Investigation of levels and human health risk of heavy metals in tea samples marketed in Jazan, Saudi Arabia

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Abstract: Following water, tea is the most widely used beverage. Tea is regarded as a healthful beverage because of its relationship with a decreased risk of stroke, metabolic syndrome, obesity, and events related to cardiovascular disease. Tea contains a variety of trace minerals and elements that are good for human health, but if they are present in high enough amounts, some of them can be hazardous and cause cancer. This work stands out due to its thorough evaluation of heavy metal levels, and conducting a risk assessment for tea consumers in Jazan region southern of Saudi Arabia. The present research examined the content of 6 different heavy metals in 11 samples of black tea gathered from local markets in Jazan, southern Saudi Arabia: cadmium (Cd), lead (Pb), chromium (Cr), zinc (Zn), copper (Cu), iron (Fe), and zinc (Zn).

Flame atomic absorption spectroscopy (FAAS) was employed to detect heavy metals in tea samples. The Target Hazard Quotient (THQ) and combined hazard index (HI) for heavy metals in tea samples were employed for assessing the health risks to tea consumers. The study found that the median content of Cd, Pb, Cr, Zn, Cu, and Fe in the investigated samples is 1.31, 2.33, 16.07, 27.88, 5.52, and 120.4 µg/g, respectively. We compared the concentration of heavy metals in the samples under examination to the World Health Organization's (WHO) maximum allowable levels. The findings demonstrated that the mean levels of Cu, Fe, Pb, Cd, and Zn in tea samples were significantly below the maximum permitted values. Conversely, Cr level in tea samples exceeded the maximum allowable values. THQ mean values were 0.003, 0.05, 0.16, 0.19, 0.69, and 0.8 for Cr, Fe, Zn, Pb, Cu, and Cd, respectively. The THQ values of all metals under investigation were found to be below the permitted limits (less than one). These data show that the individual heavy metals present in the investigated samples do not pose any carcinogenic health concerns. In contrast, the HI value for various heavy metals was determined to be greater than one (1.91) indicating that regular tracking of the levels of heavy metals in black tea samples is required.

Keywords: Evaluation; Heavy metals levels; Tea; Non-carcinogenic health risk

دراسة مستويات ومخاطر المعادن الثقيلة على صحة الإنسان في عينات الشاي المسوقة في جازان، السعودية

الملخص: بعد الماء، يعد الشاي هو المشروب الأكثر استخدامًا. يعتبر الشاي مشروبًا صحيًا بسبب علاقته بانخفاض خطر الإصابة بالسكتة الدماغية ومتلازمة التمثيل الغذائي والسمنة والأحداث المرتبطة بأمراض القلب والأوعية الدموية. يحتوي الشاي على مجموعة متنوعة من المعادن النادرة والعناصر المفيدة لصحة الإنسان، ولكن إذا كانت موجودة بكميات عالية بما فيه الكفاية، فقد يكون بعضها خطيرًا ويسبب السرطان. يبرز هذا العمل بسبب تقييمه الشامل لمستويات المعادن الثقيلة، وإجراء تقييم المخاطر لمستهلكي الشاي في منطقة جازان جنوب المملكة العربية السعودية. تناول البحث الحالي محتوى 6 معادن ثقيلة مختلفة في 11 عينة من الشاي الأسود التي تم جمعها من الأسواق المحلية في منطقة جازان، جنوب المملكة العربية السعودية: الكادميوم (Cd)، الرصاص (Pb)، الكروم (Cr)، الزنك (Zn)، النحاس (النحاس)، والحديد (Fe)، والزنك (Zn).

تم استخدام مطيافية الامتصاص الذري باللهب (FAAS) للكشف عن المعادن الثقيلة في عينات الشاي. تم استخدام حاصل الخطر المستهدف (THQ) ومؤشر الخطر المشترك (HI) للمعادن الثقيلة في عينات الشاي لتقييم المخاطر الصحية على مستهلكي الشاي. وجدت الدراسة أن متوسط محتوى الكادميوم والرصاص والكروم والزنك والنحاس والحديد في العينات التي تم فحصها هو 1.31، 2.33، 16.07، 27.88، 5.52، و120.4 ميكروجرام/جرام، على التوالي. قمنا بمقارنة تركيز المعادن الثقيلة في العينات قيد الفحص بالمستويات القصوى المسموح بها من قبل منظمة الصحة العالمية. وأظهرت النتائج أن متوسط مستويات النحاس والحديد والرصاص والكادميوم والزنك في عينات الشاي كانت أقل بكثير من القيم القصوى المسموح بها. وعلى العكس من ذلك، تجاوز مستوى الكروم في عينات الشاي الحد الأقصى للقيم المسموح بها. كانت القيم المتوسطة لـ THQ هي 0.003 و0.05 و0.16 و0.19 و0.69 و0.8 لكل من Cr وFe وZn وPb وCu وCd على التوالي. تبين أن قيم THQ لجميع المعادن قيد التحقيق أقل من الحدود المسموح بها (أقل من واحد). تظهر هذه البيانات أن المعادن الثقيلة الفردية الموجودة في العينات التي تم فحصها لا تشكل أي مخاوف صحية مسرطنة. في المقابل، تم تحديد قيمة HI لمختلف المعادن الثقيلة لتكون أكبر من واحد (1.91) مما يشير إلى أن التتبع المنتظم لمستويات المعادن الثقيلة في عينات الشاي الأسود مطلوب.

1. Introduction

Drinking tea is almost as common as drinking water throughout the world. Every day, almost 18 billion teacups are drunk worldwide [1-3]. Arabian adults usually have three or four cups of tea a day. Tea can be categorized as black, white, green, oolong, dark, or yellow tea depending on the method of fermentation and the mix of flavor characteristics [4]. The most common forms of tea are green and black types [5]. Black tea is created by fermentation but green tea is created by drying and roasting. Numerous studies have demonstrated the link between reasonable tea drinking and reduced blood cholesterol, the prevention of LDL oxidation, Skin cancer, Parkinson's disorder, infarction of the heart, heart attack, and stroke, and a decreased risk of cardiovascular and cancer diseases [6-8]. This is so because a variety of chemicals, such as heavy metals, polyphenols, caffeine, and fluorides, are beneficial to the human body [9]. Several studies have demonstrated that the tea plant is contaminated with a variety of dangerous heavy metals, such as nickel, lead, cadmium, mercury, chromium, and others which affect the tea quality [10-15]. There are several reasons why heavy metals naturally accumulate into tea leaves, including contaminated soil, the use of pesticides and fertilizers, the environment in which the tea is grown, mining, agricultural runoff, contaminated irrigation water, the provenance of the tea, and the manufacturing processes [16-20]. Some heavy metals are harmful and carcinogenic, however acceptable quantities of Cr, Fe, Co, Ni, and Zn are necessary for organism growth and may be used as a treatment for a variety of diseases [21-24]. Chromium, for instance, reduces fat and cholesterol and regulates insulin and blood sugar levels; however, prolonged contact with elevated chromium concentrations causes lung cancer [25]. Moreover, lead and cadmium pose a serious risk to human health, especially to the neurological urinary systems, which can lead to troubles with the kidneys, hearts, bones, and nervous systems [26]. All of these minerals can be released by tea leaves into the infusion. [17]. One key biological feature of heavy metals is their potential to bioaccumulate. This makes bioaccumulation an essential component of hazard evaluation methods. Thus, excessive consumption of heavy metals can lead to a number of illnesses, including weakening nails, oily skin, pigmentation, skin discoloration, hyperactivity in autistic children, and hair loss, particularly in women. They may potentially result in toxicity, hyposmia, coma, or even death [27-29]. For these reasons, determining the concentration of heavy metals in tea is vital to human health and ought to be done regularly. Several studies were conducted to determine the hazardous metal content in various samples of tea leaves. Many investigations have been conducted in Asia to evaluate the content of hazardous elements in tea samples. Wu, Xiaoling, et al. [30] examined the health problems associated with tea drinking in China and the presence of heavy metals in tea leaves. An additional study [31] found that 30 distinct Chinese teas had Pb contents ranging from 0.26 to 3.2 mg kg^{-1}, and Cd contents ranging from 0.0059 to 0.085 mg kg^{-1}. Another study [32] discovered that various heavy metal concentrations (Al, Pb, Cd, Hg, and As) were near the upper levels published by (WHO).[33]. Pourramezani et al.[23] concluded that drinking tea posed no danger of heavy metal exposure because the hazard index of Indian and Sri Lanka teas was non-significant for tea consumers.

Moreover, Pb and Cd contents in tea brands in Saudi Arabia ranging from 0.3–2.2 to 0.32–2.17 µg g^{-1}, correspondingly, according to Ashraf and Mian [5].According to WHO-acceptable standards, these levels are safe for ingestion by humans.

Analytical techniques such as anodic stripping voltammetry (ASV), graphite furnace atomic absorption spectrometry (GFAAS), and inductively coupled plasma-mass spectrometry (ICP-MS) are employed for assessing the concentration of heavy metals [34-36] but these instruments are expensive. However, Flame Atomic Absorption Spectrometry (FAAS) is more widely available and reasonably priced. This work stands out due to its thorough evaluation of both individual and multiple heavy metal levels, comparing them with international standards and conducting a risk assessment for tea consumers in Jazan region southern of Saudi Arabia. The goal of this research aimed to measure the heavy metals content, particularly Cd, Pb, Cr, Zn, Cu, and Fe, in 11 samples of black tea using FAAS. The study examined the potential health risks associated with heavy metals in Saudi tea consumers, utilizing the HQ and HI to calculate human health hazards.

2. Materials and methods

2.1. Reagents and materials

Standard solutions and tea samples were prepared using ultrapure deionized water generated by a Milli-Q purification system. Tea sample preparation for analysis was done using HPLC-grade nitric acid, which was purchased from Sigma-Aldrich. Typical stock solutions of Zinc, Cu, Fe, Pb, Cd, and Cr were purchased from Analytik Jena in Germany at a concentration of 1000 mg/L in 0.5% (v/v) HNO_3. Every day, a specific volume of stock solution was dissolved in MilliQ water to prepare fresh working standards solutions within the appropriate concentration range for each element. All of the glassware used in the standard solution preparation was thoroughly cleaned with MilliQ water, allowed to air dry, and then immersed in 10% nitric acid for an entire night before being used.

2.2. Apparatus

The content of heavy metals in tea samples was determined using FAAS novAA 350, Analytik Jena, Germany. Table 1 shows the measuring conditions for each heavy metal.

Table 1. Operating conditions for measuring each heavy metal with an atomic absorption spectrophotometer.

	Cu	Fe	Pb	Cd	Zn	Cr
λ (nm)	324.8	248.3	283	228	213.9	357.9
Slit (nm)	1.4	0.2	1.4	1.4	0.5	0.2
Lamp current (mA)	2	4	2	2	2	4
Flame	Air-C_2H_2					

2.3. Tea samples, collection, preparation, and analysis

Eleven of the most popular tea samples were randomly selected from supermarkets in the Saudi Arabian southern city of Jazan. Samples were prepared for chemical analysis using the standard protocol outlined in the Association of Official Agriculture Chemists [37]. The samples were easily dried at 100°C, then ignited at 450°C for 10 hours till white ashes formed. After adding five milliliters of 6M HCl, the sample was allowed to evaporate until completely dried. The remaining substance was dissolved with 0.1 M nitric acid. The same protocol was applied to blanks. FAAS was used to assess the diluted digested samples. First, use the device software to choose the desired elements for analysis as well as a suitable wavelength for the selected element. Calibrate the instrument by inserting a blank and standards, creating a standard curve, and then analyzing the unknown samples.

2.4. Characterization of the health risks

An important metric for evaluating the risks to long-term health is the metals daily intake determination which can be calculated using the equation (1) in Table S1[38]. On the other hand, the prospective risk implications of different metals can be statistically assessed via the target hazard quotient. equation (2) in Table S1 is used to compute the target hazard quotient [39, 40]. In this study, the total risk potential of non-carcinogenic effects of being exposed to a combination of heavy metals was evaluated using (HI) using equation (3) in Table S1 [13, 38]. The cumulative non-carcinogenic health risk of several heavy metalsis determined using Equation 3 [41]. It was considered that the non-carcinogenic risk would be tolerable if the THQ value was less than one. Greater THQ levels correspond to more risk [42]. On the other hand, if the HI is less than one, there is no risk of cancer from the exposure dose. Heavy metal exposure doses that are harmful to human health are highly likely to occur if the HI is greater than one. When the HI value exceeds 10.0, there is a prolonged harmful impact on human health [43].

3. Results and discussion

Stock solutions of 1000 mg/L for each of the selected heavy metals were diluted using 0.5% (v/v) HNO_3 solution to create a series of standard solutions for establishing the calibration curves for the metals. Figure 1 illustrates the obtained calibration curves for the heavy metals under investigation. The correlation coefficient (R^2) was used to assess the calibration curves' linearity. The calibration curves for all heavy metals under consideration were found to have good linearity within their concentration range.

3.1. Heavy metal concentrations in tea samples

Figure 2 indicates the concentrations of the heavy metals Cu, Fe, Zn, Pb, Cd, and Cr in the tea samples under investigation. The ability of this plant to accumulate Tea's capacity for gathering metals is demonstrated by the overall levels of metals evaluated in tea samples.

Based on the average metal content in tea samples, the metals can be ranked as follows: Fe > Zn > Cr > Cu > Pb > Cd. Cu levels in the samples investigated varied from 1.9 to 14.5 µg/g, with a mean of 5.52 ug/g. Our findings are consistent with the average Cu concentration (5.5 µg/g) obtained by Kilicel F.[44].

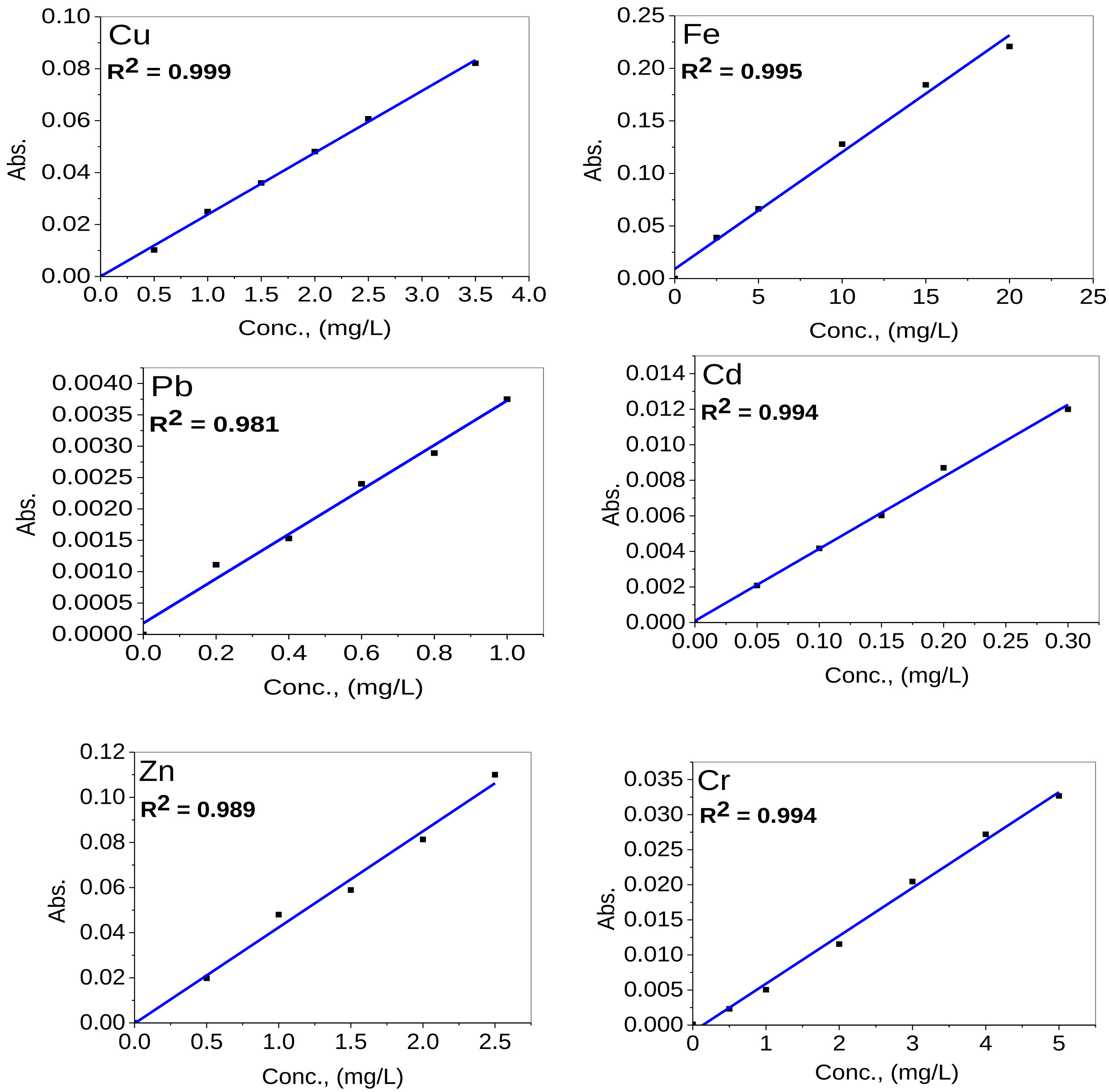

Figure 1: Standard curves for Cu, Fe, Pb, Cd, Zn, and Cr.

The average concentration of Cu in tea samples in the current study was lower than the upper limits established by the World Health Organization (10 µg/g) [43, 44] and several other countries, such as China (60 µg/g), Japan (100 ug/g), and the United States (150 µg/g) [44].Our findings showed good agreement with values reported in the literature for tea samples from Turkey, China, Japan, India, and the United States. [3, 19, 34, 36, 45] Furthermore, the level of Cu identified in the tea samples under investigation agrees with the results of Abdallah A. Shaltout et al. [46] who stated that samples taken from Saudi Arabia had Cu levels ranging from 7.5 to 28.3 µg/g. The wide range in Cu concentration seen in the teas may be explained by the various types and quality levels, Cu pollution comes from fungicides and growing regions of the teas [47].

The first element detected at the greatest level in the samples of black tea that were being examined was iron. The iron content ranged widely between 53.5 to 223.9 µg/g in the various tea samples, with an average of 120.4 µg/g. All tea samples had an acceptable level of Fe, even if it was below the allowable threshold of 450 µg/g. [48]. The Fe level in the samples under investigation was found to be consistent with the findings of Al-Othman et al. [49] who found that the levels of Fe in the examined samples were 46 and 348 µg/g, respectively. On the other hand, Waqar Ashraf and Atiq A. Mian [5] have observed that the mean value of Fe was 250.5 µg/g, with levels ranging from 88.7–946.7 µg/g. Additionally, Fe levels in the investigated samples varied from 74 and 1,000 µg/g, according to Pedro et al.[2]

Zinc concentrations fluctuated between 6.7 and 46.8 µg/g, averaging 27.88 µg/g. When the zinc content is below the 50 µg/g allowable limit, all tea samples are considered acceptable.[48, 50]. Our findings matched with those of Federico et al. [49], who found that samples of black tea had an average Zn concentration of 29 µg/g. [51]. POPOVIĆ, Slađana, et al. obtained similar results, determining that the Zn level varied between 19.06 and 43.97µg/g.[52]. Furthermore, Kilicel F. [44] discovered that samples of black tea had an average Zn concentration of 31.99 µg/g.

Concentrations of Pb were 0.01 to 12.5 µg/g, averaging 2.33 µg/g. This is consistent with Al-Oud, S. [53], who observed that Pb levels in Saudi Arabia tea samples varied between 0.03 and 14.84 µg/g. According to a different study, the mean Pb content is 3.04 µg/g [54]. Except for the T11 sample, all samples had acceptable Pb levels that were below the WHO-permitted standard of 10 µg/g [33] as well as other countries including China (5 µg/g), India (10 µg/g), and Thailand (10 µg/g). This rise could be attributable to commercial flavor and color additives containing different amounts of various heavy metals. [55]. Previous investigations have shown that Pb levels ranged from 0.03 to 14.84 µg/g, 0.198 to 6.345 µg/g, and 0.26 to 0.83 µg/g [18, 53, 56]. The primary sources of Lead in tea are the growing mediums, such as soil. Pb contamination in soil is frequently related to industrial operations, agricultural activities (application of pesticides), and urban activities (the burning of gasoline). Pb is more accessible for tea root intake in extremely acidic soils. Another way that Pb can contaminate tea is through deposits from the contaminated air into the plant's leaves [47].

Cadmium levels in black tea samples varied between 0.92 and 3.98 ug/g, having a mean value of 1.31 µg/g. This was considered acceptable since the amount was below the WHO-recommended threshold of 3 µg/g. [33]. All of our findings on Cd in samples other than T3 are consistent with those of Alwan W. [55] who found that Cd concentrations ranged from 0.18 to 2.43 µg/g. Furthermore, our results for Cd in all samples except T3 are consistent with those of POPOVIĆ, Slađana, et al.[52] who discovered that the range of Cd concentrations was 0.077 to 0.92 µg/g. Franklin et al. [57] She stated that phosphatic fertilizers had 4.9–5.5 ug/g of Cd and zinc sources contained 11.8–50.9 ug/g. It seems that the sources of the lead in tea came from phosphatic and zinc fertilizers.

Chromium (Cr) is extremely harmful to human health, causing severe damage to the kidneys and lungs. All of the black tea samples had chromium contents ranging from 5.6 to 40.2 µg/g, with an average of 16.07 µg/g. Every tea sample had an unacceptable level of Cr, defined as one where the level exceeds the allowable limit of 1.3 µg/g. [48]. The results we got were consistent with those of Falahi, E. [58] who reported a mean concentration of Cr of 15.9 µg/g. The nine commercial teas that were gathered from different countries had a Cr level that ranged from 19.8 to 129.1 µg/g [17]. According to Ferrara et al., black tea samples had chromium levels between 17.9 to 115.4 µg/g, which is in line with the Cr level found in our investigation [59]. On the other hand, Mandiwana et al. [60] discovered that the chromium concentration in samples of black tea varied from 0.28 to 14.0 $\mu g\ g^{-1}$. Additionally, maximum Cr was discovered in Turkish tea samples by Narin et al. [36] at 16.9 ug/g. Cr is mostly introduced into the black tea manufacturing process through the CTC rollers, where it is regarded as a local contaminant [56]. According to Seenivasan et al., the sharpening of crush, tear, and curl rollers employed in the production of tea was mostly associated with the Cr content of tea. [17]. Moreover, it has been noted that Cr speciation in soil determines whether plants are contaminated with Cr [61]. The variation in Cr concentration across tea from different production sites may help to explain this in part.

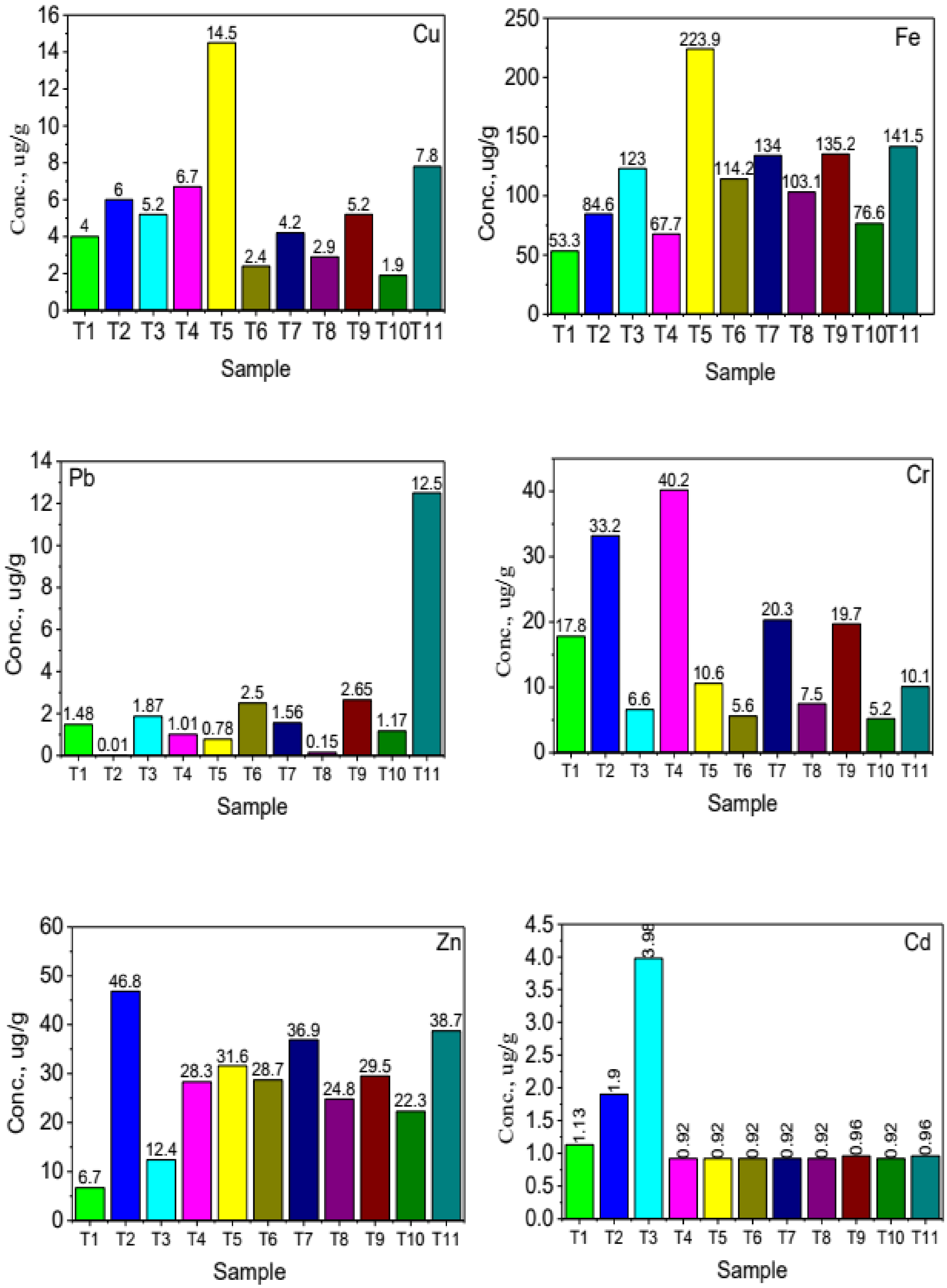

Figure 2. Concentration of Cu, Fe, Pb, Cd, Zn, and Cr in tea samples (µg/g)

3.2. Assessment of health risk

3.2.1. Singular heavy metal risk

The target hazard quotient (THQ) values for each heavy metal in each sample under investigation are estimated to evaluate non-cancer health risks. The target hazard quotient (THQ) evaluates the possibility of being exposed to a trace element with known negative impacts on humans [62]. Figure 3 depicts the THQ values for every heavy metal in each sample and the mean THQ values. The order of the mean THQ values after consuming prepared tea was Cd > Cu > Pb > Zn > Fe > Cr. The THQ values for each heavy metal in all samples under investigation ranged from 0.002 to 0.051 averaging 0.003, 0.02 to 0.1 averaging 0.05, 0.04 to 0.27 averaging 0.16, 0.001 to 1.05 averaging 0.19, 0.16 to 1.17 averaging 0.69 and 0.56 to 2.42 averaging 0.8 in case of Cr, Fe, Zn, Pb, Cu, and Cd, respectively. For all heavy metals in all of the samples under evaluation, THQ mean values were found to be less than 1 (THQ < 1). However, the THQ values were 1.1 and 2.42 in the T_2 copper sample and T_3 cadmium sample. The THQ values for Cu, Cd, Fe, and Zn in tea samples were less than 1, which is consistent with the findings of Gogoi b. et al [63]. Similar results showed that drinking tea does not expose people to heavy metal hazards [13, 64]. Additionally, comparable findings were made by S. Wali Alwan [55], who showed that the THQ values for Pb and Cd were less than 1 Moreover, THQ values for Cd and Cr were less than 1, according to Sahar G. et al [65]. According to these results, it is unlikely that consuming tea leaves regularly would hurt a Consumer's health.

3.2.2 Accumulated hazard of several heavy metals

The HI values for different metals can be used to express their accumulated non-carcinogenic effects. Figure 4a indicates the hazard index scores of all metals that were examined through the consumption of tea samples. Figure 4b shows the mean THQ values for the investigated heavy metals and their corresponding HI value. It was indicated that the HI values in both cases were greater than 1. As shown in Figure 4a, HI was ranged from 1.04 in T_1 to 3.02 in T_3 samples. Our findings concurred with those of previous studies [13, 20, 66] which revealed that the HI value was greater than 1. Moreover, S. Wali Alwan [55] reported comparable results, determining that the HI values for Cd, Pb, and Ni in tea samples varied from 0.34 to 23.4. Additionally, HI values for Cu, Zn, Ni, Cr, Cd, Pb, As, Hg, and Al in tea samples ranged from 0.16 to 1.7, according to another investigation [13]. These findings suggest that there are non-carcinogenic health risks linked with the multiple heavy metals found in the examined samples.

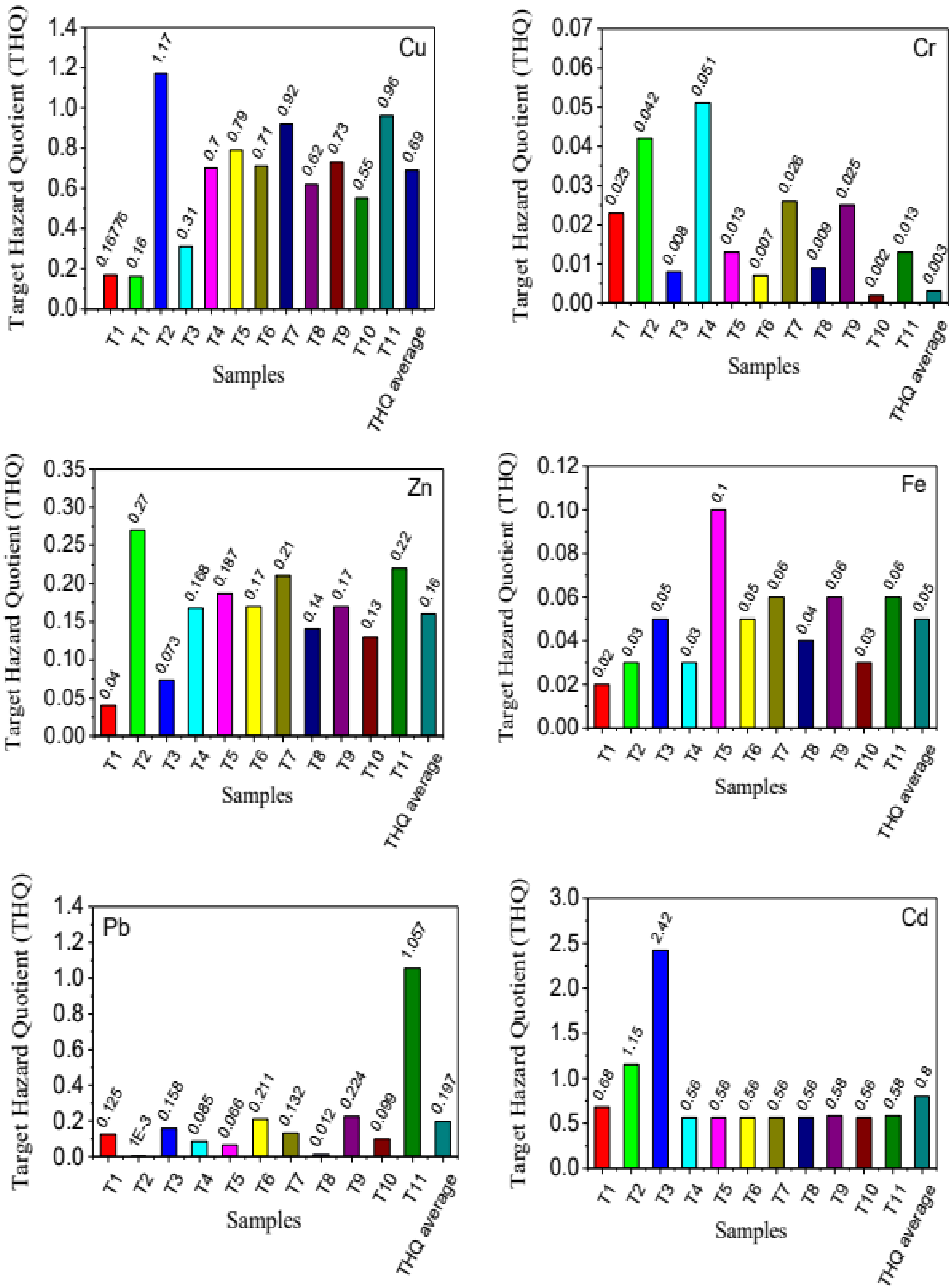

Figure 3. Target Hazard Quotient and its average for the selected heavy metals; Cu, Fe, Pb, Cd, Zn, and Cr in the examined samp

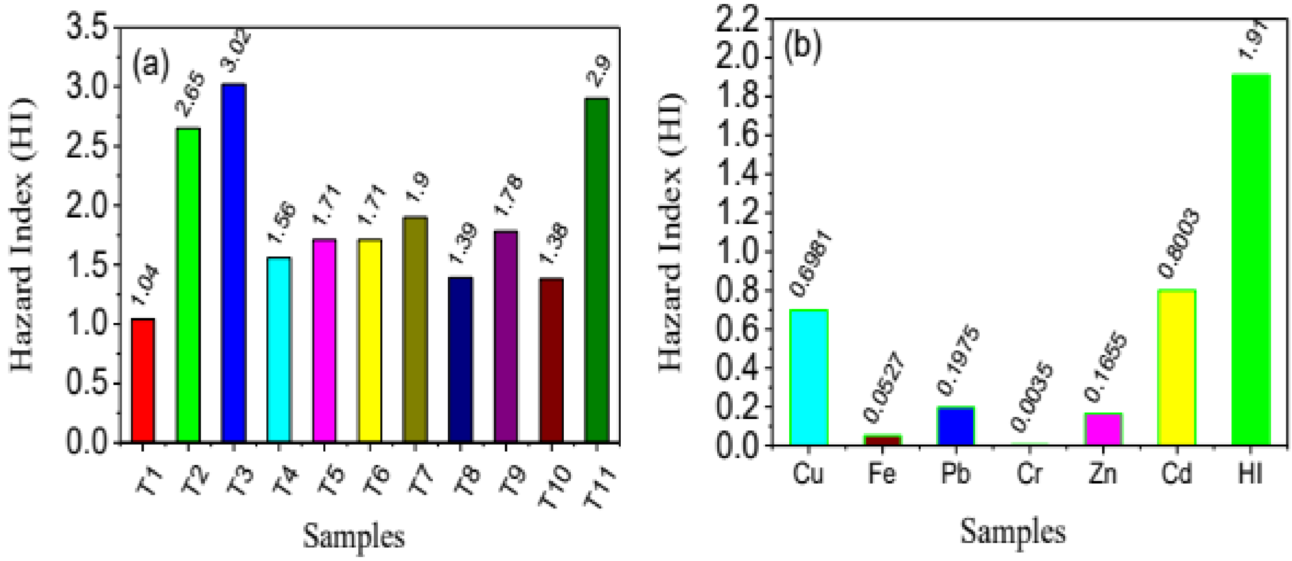

Fig.4. a) Hazard Index, and b) THQ $_{average}$ and corresponding HI for; Cu, Fe, Pb, Cd, Zn, and Cr.

4.Conclusion

This study monitored the level of several heavy metals, including Cu, Fe, Pb, Cd, Zn, and Cr, in tea samples gathered from Jazan local markets in southern Saudi Arabia. According to the findings, the average level of the heavy metals in tea samples under investigation Cu, Fe, Pb, Cd, and Zn was found to be below the maximum allowable limit. Conversely, the mean concentration of Cr exceeds the maximum permitted thresholds, and it is crucial to keep an eye on its level in tea brands. Furthermore, THQ mean values were 0.003, 0.05, 0.16, 0.19, 0.69, and 0.8 for Cr, Fe, Zn, Pb, Cu, and Cd, respectively accordingly. The THQ values of every metal under investigation were found to be below the allowable limits (less than one) in all samples indicating the individual heavy metals have no health risk to tea consumers. Conversely, the HI value was found to be higher than the allowable limits (less than 1). These findings suggest that the different heavy metals found in the investigated samples can carry non-carcinogenic health risks and highlight the importance of regularly monitoring the concentration of heavy metals in tea samples.

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