other studies [2,61]. In particular, for children in all the land-use areas, the toxic metals (Cr, Pb, and As) might be a concern for non-carcinogenic health risks due to their higher HQ values compared to those of other toxic metals. However, the total concentrations of As were lower than other toxic metals concentrations (Table 2) and the percentage of bioaccessibility was higher (Figure 3). Therefore, if children are exposed to a certain amount of roadside dust, Cr, Pb, and As could cause potential health risks such as neurological or disability disorders, loss of appetite, respiratory and cardiovascular tract disorders, hematopoietic disorders, weakness, and mental disorders [27,62,63]. In addition, children have a higher digestion capacity, absorption rate, and hemoglobin sensitivity to toxic metals, which are also regarded as health risks in this age group [64]. As a result, when children go outside, they must keep their hands and mouth clean and refrain from "hand eating." Similarly, toxic metals Cr, As, and Pb contributed the most to HQs in adults. The highest contributions of the HQs of Cr (17.5–28.1%), As (23.8–27.3%) and Pb (18.9–26.29%) to HI for adults and children in four land-use areas, respectively. The urban area ranking based on the HI values followed the same pattern for both adults and children (PRA > CA > SRA > UGA). In this study, the maximum HI values for toxic metals in PRA were regarded as reflecting high toxic metal concentrations in the roadside dust (Table 2).

In this study, the carcinogenic risks posed by Cr, Ni, As, and Pb were investigated and the results are presented in Figure 6. On the other hand, the carcinogenic risks of Mn, Co, Cu and Zn were not calculated due to the lack of carcinogenic slope factor values in the ingestion pathway. The carcinogenic risk (CR) caused by As and Pb in roadside dust in various land-use areas of Dhaka city was found to be less than 1×10^{-6}, indicating that there is no obvious serious risk from exposure to toxic metals for adults and children.

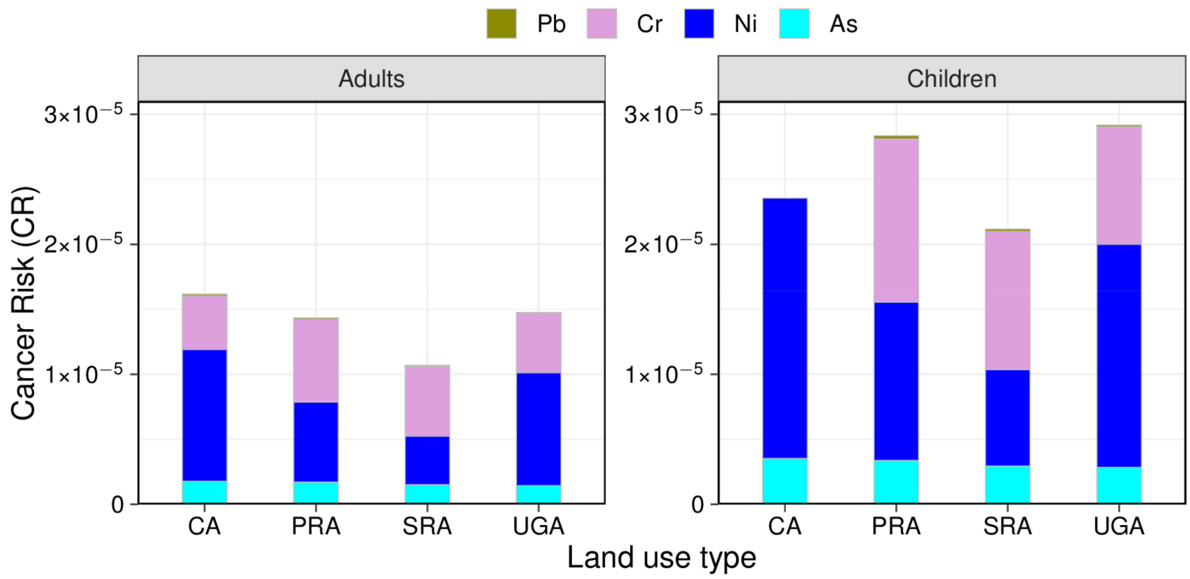

Figure 6. Carcinogenic risk of elements (Pb, Cr, Ni and As) in roadside dust in Dhaka city. CA = commercial area, PRA = planned residential area, SRA = spontaneous residential area, UGA = urban green area.

Previous studies have shown that As might not be a cause of carcinogenic risk in the institutional dust of Dhaka city [15] but it is considered as a cancer risk through inhalation [10]. The carcinogenic risks of Cr in SRA (for children) and Ni in CA (for both adults and children), PRA (for children) and UGA (for children) were found to be within the tolerable range of 10^{-6} to 10^{-4}. In addition, Ni and Cr have been listed as carcinogens by the International Agency for Research on Cancer. Previous studies have noted that Cr might be a cause of the cancer risk of the roadside dust in Dhaka city [10,13], and also that the tannery industries are the main source of Cr in the road dust of Dhaka city [65]. Based on the average CR values of Cr, Ni, As and Pb, the CA (for adults 4.25×10^{-6} and for children 8.42×10^{-6}) showed a higher cancer risk than that of other land-use areas. Concerning the cancer risk for both adults and children, the mean maximum CR levels of Ni were observed in CA (1.01×10^{-5} for adult and 2×10^{-5} for children) and UGA (8.6×10^{-6} for adults and 1.7×10^{-5} for children). On the other hand, the average maximum CR levels of Cr were observed in PRA (6.35×10^{-6} for adults and 1.25×10^{-5} for children) and SRA (5.37×10^{-6} for adults and 1.06×10^{-5} for children) of the roadside dust in Dhaka city. In addition, the average maximum CR levels of Ni (CA and UGA for children and CA for adults) and Cr (PRA and SRA for children) were within the range of tolerable cancer risk (10^{-6} to 10^{-4}), indicating that residents might be at significant risk of cancer if they were continuously exposed to the road dust.

4. Conclusions

This is the first study to investigate the bioaccessible concentrations and assess health risks based on the bioavailability of toxic metals in roadside dust in Dhaka city, Bangladesh. Mn, Zn, and Cu had the highest bioaccessible concentrations in the gastric and intestinal phases of roadside dust for all land-use areas. However, As in the gastric phase and Co in the intestinal phase had the highest bioaccessibility, but with low bioaccessible concentrations in samples from the four land categories. Although the results detected no significant carcinogenic or non-carcinogenic risks, mitigation policies are still suggested in order to reduce the health risks posed by roadside dust imbued with toxic metals in Dhaka city. The actual health risks posed by toxic metals in the vicinity of Dhaka city exceeded the estimates of this study and demand increased attention. Finally, this study suggests that more bioaccessibility research should be conducted based on particle sizes, seasonal variations, and the use of several bioavailability methodologies so as to acquire more realistic and precise results regarding toxic metal bioavailability in the roadside dust of Dhaka city.

Author Contributions: Conceptualization: Q.W. and M.H.R.; methodology: Q.W. and W.W.; software: M.H.K. and M.H.R.; validation: Q.W., W.W. and Y.I.; formal analysis: M.H.K.; investigation, M.H.K.; resources: Q.W., Y.I. and M.H.R.; data curation: M.H.K. and M.H.R.; writing—original draft preparation: M.H.K.; writing—review and editing: M.H.R., Q.W., W.W. and Y.I.; visualization: M.H.K. and M.H.R.; supervision: Q.W. and W.W.; project administration: Q.W.; funding acquisition: Q.W. All authors have read and agreed to the published version of the manuscript.

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