ORIGINAL RESEARCH ARTICLE

Determination of Ni, Cu, Cd, Zn, Pb, Cr and Mn in some black and green tea leaves and their infusions available in Bangladeshi local markets

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ABSTRACT

Tea is a highly common beverage worldwide. In addition to other particles, it contains a range of minerals and trace elements. Even though many are necessary for humans, some could be hazardous. Scientists have long been interested in the toxicity of trace metals and how they affect human health. By using Flame Atomic Absorption Spectrophotometer, the amounts of Ni, Cu, Cd, Zn, Pb, Cr, and Mn in eight black tea leaves, five green tea leaves, and their infusions were examined. The tea samples were collected from Bangladeshi local markets. The infusion time in boiling water was 5 and 10 minutes. Results revealed that the concentration ranges of Ni, Cu, Cd, Zn, Pb, Cr and Mn in black and green tea leaves and their infusions of 5 minutes and 10 minutes were 1.82 to 41.32, 13.54 to 29.87, 0.48 to 2.16, 0.30 to 36.52, 0.04 to 3.89, 0.00 to 4.95 and 112.67 to 187.98 ppm respectively. Concentrations of the elements found in samples were compared with maximum values reported by World Health Organization (WHO). To verify the accuracy of the work, standard reference material (SRM) and certified reference material (CRM) were also tested alongside tea samples. Geographical fluctuations, seasonal changes, chemical characteristics of the soil of the cultivated area, plant age, manufacturing, and packaging process are responsible for the variances in metal content of several tea brands. Cd, Pb, Cr, and Ni's target hazard quotients (THQ) and the hazard index (HI) from drinking tea were both below one, indicating no risk to human health.

Keywords: Black Teas; Green Teas; Metal Concentrations; Tea Infusions; Flame Atomic Absorption Spectrophotometer

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1. Introduction

One of the oldest, most consumed, and non-alcoholic beverages in the world is tea (Camellia sinensis L), which is produced from the leaves of tea plants^[1]. Teas have proven economic, health, and social benefits. Tea comes in a variety of flavors, including black, green, lemon, ginger, and honey. The most often consumed varieties among them are black and green teas^[2].

Though tea infusion is a very poor supply of protein, vitamins, and carbs, it is a good source of dietary metals and polyphenols that bind to metals to reduce free radicals^[3]. Green tea has more potent antioxidants than other types of tea, including vitamin E and catechins. It is thought that tea catechins can interact with reactive oxygen species, which may be important in the development of cancer, by stopping a chain of oxidative reactions, thereby reducing, or even preventing the development of cancers like lung, prostate, and breast cancer^[4–7]. Due to their powerful antioxidant activity, catechins not only strengthen the immune system but also effectively kill germs, restrict plaque and cavity growth,

and prevent an excessive buildup of blood cholesterol^[8–11].

Tea leaves also contain essential minerals at major, minor, and trace levels, including zinc, manganese, iron, copper, magnesium, titanium, aluminum, strontium, bromine, sodium, potassium, phosphorous, iodine, and fluorine^[12]. However, excessive consumption can lead to a number of health issues^[13]. The optimal concentration required for multipurpose varies greatly depending on the element and the age and gender of the consumers^[14]. The study of essential and heavy metals in tea plays important roles on the human body in a complex metabolic pathway, and they should be consumed in adequate amounts, as deficiencies or excesses may cause diseases^[15]. Both metallic and non-metallic components are necessary for the human body's healthy growth, development, and optimal operation. It is therefore crucial to determine the presence of these elements in beverages, water, food, plants, and soil^[16–18].

Nowadays, measuring the total metal content of tea is far more popular than measuring the metal content of infusions. The metals that have been determined vary between investigations since some concentrate on determining many metals while others just look for harmful components. Iraq worked on the concentrations of Cd, Pb, and Ni in several brands of tea leaves and tea infusion offered in the marketplaces in Babylon Province in 2022^[19].

According to another paper published in Iran, 160 samples of bagged, and loose-leaf black and green tea were tested in 2020 using a graphite furnace atomic absorption spectrophotometer (GF-AAS) to determine the concentration and health risk of hazardous metals like Cu, Zn, As, Cd, and Pb^[20]. This article analyzed the essential and hazardous components of tea leaves and the beverages made from them. The amount of metal varies from one country to the next due to several variables, including environmental standards, geographic characteristics, and human engagement in those places. Therefore, toxic and micronutrient levels in processed tea leaves and their infusions are therefore extensively monitored by numerous researchers and evaluated the health risks associated with their consumption globally. In Bangladesh, heavy metals in food and drinks

are frequently monitored; however, very little study was carried on tea samples. The goal of this study is to update information and find out how much Ni, Cd, Pb, Cr, Cu, Zn, and Mn are contained in the eight black and five green tea leaves and their infusions of 5 minutes and 10 minutes. Secondly, the health risk assessment was conducted by consuming these teas. Finally, the measured values were compared to the standard reference materials, tomato leaves NBS-SRM 1573 and tea leaves NIES-CRM 7, and to WHO recommendations for drinking water.

2. Materials and methods

2.1 Sample collection and preparation

A total of 13 commercially available teas (8 black and 5 green teas) were taken from the local shops of Bangladesh. They were chosen in this study because all were very common and were well known in Bangladesh. Glassware used were washed with detergent solution followed by 10% (v/v) HNO₃ and then rinsed with tap water and finally with deionized water and dried by an oven (Memmert 30 °C–200 °C) before use. Distilled water was also used as the solvent to prepare the solutions. Each sample was weighed by a Sartorius Analytical Balance (CP 225 D). The diluted digests were analyzed using Flame Atomic Adsorption Spectrophotometer (FAAS) supplied by Shimadzu (AAS-6800).

2.2 Reagents

All reagents used in this study were of analytical grade. H₂O₂ was procured from Sigma Aldrich (USA), HNO₃ and HClO₄ were supplied by Merck, Germany. The standard solution of Ni, Cu, Cd, Zn, Pb, Cr, and Mn was purchased from Wako Pure Chemical Industries Ltd.

2.3 Material preparation

The essential and heavy metals were analyzed in eight black teas and five green teas available in Bangladeshi local markets which were popular and widely consumed today. In this study, two methods were employed to prepare tea samples in order to find out the actual amount of mineral components and trace metals in tea and tea extracts by FAAS. For each method, three replicates for each sample were

completed. This digestion procedure was validated by using the standard reference materials of tomato leaves (NBS-SRM 1573) and the reference certified material of tea leaves (NIES-CRM 7).

2.3.1 Total contents of metals

One gram (1 g) of each tea sample was digested using 12 cm³ of a mixture (3:1 v/v) of concentrated HNO₃ and HClO₄ acids. The digested samples were filtered and transferred to a 100 mL volumetric flask, and the volume was adjusted to the mark with 5% HNO₃ acid. Tomato leaves NBS-SRM 1573 of standard reference material and tea leaves NIES-CRM 7 of certified reference material were also digested by the same technique.

2.3.2 Hot water extraction of tea samples

Infusions for metal analysis were determined by having a portion of 1 gm of tea boiled in 50 mL of distilled water for 5 and 10 minutes respectively and filtered. The filtrate samples were evaporated to near dryness and digested with concentrated HNO $_3$ and HClO $_4$ (9 + 3) mL. The final volume of the solution was made up to 100 mL. Following digestion 10 drops of H $_2$ O $_2$ were added and centrifuged. The acid digested sediments were filtered and capped until measured by FAAS.

2.4 Human health risks

Identifying the levels of heavy metals (Cd, pb, Cr, and Ni) in commonly used tea in open markets in Bangladesh was the aim of this study, which also aimed to evaluate the situation surrounding the assessment of health concerns. We utilized the following equation to determine the average daily intake (ADI) of heavy metals (g·person⁻¹·day⁻¹) of Cd, Pb, Cr and Ni by the human participants through consumption of black and green teas^[21].

$$ADI = (C \times IR \times TR)/(BW \times 1,000)$$

where.

C stands for metal concentration in tea leave;

IR stands for tea ingestion rate (11.4 g·person⁻¹·day⁻¹);

TR is the metal transfer rate from tea leaves to tea infusion;

TR = Metal in tea infusion/metal in tea leaves. Extraction efficiency (EE) can also be calculated as a percentage of TR.

BW is bodyweight, 60 kg for adults' perspective to Bangladesh^[22,23].

To discover non-carcinogenic health problems in tea drinkers, the Target Hazard Quotient (THQ) of each metal was calculated. There is no considerable risk of non-carcinogenic consequences for consumers if the THQ value is less than one. The following calculation calculates the single contaminant's hazard quotient:

THQ = Exposure dose/reference dose where, reference dose = 5.0×10^{-4} for Pb, 1.5×10^{-3} for Cd, 3×10^{-3} for Cr and 2×10^{-2} mg·kg⁻¹·bw·d⁻¹ for Ni respectively^[24–26].

In order to determine the overall danger for a combination of heavy metals, the hazard index of the heavy metals is computed using the equation below. Target Hazard Quotient (THQ) values are determined for each metal in a tea sample, and the summation hazard of heavy metals is utilized to extract the Hazard Index (HI) value.

$$HI = THO(Cd) + THO(Pb) + THO(Cr) + THO(Ni)$$

2.5 Method validation

Using the tomato leaves NBS-SRM 1573 of standard reference material and tea leaves NIES-CRM 7 of certified reference material, the accuracy of the total element concentrations was evaluated.

3. Results and discussion

3.1 Metal concentrations in tea leaves and tea infusions

Every sample of this study was very popular in local market of Bangladesh. Eight black teas and five green teas are each represented in **Table 1** with information using code number, such as A and B. With comparisons of tea leaves, tea infusions, and boiling times, **Table 2** and **Table 3** provide an overview of the concentrations of hazardous (Ni, Cd, Pb, and Cr) and essential (Cu, Zn, and Mn) elements in black and green teas that were found in this study. All the metals were present at $\mu g \cdot g^{-1}$ level in tea leaves and $\mu g \cdot m L^{-1}$. **Table 4** shows the comparison of our results with WHO standard value of drinking water to measure its quality.

Table 1. Tea contents according to manufacturers' information

Sample No.	Sample ID	Origin	Shape	Quantity				
Black teas	Black teas							
1	Black A	Bangladesh	Packet	10 gm				
2	Black B	Bangladesh	Packet	100 gm				
3	Black C	Bangladesh	Packet	100 gm				
4	Black D	Bangladesh	Packet	50 gm				
5	Black E	Bangladesh	Packet	100 gm				
6	Black F	Bangladesh	Packet	100 gm				
7	Black G	Bangladesh	Packet	100 gm				
8	Black H	Bangladesh	Packet	100 gm				
Green teas								
9	Green A	Bangladesh	Tea bag	40 tea bags with 60 gm				
10	Green B	Bangladesh	Tea bag	50 tea bags with 100 gm				
11	Green C	Bangladesh	Tea bag	100 tea bags with 140 gm				
12	Green D	Bangladesh	Packet	100 gm				
13	Green E	Bangladesh	Packet	100 gm				

Table 2. Comparison of tea leaves and their infusions for 5 minutes and 10 minutes boiling

Average	Ni	Cu	Cd	Zn	Pb	Cr	Mn
Tea leaves	27.66 ± 0.56	22.06 ± 0.48	1.04 ± 0.18	17.47 ± 0.79	1.69 ± 0.45	4.23 ± 0.11	149.18 ± 4.16
5 min	21.57 ± 0.27	21.53 ± 0.64	0.55 ± 0.44	14.47 ± 0.46	0.45 ± 0.13	0.15 ± 0.24	143.80 ± 3.67
10 min	20.90 ± 0.14	18.34 ± 0.41	0.70 ± 0.47	12.22 ± 0.28	1.02 ± 0.29	0.78 ± 0.38	145.27 ± 3.72

Values are given mean \pm SD and units are $\mu g \cdot g^{-1}$ for tea leaves and $\mu g \cdot m L^{-1}$ for tea infusions.

Table 3. Comparison of black tea and green tea

Average	Ni	Cu	Cd	Zn	Pb	Cr	Mn	
Black tea leaves	29.84 ± 0.28	21.16 ± 0.12	1.06 ± 0.49	21.58 ± 0.68	1.39 ± 0.27	4.10 ± 0.54	152.23 ± 3.51	
Green tea leaves	21.14 ± 0.84	22.06 ± 0.41	1.95 ± 0.24	5.13 ± 0.28	3.61 ± 0.29	0.32 ± 0.19	130.04 ± 3.49	
Boiling for 5 min	Boiling for 5 minutes							
Black tea	26.57 ± 0.46	20.75 ± 0.57	0.55 ± 0.67	18.41 ± 0.49	0.39 ± 0.27	3.78 ± 0.49	145.81 ± 2.57	
Green tea	6.56 ± 0.86	23.87 ± 0.67	0.52 ± 0.22	2.65 ± 0.64	1.08 ± 0.29	0.30 ± 0.46	137.80 ± 2.64	
Boiling for 10 minutes								
Black tea	24.06 ± 0.29	16.72 ± 0.57	0.72 ± 0.15	15.21 ± 0.27	0.96 ± 0.54	4.45 ± 0.59	148.52 ± 3.10	
Green tea	11.40 ± 0.51	23.19 ± 0.63	0.76 ± 0.72	3.26 ± 0.42	1.38 ± 0.43	0.50 ± 0.52	135.53 ± 3.24	

Values are given mean \pm SD and units are $\mu g \cdot g^{-1}$ for tea leaves and $\mu g \cdot m L^{-1}$ for tea infusions.

Table 4. Comparison of metal contents in both tea leaves and tea infusions with WHO standard value of drinking water (ppm)

Elements	Min	Max	Mean	SD	WHO
Ni	1.82	41.32	23.38	0.14	0.07
Cu	13.54	29.87	18.64	0.54	2.00
Cd	0.48	1.16	0.70	0.13	0.003
Zn	0.30	36.52	14.72	0.18	5
Pb	0.04	3.89	3.55	0.72	0.01
Cr	0.00	4.95	3.09	0.16	0.05
Mn	112.67	187.98	142.75	4.89	-

SD = Standard Deviation

A zinc inadequacy can result in stunted growth and decreased bone development. At greater concentrations, it can harm humans, especially if zinc oxide fumes are inhaled. Ailment can occasionally develop from eating acidic food prepared in Zn galvanized containers^[27]. Evidence found that the concentration of this element in tea samples was 9.1 to 15.1 µg·g⁻¹ with a mean concentration of $12.2 \pm 2.2 \, \mu g \cdot g^{-1}$ in Meghalaya, India^[28]. The Zn content in tea samples in our study was $17.47 \pm 0.79 \ \mu g \cdot g^{-1}$ which is very similar to that investigation, while in tea aqueous extracts of 5 and 10 minutes it was 14.47 ± 0.46 $\mu g \cdot mL^{-1}$ and $12.22 \pm 0.28 \,\mu g \cdot mL^{-1}$ respectively. The Zn contents in tea samples ranged from 23.47 mg·kg⁻¹ to 52.32 mg·kg⁻¹ with an average value of 39.55 mg·kg⁻¹ were reported in Vietnam^[29]. In black tea, it was $21.58 \pm 1.68 \ \mu g \cdot g^{-1}$ in our analysis, whereas in the same content of green tea it was 5.13 $\pm 0.28 \,\mu g \cdot g^{-1}$ which was lower than the black tea.

Cd is a highly poisonous metal with no beneficial effects on plants or animals. It can be accumulated in the kidneys and lead to renal problems, high blood pressure, and harm to the nervous system^[30]. The concentration of Cd found in tea samples ranged from 0.48-1.16 µg·g⁻¹ with an average of $0.70 \,\mu g \cdot g^{-1}$. The lowest value of cadmium was found in Black G which was 5-minute boiled infusion and the highest was also found from Black G tea leaves might be due to the less solubility of Cd in tea extract. The average value of Cd in tea leaves, 5-minute and 10-minute boiled infusions were $1.04 \pm 0.18 \, \mu g \cdot g^{-1}$, $0.55 \pm 0.44 \ \mu g \cdot mL^{-1}$, and $0.70 \pm 0.47 \ \mu g \cdot mL^{-1}$ respectively. Cd concentration of black tea samples and their 5-minute and 10-minute boiled infusions were $1.06 \pm 0.49 \ \mu g \cdot g^{-1}$, $0.55 \pm 0.67 \ \mu g \cdot mL^{-1}$, and $0.72 \pm 0.15 \ \mu g \cdot mL^{-1}$ respectively, whereas for green tea samples the concentration of Cd were very similar: $1.95 \pm 0.24 \ \mu g \cdot g^{-1}, \, 0.52 \pm 0.22 \ \mu g \cdot mL^{-1}$ and 0.76 \pm 0.72 µg·mL⁻¹ respectively (**Tables 2** and **3**). In a previous study, heavy metal content of black teas from Tamil Nadu, India was measured, and the concentration of cadmium was found to be 0.89 ± 0.10 $\mu g \cdot g^{-1}$ to 1.59 \pm 0.26 $\mu g \cdot g^{-1}$ which was very similar with our study^[31].

Cu is a very useful element, but too much of it in the body can lead to "Wilson illness", which can cause lung issues, acute anemia, nausea, and vomiting. A lack of this element can cause cardiovascular illness, whilst an excess can cause hypertension^[32]. Additionally, copper is known to work as an antioxidant enzyme. In 2021, Cu was detected in green teas from Kakara district of Taraba, Nigeria and the concentration of Cu was found to be $0.06-0.20 \,\mu g \cdot g^{-1[33]}$. In our study, the range of Cu was found to be 13.54 to 29.87 ppm with an average value of 18.64 ± 0.48 ppm which was higher than that found in Taraba, Nigeria but similar to that found in China (14.90-26.10 μg·g⁻¹)^[34]. In the case of black and green teas, the average values were very near, which were $21.16 \pm$ $0.12 \,\mu g \cdot g^{-1}$ and $22.06 \pm 0.41 \,\mu g \cdot g^{-1}$ respectively. And the average values of their 5-minute boiled infusion were found to be $20.75 \pm 0.57 \,\mu \text{g} \cdot \text{mL}^{-1}$ (black teas) and $23.87 \pm 0.67 \,\mu \text{g} \cdot \text{mL}^{-1}$ (green teas) and that of 10minute boiled infusion were $16.72 \pm 0.59 \ \mu g \cdot mL^{-1}$ (black teas) and 23.19 \pm 0.63 μ g·mL⁻¹ (green teas). The permissible limit of Cu for drinking water is 2 ppm recommended by WHO, in case of some samples, it exceeded in this study.

Even though it may only be present in very little amounts, Pb is extremely hazardous. Several organ systems are impacted by PB. Similar to Cd, this element is biologically unnecessary and has a substantial harmful threat^[31]. The mean Pb content in black tea was $1.39 \pm 0.27 \,\mu\text{g}\cdot\text{g}^{-1}$ whereas in green tea it was found higher at 3.61 \pm 0.29 μ g·g⁻¹, which is lower than Chinese tea (where Pb levels ranged from 0.48 $\mu g {\cdot} g^{-1}$ to 10.57 $\mu g {\cdot} g^{-1}$ and average was 3.04 μg·g⁻¹)^[35]. But this value was higher than that from the survey in Nigeria in 2010, in which Pb was reported in black teas $(0.03 \pm 0.00 \,\mu\text{g}\cdot\text{g}^{-1})$ as well as its 10-minute boiled infusion $(0.00 \pm 0.00 \,\mu \text{g·mL}^{-1})$ where the same content in green tea was higher 0.27 $\pm 0.02 \ \mu g \cdot g^{-1}$ and $0.00 \ \mu g \cdot m L^{-1}$ in its infusion^[36]. In our study, Pb content in 5-minute and 10-minute infusions were found to be $0.39 \pm 0.27 \,\mu \text{g} \cdot \text{mL}^{-1}$ and $0.96 \pm 0.54~\mu g{\cdot}mL^{-1}$ for black teas and 1.08 ± 0.29 $\mu g \cdot m L^{-1}$ and $1.38 \pm 0.43 \ \mu g \cdot m L^{-1}$ for green teas respectively where the total content of Pb in tea leaves for both black and green teas was $1.69 \pm 0.45 \,\mu g \cdot g^{-1}$.

Airborne exposure Ni fumes and dust can harm the respiratory system and lead to lung cancer. Nickel is a renowned human carcinogen, and the most prevalent antioxidant in the human body— α tocopherol—is affected in terms of its action^[37]. In a previous study, the Ni content in black and green teas from Ibadan, Nigeria was 3.50–8.00 µg·g^{-1[38]}. In our study, it ranged from 1.82 ± 0.55 ppm to 41.32 ± 0.30 ppm for both black and green teas. Ni in the infusion samples ranged between 2.89 µg·mL⁻¹ and 22.6 μg·mL⁻¹ reported by other researchers in India^[28]. In our experiment, it was $21.57 \pm 0.27 \,\mu g \cdot mL^{-1}$ and $20.90 \pm 0.14 \,\mu \text{g} \cdot \text{mL}^{-1}$ for boiled infusions lasting 5 and 10 minutes respectively. Moreover, nickel content in black tea $(29.84 \pm 0.28 \,\mu\mathrm{g}\cdot\mathrm{g}^{-1})$ was slightly higher than that in green tea (21.14 \pm 0.84 μ g·g⁻¹). However, it is a subject of concern that in some samples Ni content exceeded WHO standard limit (0.07 μg·g⁻¹) for both black teas and green teas. Ni is mainly obtained by applying poor-quality fertilizers and micronutrients to the soil and leaves^[39]. Ni is a hazardous element; hence tea fields must be checked for heavy metal impurities.

Cr lowers fatty acids and cholesterol and controls blood sugar and insulin levels, but long-term exposure to high chromium levels in humans leads to lung cancer^[30]. In earlier investigations in Puan County, Guizhou Province, China, the range of Cr concentration in green tea samples was found to be 121 μg·g⁻¹ and in Iran in black tea samples it was found to be $0.9-3.9 \,\mu \,\mathrm{g} \cdot \mathrm{g}^{-1}$ whereas in infusion of 5.0 g of black tea in 150 mL water it was 0.4-0.8 μg·mL^{-1[34,37]}. In our study, the Cr content in tea samples was $3.09 \pm 0.16 \ \mu g \cdot g^{-1}$ whereas in tea aqueous extracts of 5 minutes and 10 minutes it was 3.78 \pm $0.49~\mu g \cdot mL^{-1}$ and $4.45 \pm 0.59~\mu g \cdot mL^{-1}$ for black teas whereas $0.30 \pm 0.46 \ \mu g \cdot mL^{-1}$ and 0.50 ± 0.52 µg⋅mL⁻¹ for green teas. The Cr content in black tea was $4.10 \pm 0.54 \,\mu g \cdot g^{-1}$ which was greater than the same content in green tea $0.32 \pm 0.19 \,\mu\text{g}\cdot\text{g}^{-1}$, which may be due to the manufacturing process. It might be present in both hexavalent and trivalent forms in teas. In some tea samples, it was above the permissible limit referred by WHO showed in Table 4.

Mn is necessary for both human and animal skeletal development, growth, and reproduction. However, it is unclear how this component works in humans^[17]. In India, an investigation showed Mn content in black tea was $709.0 \pm 14.18 \ \mu g \cdot g^{-1}$ and in

green tea was $508 \pm 44.034 \, \mu g \cdot g^{-1}$ and in Puan County, Guizhou Province, China, the concentration of Mn in tea leaves was 194-4,610 µg·g⁻¹ which were less in our study 152.23 \pm 3.51 $\mu g \cdot g^{-1}$ and $130.04 \pm 3.49 \ \mu g \cdot g^{-1}$ for black tea and green tea respectively^[28,34]. For infusion, it was 145.81 ± 2.57 $\mu g \cdot mL^{-1}$ and $148.52 \pm 3.10 \ \mu g \cdot mL^{-1}$ for black tea, and $137.80 \pm 2.64 \ \mu g \cdot mL^{-1}$ and 135.53 ± 3.24 µg⋅mL⁻¹ for green tea with boiling period of 5 and 10 minutes respectively. Like other investigations, our study revealed that the concentration of Mn was the highest among other available elements in tea. The reason for this phenomenon is that Mn is easily accumulated in tea leaves. The migration rate of Mn is found to be higher in acidic soil, especially when pH is below 4.5. In addition, tea plants have a significant translocation factor with Mn, which allows the mobility of Mn from the roots to the ground above parts and leaves tissue under the transpiration stream^[40]. Also, the roles of genes whose expression is responsible for Mn's dispersion, absorption, translocation, and transformation in tea plants can be used to explain the high potential of Mn to accumulate in old tea leaves^[41].

In a review, the micronutrients, Mn and Zn were measured to be $0.05 \ \mu g \cdot L^{-1} - 1,980.34 \ mg \cdot L^{-1}$ and 1.12–2.23 µg·L⁻¹ respectively in tea infusions^[42]. In our investigation, the levels of metallic components in tea samples were in the following order: Mn > Ni > Cu > Zn > Cr > Pb > Cd. The obtained results demonstrate that the tested brands of both black and green teas have significantly different amounts of heavy metals. In black teas, Ni, Zn, Mn, and Cr were higher than that in green teas; Cu and Cd were very similar in both types of teas; but Pb showed higher value in green teas compared to black teas. The outcome demonstrates that hazardous metals Cd, Pb, and Cr were less soluble in tea aqueous extract while Ni, Cu, Zn, and Mn were more soluble.

In many countries, the heavy metal contents exceeded the permissible limit of WHO. In Iraq, Pb in tea leaves and their infusions was higher than the level of parameters recommended by WHO. The contents of toxic elements Cd, Pb, and Ni in tea leaves in that country were 0.18–2.43 ppm, 0.3–

17.24 ppm, and 3.7–9.3 ppm whereas in tea infusions the contents were 0.022–0.4 ppm, 0.4–7.25 ppm, and 0.008–4.4 ppm respectively^[19]. Pb and Cd in tea contents in Iraq were also higher than that in our study. On the other hand, Ni and Cd in the present study were higher than the Kingdom of Saudi Arabia whereas Pb was lower. The metal contents of fourteen common marked brand tea leaves (both black and herb teas) in Saudi Arabia were 2.46–8.90 $\mu g \cdot g^{-1}$ for Ni, 0.03–14.84 $\mu g \cdot g^{-1}$ for Pb and N.D–0.37 $\mu g \cdot g^{-1}$ for Cd. However, Pb and Cd were too low to be detected in their infusions

where Ni was $0.55-5.30 \,\mu\text{g}\cdot\text{g}^{-1[43]}$.

3.2 Method validation

The accuracy of the total element concentrations was assessed using the tomato leaves NBS-SRM 1573 and tea leaves NIES-CRM 7, and the results are shown in **Table 5**. This exhibits very little differences among metals. The recoveries for tomato leaves NBS-SRM 1573 and tea leaves NIES-CRM 7 were also examined, and the results were compatible with those of prior studies that had SRM Apple Leaves 1515 as their standard reference^[44].

Table 5. Results obtained for the standard reference materials with certified value ($\mu g \cdot g^{-1}$)

SRM/CRM	Element	Certified value	Measured value	Recovery (%)
Tomato leaves	Ni	4.70 ± 0.14	4.32 ± 0.43	91.91
NBS-SRM 1573	Cu	4.70 ± 0.14	7.47 ± 0.74	158.98
	Cd	1.52 ± 0.04	1.88 ± 0.45	123.68
	Zn	30.9 ± 0.7	28.43 ± 0.50	92.01
	Cr	1.99 ± 0.06	1.36 ± 0.57	68.34
	Mn	246 ± 8	320.83 ± 4.21	130.08
Tea leaves	Ni	7.89 ± 0.52	9.87 ± 0.48	125.09
NIES-CRM 7	Cu	9.48 ± 0.76	11.87 ± 0.67	125.21
	Zn	31.9 ± 2.2	27.32 ± 0.46	85.64
	Mn	704 ± 52	621.32 ± 4.08	88.25

3.3 Human health risks

As tea is prepared, not all of the heavy metals found in tea leaves are entirely transported into the tea infusion. The metal transfer rate (TR) from tea leaves to hot infusions should therefore be taken into account. As a result, TR values were also used to calculate the anticipated daily consumption of each metal. A high TR value was found in the examined tea samples, which were ranked as Mn > Cu > Ni > Zn > Cd > Pb > Cr for a 10-minute infusion time and Cu > Mn > Zn > Ni > Cd > Pb > Cr for a 5-minute infusion period. When the infusion period was increased from 5 to 10 minutes, the TR values of Mn and Ni increased by 1.04% and 2.56%, respectively. Due to the fact that Mn, Zn, and Cu are necessary for both people and tea plants, the health risk associated with these elements was not considered. The average daily intake (ADI) of Cd, Pb, Cr, and Ni components was lower than the reference dosage, indicating that adults who routinely consume heavy

metals in tea will not have any harmful health effects.

Pb > Ni > Cd > Cr for a 5-minute infusion period and Ni > Pb > Cr > Cd for a 10-minute infusion period, respectively, were the mean THQs of each metal related with ingesting tea infusions in more detail represented in Tables 6 and 7 respectively. Each metal's THQs were lower than its RfD values. Though the HI values for the 5- and 10-minute infusions were both less than one, more emphasis should be given to the health risks associated with tea infusions because Bangladesh has seen relatively little research in this area. According to this study's findings, drinking tea with heavy metals (Cd, Pb, Cr, and Ni) does not pose any dangers to human health. This finding is consistent with earlier research in China where the THQ for the elements Cd, Pb, Tl, Hg, As, Sb, Cr, and Ni and the hazard index (HI) from tea consumption were less than one^[40].

Table 6. ADI, EE, THQ, and HI of heavy metals of 5-minute boiled infusions

5-minute infusion	ADI	EE (%)	THQ
Ni	4.10E-03	77.98	2.05E-01
Cd	1.05E-04	52.90	6.97E-02
Pb	8.55E-05	26.80	1.71E-01
Cr	2.85E-05	35.50	9.50E-03
HI			4.55E-01

Table 7. ADI, EE, THQ, and HI of heavy metals of 10-minute boiled infusions

10-minute infusion	ADI	EE (%)	THQ
Ni	3.97E-03	75.60	1.99E-01
Cd	1.33E-04	67.30	8.87E-02
Pd	1.94E-04	60.40	3.88E-01
Cr	1.48E-04	18.40	4.94E-02
HI			7.24E-01

Researchers in Bangladesh measured the concentrations of Cd, Cr, Pb, As, and Se in fresh tea leaves, processed (black) tea leaves, and soil from tea plantations. Processed (black) tea leaves included the following amounts of heavy metals: Cd (0.04–0.16 μ g/g), Cr (0.45–10.73 μ g/g), Pb (0.07–1.03 μ g/g), however health risk assessment was not done^[45]. In our investigation, the ranges for Cd, Cr, and Pb were 0.48–1.16, 0.00–4.95 and 0.04–3.89 ppm respectively.

The result of the metal concentrations was compared with WHO guidelines for drinking water, bearing in mind that the tea is consumed significantly in less amount compared to drinking water in Bangladesh. Some elements were over the WHO standard limit, while others were within the permissible limit showed in Table 4. Although there hasn't been much research on tea samples in Bangladesh, heavy metal levels in fruits, vegetables, and other food products are often determined. HI values of some of the studied samples were less than one and some showed greater than one. In a prior study, non-cancer risk (HI > 1) was shown to be present in all the tested vegetables and fruits examined in Bangladesh, with a contribution of 19, 27, 49, and 5% for fruit vegetables, root vegetables, leafy vegetables, and fruits, respectively^[46].

4. Conclusion

For thousands of years, tea has been a significant beverage and a fundamental part of world culture. Numerous studies have demonstrated the positive health benefits of tea consumption. This study used AAS to measure the amounts of Cu, Zn, Cr, Mn, Ni, Pb, and Cd in black teas, green teas and their infusions that were readily available in the Bangladeshi local market. The obtained results were compared with the guidelines of drinking-water quality, WHO. Ni, Cd, Pb and Cr are very toxic, and in some tea samples they exceeded the maximum permissible limit according to WHO. The result also revealed that, the total content of Cu, Cd and Pb was the highest in green teas, while that of Mn, Cr, Ni and Zn was the highest in black teas. Moreover, Cd, Pb, and Cr were less soluble in tea aqueous extract while Ni, Cu, Zn, and Mn were more soluble. Metals from tea leaves to tea infusions were tested for the transfer rate (TR). In order to assess the health risk to consumers and determine the non-carcinogenic dangers, the target hazard quotient (THQ) and hazard index (HI) of the investigated heavy metal consumption in the tea were calculated. Each metal had a value of less than 1, which is considered safe for human ingestion. Many factors including environmental and anthropogenic activities are contributing to the metal's accumulation in tea leaves. As a result, this investigation will give us a better idea in taking further steps to improve the quality of black and green teas in the state. Toxic metal levels in all food must be measured and monitored as part of the quality control and food safety procedures in Bangladesh.

Conflict of interest

The authors declared no conflict of interest.

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