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RESEARCH ARTICLE
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ESTIMATION AND HEALTH RISK ASSESSMENT OF SELECTED HEAVY METALS (Cd, Cr, Pb, Cu and Ni) IN CHILDREN TOYS

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Abstract
The potential risk that heavy metals cause during the early stages of childhood development makes it a global health concern. The main aim of this study is to determine the concentration of cadmium, chromium, nickel-copper, and chromium (VI) to carry out the dose-response assessment, evaluating questionnaire responses given by children's parents in Lahore, Pakistan and calculate hazard quotient (HQ) to evaluate whether concentrations are above or below permissible limit. The concentrations detected by AAS in digested samples ranged from 83.7 mg/kg to -0.087 mg/kg for Cd, 806 mg/kg to -0.05 mg/kg for Cr (VI), 1001 mg/kg to -0.008 mg/kg for Pb, 822 mg/kg to -0.07 mg/kg for Cu, and 3000 mg/kg to -0.9 mg/kg for Ni. 40% samples for Cd, 5% samples for Cr (VI), 55% samples for Pb, and 5% samples for Ni exceeded the EU limit. Based on the HQ values detected, the trend of concentration of heavy metals exceeding EU regulations was Pb > Cd > Cr (VI) > Ni. Copper did not exceed any regulation yet present in many samples. Based on data from questionnaire responses and dose-response assessment using hazard quotient, heavy metal poisoning has been confirmed as a significant hazard because there are several health problems linked with it. Once heavy
metals get into the living organisms, and the organism is exposed many times heavy metals are bioaccumulated. Hence, it can be harmful to mental health and the functioning of the central nervous system.

Keywords- Heavy metal poisoning, Hazard Quotient, bioaccumulation, low-priced toys.

INTRODUCTION:

Determination of toxicity of metals in living organisms is an important issue to consider in the field of agriculture, food, protection of the environment and materials of premium quality (Mustafa Soylak, Sibel Saraçoğlu, Mustafa Tüzen, & Durali Mendil, 2005), (Muwaffak Al Osman, Fei Yang, & Isaac Yaw Massey, 2019). Playing with toys is an essential part of childhood. Different styles of children playing with toys lead to exposure via chewing, licking, sucking or ingestion which results in the leaching of heavy metals through saliva from toys (Kumar, 2007, Tangahu, 2011). This exposure is due to the leaching of heavy metals from the surface of toys (Ahmad, N., 2014). The most crucial scenario occurs when children ingest very small fragments of broken toys or painted coating or as a whole.

In this case, the consumed object gets exposed to the gastrointestinal tract or contacts the saliva in the mouth. A low level of pH with raised temperature and internal salts and enzymes for digestion in the stomach and saliva enhances the solubility of toxic metals in the digestive tract.

Plastic toys contain a wide range of additives to plasticize, stabilize, cure, antioxidize and color the products. The addition of metal stabilizers in toys is essential because they consist of chlorine that forms hydrochloric acid after reaction with free hydrogen ions resulting in degradation of the product if these metal stabilizers are not used in manufacturing. During the developmental and physiological growth of young children i.e., high metabolic rate, low body mass and fast growth rate they are highly sensitive and vulnerable to various health issues either carcinogenic or non-carcinogenic after having contact with items contaminated with various metals (Becker, 2010; Abhay and Prashant, 2007). The bioavailability of metal from an item can be estimated through in vitro and in vivo assessing procedures. Testing via in vivo methods involves experimentation on living bodies: hence, it has limitations due to ethics and extremely high expense. So, preference is given to in vitro i.e., experiments in labs are preferred.

Excessive and long-term exposure to Cadmium (Cd), Chromium (Cr,) Lead (Pb), Copper (Cu), and Nickel (Ni) can be dangerous due to their capability to bio accumulate. They can harm our lungs, kidneys, brain, hepatic system and other organs. Lead is a potential carcinogen, and its exposure has been linked to problems related to intense kidney and brain damage which sooner or later may cause death (WHO, 2018). However, Cadmium and its compounds being carcinogenic in nature can have adverse impacts on the stomach e.g. diarrhea and emesis through ingestion. Even a low level of long-term exposure leads to probable kidney problems, respiratory diseases and weak bones (Saracoglu, S, 2012). Oral exposure to Cr (VI) in humans induces hepatotoxicity, which can develop into early-stage liver cancer and trigger the likelihood of cancer patients (Kara and Frankowski, 2018). Nickel is used in toys because it resists corrosion and has a high conductivity to electricity specifically used in model toys i.e., railway or battery related. (Use of nickel allowed in toys, 2014). According to the International Agency for Research on Cancer (IARC), some nickel compounds are known to cause cancer in humans. Wilson's illness is a rare hereditary disorder that results in prolonged exposure to Cu toxicity in the body, gastrointestinal discomfort, anemia, and damage to the kidney and liver (Brewer 2010).

Many cases in the past have been reported of serious immediate or long-term adverse consequences, including death when children were exposed to harmful chemicals through the ingestion of different consumer items. (Akimzhanova, Z., Guney, M., Kismelyeva, S., Zhakiyenova, A., and Yagofarova, A., 2020) assessed toxic elements in plastic toys in Asian markets of Kazakhstan. Most of the samples had heavy metals more than permissible limits mentioned in the United States, European Union and Canadian standards or legislation. The results were evidence of the widespread issue of heavy metal contamination in children's accessories and toys of underdeveloped countries.

Previously, only the likelihood of harmful contaminations in cheap children's plastic toys was assessed in Karachi. Due to the unavailability of National quality control standards limits for toys in Pakistan or the absence of authority for providing toy quality guidelines to ensure the safety of children, obtained results will be compared with the regulations provided by other countries of the world i.e. United States Consumer Product Safety Commission (U.S.CPSC) toy safety F963-11, European Union (EU) Toy Safety Directive (European Council 2009), Canadian limits

(Government of Canada,2016-2018), and Bureau of Indian Standards (BIS) IS:9873 permissible limits. Hence absence of any published research on the bioaccessibility of heavy metals in children's toys in Pakistan coupled with no national safety standard limits set for toys in Pakistan has necessitated this research project.
The prime research question will be answered, followed by a set of following sub-questions:
- How to evaluate total metal concentrations (Pb, Cd, Cr, Cu and Ni) in low-priced children's toys available in Lahore local markets? Compare results with the European Union, United States, Canada, and Bureau of India’s Toy Safety Regulations.
- Characterize health risk assessment based on AAS analysis of total metal concentration and questionnaire responses.

METHODOLOGY

Sample Collection

A total of twenty plastic toys for children under the age of five were sourced from various local shops in Lahore, Pakistan. The area of the samples collected is shown in Figure 1. Low-cost samples were given preference during the selection process. This was due to earlier studies that claimed that less expensive toys may contain higher quantities of metals due mostly to the recycling of contaminated materials or the absence of raw materials regulation (Kang and Zhu, 2013; Weidenhamer and Clement, 2007a,b). Collected plastic toys featured rattles, playdough or clay, teethers, baby's hard car toys, and soft animal and fruit toys. Most of the selected toys and clays had "Made in China" labels, and the rest of the toys had local production plants.
Cadmium (Cd), Chromium (Cr), Lead (Pb), Copper (Cu) and Nickel (Ni) were examined for their potential health risks in terms of total metal content.

Preparation and Analysis of Samples

With scissors and cutters, plastic toys were broken up into tiny pieces (0.5 cm ×0.5 cm), and paint coating samples and brittle/pliable toys were ground into powder. In porcelain crucibles, 0.5g of each sample was weighed, and then by adding 10 ml of analar-graded concentrated hydrochloric acid, nitric acid, and perchloric acid each sample was heated until fuming stopped. The mixture was diluted to the proper volume in a 50 ml volumetric flask. Following that, it was filtered through the Whatman filter paper. After that, samples were examined using flame atomic absorption spectroscopy for the presence of Ni, Cu, Cd, Pb, and Cr (VI) (FAAS).
These methodologies were used in prior studies (Oyeyiola, 2017, Terry Mohammed, et al., 2020). The sample preparation and analysis of samples were conducted at the Pakistan Council of Scientific & Industrial Research (PCSIR) in Lahore.

The permissible limit of heavy metal concentration

Regulations set by the Indian Bureau for Toy Safety (BIS), Canadian, United States and EU Directive in Table 3.2, 3.3, and 3.4 were compared with the results of samples induced by AAS.

Health Risk Assessment Study

The USEPA was used to adapt the health assessment for carcinogenic and non-carcinogenic risks (2009). Nickel, Chromium Cr (VI), and lead are proven carcinogens, whereas copper is known to be non-carcinogen in human beings (The Risk Assessment Information System., 2007).The United States developed models for assessing health risks (U.S EPA, 1991).
This assessment is carried out to calculate the dose of a toxic substance exposed to a person and the possible health effects in response to that dose.
In the case of heavy metals present in children's toys, the dose is exposed through ingestion because newborns, toddlers, and young children frequently mouth toys. Here risk assessment is carried out using the method described in previous research (Terry Mohammed, et al., 2020).
The average daily dose of ingestion (ADD) was used to assess the non-carcinogenic health risk (Ismail et al., 2017):

$$ADD = \frac{C_{sample} \times IR \times EF \times ED}{BW \times AT} \quad \text{(Equation 1) Here,}$$

ADD = Average Daily Dose of Ingestion (mg/kg/day)
C_{sample}= Heavy metal Concentration in the sample (mg/kg)
IR = Intake/Ingestion Rate (kg/day)
EF = Exposure Frequency (days/year)
ED = Exposure Duration (year)
BW = Body Weight (kg)

AT = Averaging Time (period over which exposure is averaged) (days)

$$HQ = \frac{ADD}{RfD} \quad \text{(Equation 2)}$$

Heavy metal Concentration in the sample was determined in PCSIR in this study, while other factors were set to the default value found in (Grẑetic I. & Ghariani R. H. A., 2008) . The default value for each parameter in the health risk calculation is described in **Table 3.5**.

Exposure Evaluation by filling out Questionnaire survey:
To evaluate the exposure levels of heavy metals after using toys purchased by individuals with low and average incomes pose a health risk factor or not. 50 heads of households were given a survey questionnaire, which 90% of them filled and sent back. The socioeconomic status of the family, including occupation, level of education, and money, as well as the kind and quality of the children's toys, knowledge of the health dangers connected with certain toys, and the prevalence of related health problems, were taken into consideration while developing the questionnaire.

Statistical Analysis
The obtained results from the Atomic Absorption Spectrophotometer were analyzed. Mean concentrations and standard deviations of all samples were obtained and represented through bar charts in Origin Software. This tool was used to get help to easily identify the highest and lowest concentrations of heavy metals in different samples.

RESULTS AND DISCUSSION:
This research focuses on the determination of metal concentration in toys by using an atomic absorption spectrophotometer, comparing the results with the EU, US, Canadian and Indian regulations and the health risk assessment of metals in children's toys checking whether the hazard quotient exceeds 1 or not. There is a lack of national standards for toy safety regulation in Pakistan and the absence of Pakistan standards and quality control authority regulations, specifically on toys to ensure children's safety,
All toys tested, including those shown in their respective tables in the result section, contained health-hazardous elements Cd, Pb, Cu, Ni, and Cr (VI).
Cd, Pb and Ni were found in 40%, 55% and 5% samples exceeding the EU limits despite being classified as carcinogens. Cadmium toxicity in toys and baby products has also been described in other pieces of literature, including (Kumar and Pastore, 2007), (Guney and Zagury 2013). In the literature, almost similar conclusions for Pb have also been reported (Gul, et al., 2022), (M. Hillyer, 2014) and Turner, A. (2019). 35% of the samples exceeded the Cr (VI) regulatory limits of the European Union. It is impossible to assess Cr (VI) oral carcinogenicity. There is no evidence in the existing literature that Cr (VI) is carcinogenic when exposed orally.
Cr (VI) and Ni were also identified at exceeded concentrations, however, compared to Pb and Cd, their quantities were less. In a nutshell, there is a trend of exceeding EU regulations Pb > Cd > Cr (VI) > Ni. Copper was not exceeding any regulation yet present in a considerable amount of samples.

According to the socio-economic questionnaire responses, 54% of parents who filled out the survey had girls and 46% had boys. 32.1% of children were under 1 year old, 28.6% were between 1 and 2 years old, 21.4% were between 3 and 4 years old, and 17.8% were above 4 years old. Most of the questioned homes had two boys and two girls, and about 61% of the children were under the age of four.

The health risk is deemed unacceptable when HQ > 1 and shouldn't be ignored at HQ > 1. In the case of cadmium, almost all the samples of toys had high amounts of cadmium and its HQ value. Cd in Sample C, E, G, I, and J was HQ above 1 i.e. 1.16, 1.04, 1.08, and 1.006 respectively.

Sample C had the highest HQ (2.004mg/L) of cadmium and the least concentration was detected in sample F. While Cr (VI), most samples had a considerable hazard quotient of chromium (VI). Cr (VI) in Sample C, E, G, I, M, P, and Q was HQ above 1 i.e. 1.88, 3.583, 1.026, 0.453, 1.20, 1.257, and 0.5. Lead's HQ in Sample C, E. and M was above 1 i.e. 3.813, 1.56 and 0.8. Most of the Ni samples had negligible hazard quotient. Only sample E had a high HQ value of Ni i.e. 2. Hence, a lot of samples had above 1 HQ values of cadmium, lead, nickel and chromium (VI) indicating adverse health impacts on children. Copper assessment in samples had all the HQ values less than 1 indicating no adverse impact on children. Numerous types of research have been looked into to demonstrate the presence of heavy metals in kid's toys (Gul, et al., 2022), (Terry Mohammed, et al., 2020), (Ismail et al., 2017) and (Korfali, S. I., Sabra, R, Jurdi, M., & Taleb, R. I. , 2013)

CONCLUSION

In short, those plastic toys were collected that were inexpensive, made for young children (infants to 5 years old), and mostly desired by low-income groups. Almost all the toy samples collected from local stores of Lahore had considerable and even high amounts of Cd>Pb>Cr (VI)>Ni>Cu.

Due to the lack of regulation and controls from the regulatory system, low-cost items are readily available in local markets. However, investigations have shown that these contaminated toys are highly dangerous for children's health and may cause serious health issues and damage to their brain, kidney, bones, and nervous systems. Even a small amount in the blood may have such a negative impact on a child's health. Therefore, future research on this topic in children's toys will be beneficial in creating regulations on toy samples and raise awareness among parents regarding health of their children while buying toys.

Conflict of Interest: There was no conflict of interest between the authors.

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Tables

Heavy metals	**EU Regulation**		
	Dry, powder, brittle stuff	Sticky & liquidy stuff	Scrapped toy stuff
Cd	1.3	0.3	17
Cr	37.5	9.4	460
Pb	2	0.5	23
Cu	622.5	156	7700
Ni	75	18.8	930

Table 3.2: Permissible limit of heavy metal concentration set by the European Union (EC 2009), the United States (US CPSC) (ASTM International 2017), Canada (Gov. of Canada 2011, 2018), and BIS (Bureau of Indian Standard).

Heavy metals	**Canada Regulations**		
	Consumer products	**Jewelry**	**Coatings for toy surface**
Cd	130	-	1000
Cr	-	-	-
Pb	90	90	90
Cu	-	-	-
Ni	-	-	-

Table 3.3: Permissible limit of heavy metal concentration set by Canada (Gov. of Canada 2011, 2018).

Heavy metals	**US limits**			**BIS**
	Children products	**Soluble clay for modeling**	**Toy's Coating & substrates**	
Cd	**200 µg**	**50**	**75**	**75**
Cr	**-**	**25**	**60**	**60**
Pb	**100**	**90**	**90**	**90**
Cu		**-**	**-**	**-**
Ni	**-**	**-**	**-**	**-**

Table 3.4: Permissible limit of heavy metal concentration set by United States (US CPSC) (ASTM International 2017) and BIS (Bureau of Indian Standard).

Table-3.5. Input Parameters for Average Daily Dose

Variables in formula used	Values
Ingestion rate (IR) of a kid	0.0002 kg/day (Grẑetic I. & Ghariani R. H. A., 2008)
Exposure frequency (EF)	365 days/ year (USEPA, 2009)
Exposure duration (ED) for non-carcinogenic metals	6 years (Grẑetic I. & Ghariani R. H. A., 2008)
Exposure duration (ED) for non-carcinogenic metals	70 years (USEPA, 2009)
Body weight (BW)	15 kg (Grẑetic I. & Ghariani R. H. A., 2008)
For non-carcinogens: AT (Averaging Time)	Actual ED * 365 days per year and intake is called Chronic Daily Intake = 2190 days
For carcinogens: AT (Averaging Time)	Lifetime (70 years) * 365 days per year and intake is called Lifetime Average Daily Dose (LADD)= 25550 days

Table 3.6: Reference Dose (RfD) for oral consumption associated with non-carcinogenic risks.

Heavy Metals	**RfD** (mg/kg-d)
Cd	0.001 (US EPA, 2010)
Cr (VI)	0.003 (IRIS, 2008)
Pb	0.0035 (WHO, 1993)
Cu	0.04 (USEPA,2016)
Ni	0.02 (IRIS, 2005)

Figures

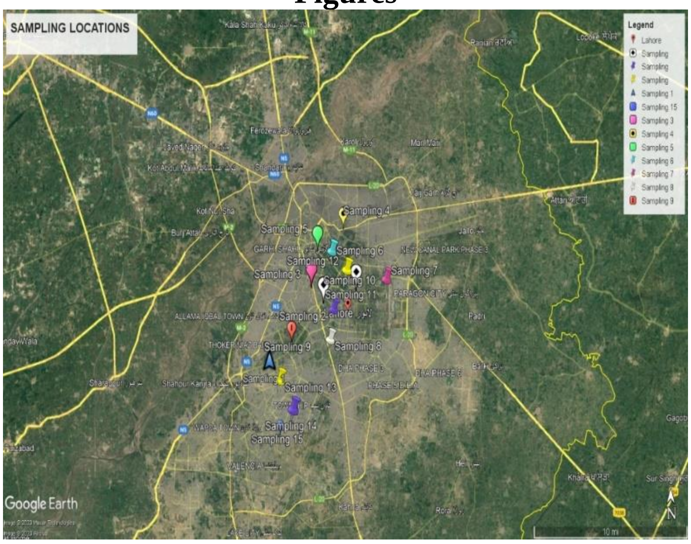


Figure 1 Location of samples collected from various local shops in Lahore, Pakistan.

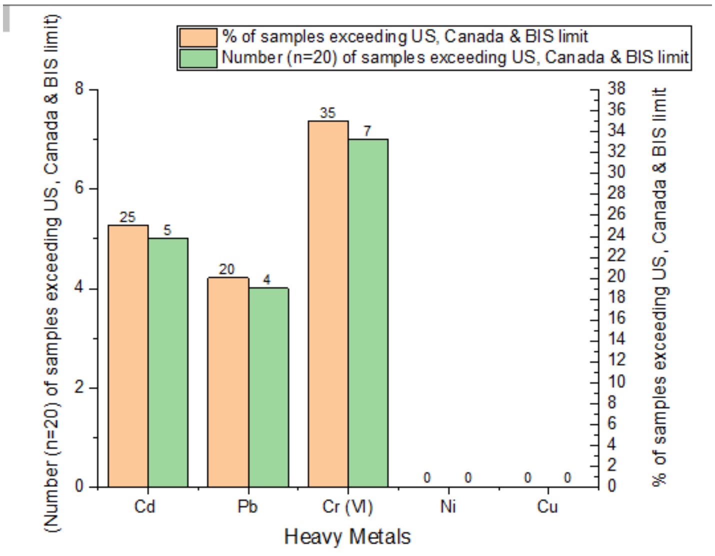
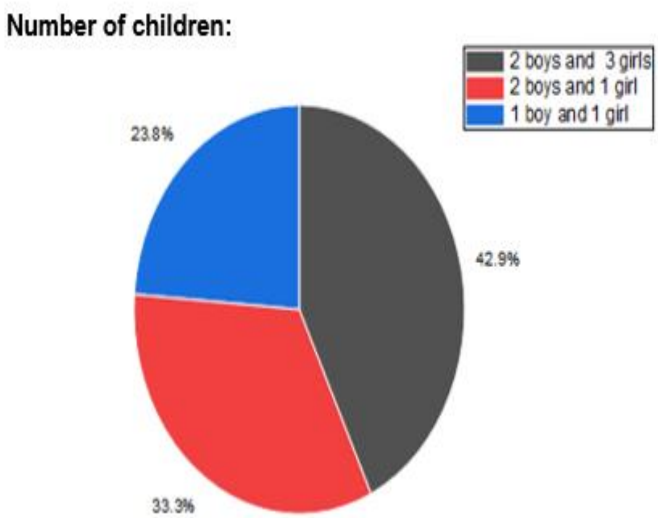
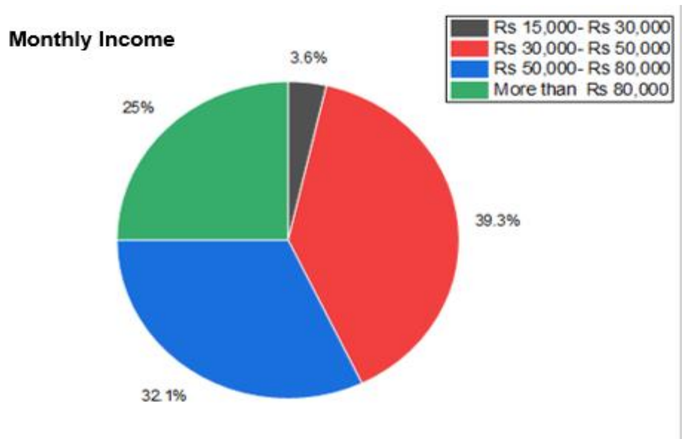


Figure 2. Samples (n=20) exceeding EU, US, Canadian and BIS regulation.





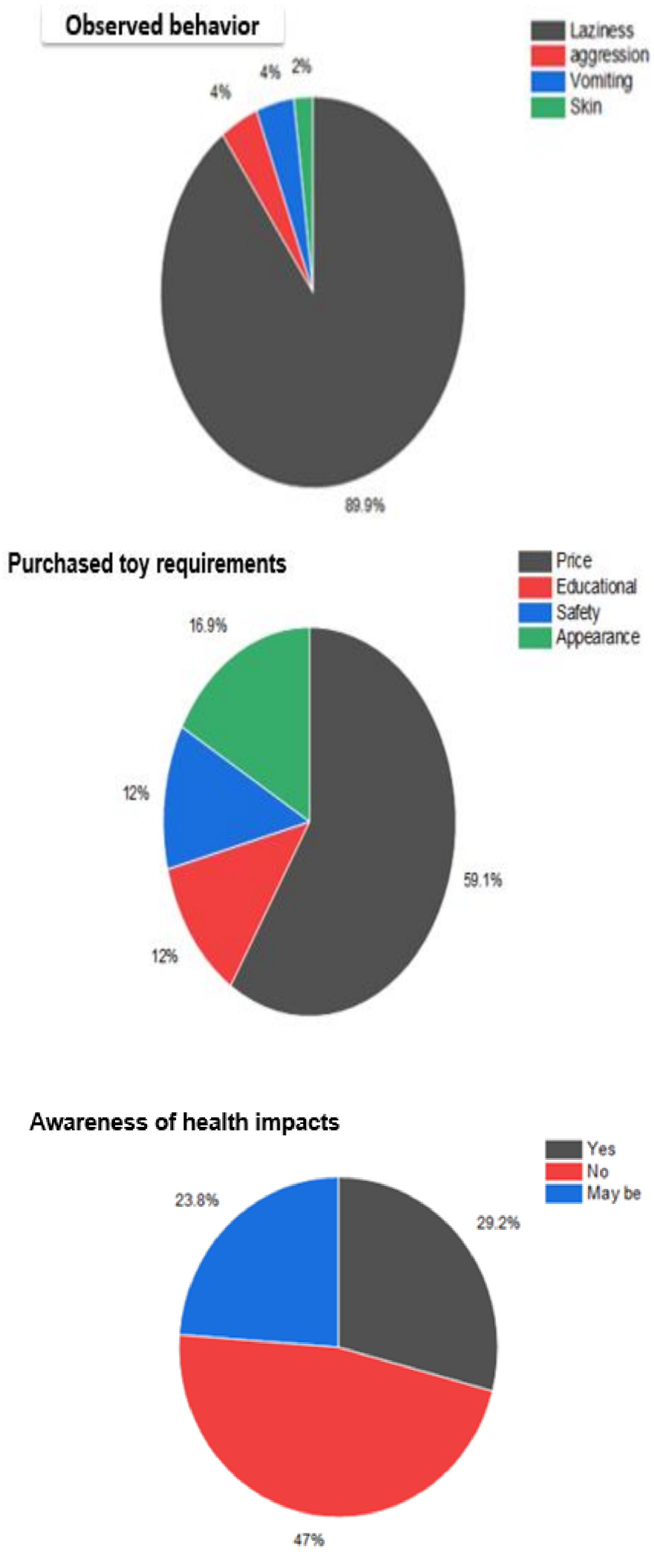


Figure 3. Exposure Evaluation by filling Questionnaire.

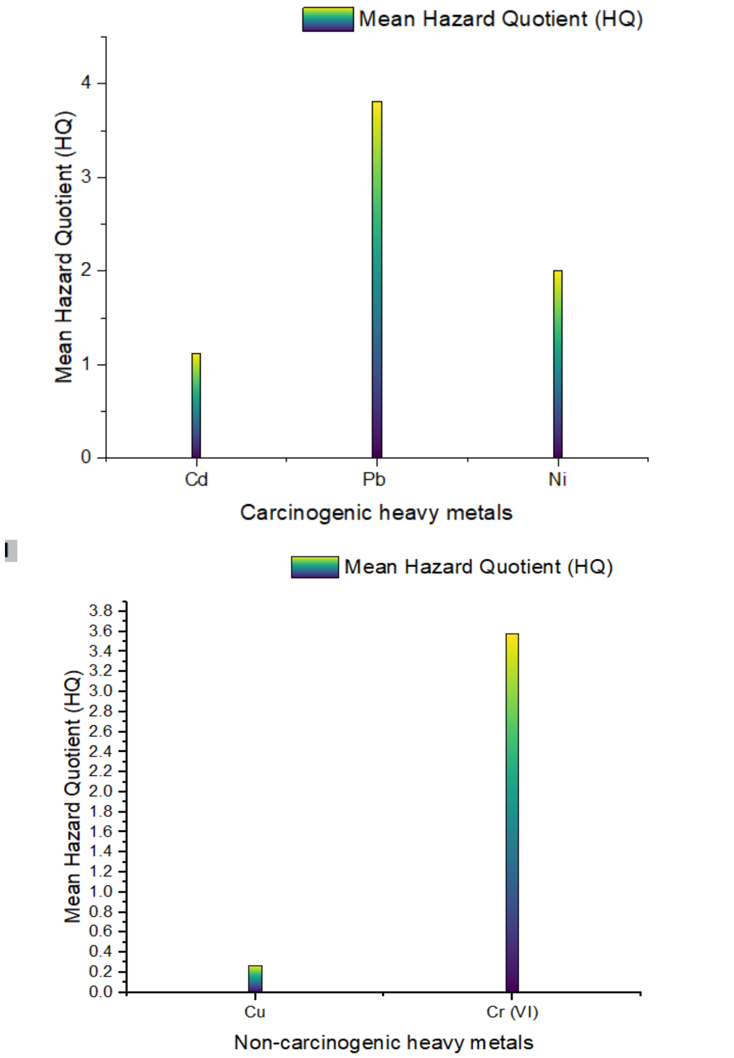


Figure 4. Hazard Quotient of heavy metals