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Micronutrients (B, Co, Cu, Fe, Mn, Mo, and Zn) content in made tea (*Camellia sinensis* L.) and tea infusion with health prospect: A critical review

Tanmoy Karak^a, Funso Raphael Kutu^b, Jyoti Rani Nath^a, Indira Sonar^a, Ranjit Kumar Paul^c, Romesh Kumar Boruah^a, Sandip Sanyal^d, Santanu Sabhapondit^e, and Amrit Kumar Dutta^a

^aUpper Assam Advisory Centre, Tea Research Association, Assam, India; ^bDepartment of Crop Science, School of Agricultural Sciences, North West University, Mafikeng Campus, Mmabatho, South Africa; ^cIndian Agricultural Statistics Research Institute, New Delhi, India; ^dDepartment of Tea Processing and Manufacturing Advisory, Tea Research Association, Tocklai Tea Research Institute, Jorhat, Assam, India; ^eDepartment of Biochemistry, Tea Research Association, Tocklai Tea Research Institute, Jorhat, Assam, India

ABSTRACT

Tea (Camellia sinensis L.) is a perennial acidophilic crop, and known to be a nonalcoholic stimulating beverage that is most widely consumed after water. The aim of this review paper is to provide a detailed documentation of selected micronutrient contents, viz. boron (B), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), and zinc (Zn) in made tea and tea infusion. Available data from the literature were used to calculate human health aspect associated with the consumption of tea infusion. A wide range of micronutrients reported in both made tea and tea infusion could be the major sources of micronutrients for human. The content of B, Co, Cu, Fe, Mn, Mo, and Zn in made tea are ranged from 3.04 to 58.44 μ g g⁻¹, below detectable limit (BDL) to 122.4 μ g g⁻¹, BDL to 602 μ g g⁻¹, 0.275 to 13,040 μ g g⁻¹ 0.004 to 15,866 μ g g⁻¹, 0.04 to 570.80 μ g g⁻¹ and 0.01 to 1120 μ g g⁻¹, respectively. Only 3.2 μ g L⁻¹ to 7.25 mg L⁻¹, 0.01 μ g L⁻¹ to 7 mg L⁻¹, 3.80 μ g L⁻¹ to 6.13 mg L⁻¹, 135.59 μ g L⁻¹ -11.05 mg L⁻¹, 0.05 μ g L⁻¹ to 1980.34 mg L⁻¹, 0.012 to 3.78 μ g L⁻¹, and 1.12 μ g L⁻¹ to 2.32 μ g L⁻¹ of B, Co, Cu, Fe, Mn, Mo, and Zn, respectively, are found in tea infusion which are lower than the prescribed limit of micronutrients in drinking water by World Health Organization. Furthermore, micronutrient contents in tea infusion depend on infusion procedure as well as on the instrument used for analysis. The proportion of micronutrients found in different tea types are 1.0-88.9% for B, 10-60% for Co, 2.0-97.8% for Cu, 67.8-89.9% for Fe, 71.0-87.4% for Mn, 13.3-34% for Mo, and 34.9-83% for Zn. From the results, it can also be concluded that consumption of three cups of tea infusion per day does not have any adverse effect on human health with respect to the referred micronutrients rather got beneficial effects to human.

Abbreviations: AAFES: Atomic absorption flame emission spectrophotometry; AAS: Atomic absorption spectrometry; CPE: Cloud point extraction; EAAS: Electrothermal atomic absorption spectrophotometry; EPMA: Electron probe microanalyzer; TAAS: Electrothermal atomic absorption spectrometry; FAAS: Flame atomic absorption spectrometry; GFAAS: Graphite furnace atomic absorption spectrophotometry; HR CS GFAAS: High-resolution continuum source graphite furnace atomic absorption spectrometry; HR-SF-ICPMS: High-resolution sector field inductively coupled mass spectrometry; ICP-AES: Inductively coupled plasma atomic emission spectrometry; ICP-ES: Inductively coupled plasma emission spectroscopy; ICP-MS: Inductively coupled plasma-mass spectrometry; ICP-OES: Inductively coupled plasma optical emission spectrometry; IDA: Substoichiometric isotope dilution; INAA: Instrumental neutron activation analysis; PGA: Neutron-induced prompt gamma-ray analysis; SWASV: Square wave anodic stripping voltammetry; TNAA: Thermal neutron activation analysis; XRF: X-ray fluorescence spectrometry

KEYWORDS

Health hazard; instruments; micronutrients; origins; tea infusion

Introduction

Micronutrients such as boron (B), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), and zinc (Zn) are essential for plant's growth and reproduction (Fageria et al., 2002; Karak et al., 2005a, Karak et al., 2005b, Das et al., 2006; Karak et al., 2006; Ryan et al., 2013). Indiscriminate use of N, P, and K fertilizers in recent decades in developing countries, and no application of micronutrients in many cases, especially in dry land agriculture (Tow et al., 2011) resulted in serious threat of micronutrient deficiency in soils and crops. According to a recent report on micronutrient status of 190 soils from different

countries of world, 49% was deficient in zinc, 31% was deficient in boron, 15% was deficient in molybdenum, 14% was deficient in copper, 10% was deficient in manganese, and 3% was deficient in iron (Graham et al., 2012). Furthermore, micronutrients cycling from plant to food play a vital role for micronutrient supplement to human health. Micronutrient deficiency in food becomes a serious concern for human health and is wide spreading globally particularly in the developing countries, which could be due to consumption of predominantly plant-based diets with poor micronutrients (Visioli and Hagen, 2007; Tripathi and Platel, 2013). According to Gupta

et al. (2008), Fe, Zn, Cu, Mn, Se, I, cobalt (Co), and Mo are the major micronutrients for human and are required in amounts 100 μ g g⁻¹ of dry matter. Most of these except Mo are often deficient in natural feed ingredients, necessitating their supplementation (Gürkan et al., 2013). However, food containing micronutrients can partially fulfill the micronutrient requirements of human, therefore, food fortification through micronutrient addition receives priority throughout the world (Martínez-Navarrete et al., 2002). According to WHO (2006), food fortification refers to the practice of deliberately increasing the content of an essential micronutrient in a food irrespective of whether the nutrient is originally present in the food (before processing) or not. The aim is to improve the nutritional quality of the food that can provide health benefit with minimum risk to health. In developed countries, micronutrient fortification in finished food product has been developed and promoted, but it is not common in most developing countries (Wei et al., 2013). Furthermore, risk-benefit-health assessment of micronutrient food fortification is essential as the requirement of micronutrients varies depending on the life stage and the population subgroups considered (Bottex et al., 2008).

Bio-fortification of commonly grown food crops with micronutrients is possible through micronutrient fertilization in growing crops, but requires nutrient management expertise (Graham et al., 2001; Wei et al., 2013; Prasad et al., 2014; da Silva Messias et al., 2015). Wei et al. (2012) reported that use of Zn fortification technique during germination an improve Zn concentration in rice, which is highly bioavailable for human intake. Iron fortification in sprouted soybean seeds during germination has been reported (Zielinska-Dawidziak and Siger, 2012). Increased uptake of soil Fe by rice plant has been achieved in some studies through genetic modification; however, there is a question for accepting this modification in several countries (Takahashi et al., 2001; Murray-Kolb et al., 2002). Zinc fortification in cereal grains through agronomic or genetic biofortification has been reported (Cakmak, 2008; Bulusu and Wesley, 2011; Velu et al., 2014). Foliar Zn application in rice cultivation can be effective in increasing grain Zn as rice plant has potential capacity for remobilization of Zn from leaves to grains; and has been widely reported in several literature (Das et al., 2004; Karak et al., 2005a; Karak et al., 2005b; Mabesa et al., 2013). Sarkar et al. (2012) suggested alternative strategy of natural and vegetarian food-based fortification for improving micronutrient status in India. The content of Fe, Zn, Cu, and Mn in the grains of Gadambalia sorghum cultivar was increased with micronutrients application (Ahmed et al., 2014).

Genetic biofortification is also a common approach for crop fortification where both traditional breeding as well as biotechnological tools may be involved (Prasad et al., 2014). Furthermore, some soil microorganisms like arbuscular mycorrhizal fungi could enhance crop quality not only through enrichment in macronutrients but also in micronutrients (Antunes et al., 2012; Pellegrino and Bedini, 2014).

From the above-mentioned literatures, it is evident that studies on the contents of micronutrient and its biofortification are mainly focused on common field crops. However crop like tea (Camellia sinensis L.) being acidophilic in nature has inherent high tendency for uptake of soluble metals from soil (Karak and Bhagat, 2010). Tea infusion is prepared from systematically processed dried tea

leaves using boiling water (Bansal et al., 2013). Made tea on the other hand is mainly produced from two kinds of tea plants, namely China type (Camellia sinensis var. sinensis) and Assam type (Camellia sinensis var. assamica). The China variety is a slow growing dwarf and shrub-like plant. It has small, erect, narrow, and serrated leaves that are dark green in color. The leaf pose is erectophile and placed at an angle of less than 50°. Assam variety is quick growing having a tall tree-type structure. Its leaves are large, broad, and mostly nonserrated with light green in color. Leaf pose is planophile (horizontal) and placed at an angle of less than 50°) (Wachira et al., 2013). Based on the processing procedures (Figure 1), tea can be divided into different kinds. These include black tea (fully oxidized during processing steps), green tea (nonfermented tea produced by drying and steaming of the fresh tea leaves), Oolong tea (partially fermented tea and lies between unfermented green tea and fermented black tea), and Pu-erh tea (prepared by full fermentation, like black tea, but it is fermented for a longer duration). The Pu-erh tea is also called as a "postfermented tea" (Jain et al., 2013; Bansal et al., 2013). It has been well documented that consumption pattern of tea infusion consists of 76.78% black tea, 20.22% green tea, and <2% of a mixture of Puerh, Oolong, and white tea (Costa et al., 2002; Lee and Foo, 2013; Lv et al., 2013b). Tea is a widely popular nonalcoholic, medicinal, nutraceutical, and stimulating beverage consumed by over twothird of the world population, the use of which as a beverage dates back to 350 AD in China (Dufresne and Farnworth, 2001; Karak and Bhagat, 2010; Karak et al., 2011; Bansal et al., 2013; Pinto, 2013; Karak et al., 2014a, b; Hayat et al., 2015).

A comparative statistics over 25-year period (1987 and 2012) of tea cultivation in the world is presented in Figure 2A-F. The economic and social importance of tea is easily understood from the fact that presently 48 countries in the world are cultivating tea, while its tremendous growth of harvesting area stands at about 49.1% within the last 25 years (FAOSTAT, 2014). Presently in the world, 3.27 million ha of land is covered by tea plant. The percent sharing of tea-harvesting area in the year 1987 by different countries are in the order China (37.83) > India (18.70) > Sri Lanka (10.06) > Indonesia (4.47) > Kenya (3.88) > Turkey (3.67) > Japan (2.72) > Myanmar (2.52) > Bangladesh (2.08) >Viet Nam (1.98) > Argentina (1.67). In 2012, however, global trend of total tea cultivated land was China (46.18) > India (18.47) > Sri Lanka (6.78) > Kenya (5.82) > Indonesia (3.74) > Viet Nam (3.54) > Myanmar (2.41) > Turkey (2.32) > Bangladesh (1.77) > Japan (1.40) > Argentina (1.16). The tremendous increase in total tea-cultivated land between the years 1987 and 2012 was greatest in Viet Nam (78.8%) followed by Kenya (50.0%) and China (22.1%). Although there was increase in the total tea cultivated area in some countries, dwindling trend in total cultivated area was observed in Japan (-48.5%) > Turkey (-36.8%) > Sri Lanka (-32.6%) > Argentina (-30.5%) > Indonesia (-16.3%) > Bangladesh (-14.9%) > Myanmar (-4.4%) > India (-1.2%) between 1987 and 2012. The world tea production in 2012 was around 4.81 million tons (FAOSTAT, 2014). Among the tea producing countries, the trend of percent contribution towards the total global production for the year 1987 was India (26.32) > China (22.66) > Sri Lanka (9.04) > USSR (6.61) > Kenya (6.60) > Turkey (5.96) > Indonesia (5.35) > Japan (4.08) > Argentina (1.92) > Iran (Islamic Republic of) (1.82) > Bangladesh (1.59) > Malawi (1.35) > Viet Nam (1.23). In 2012, it was

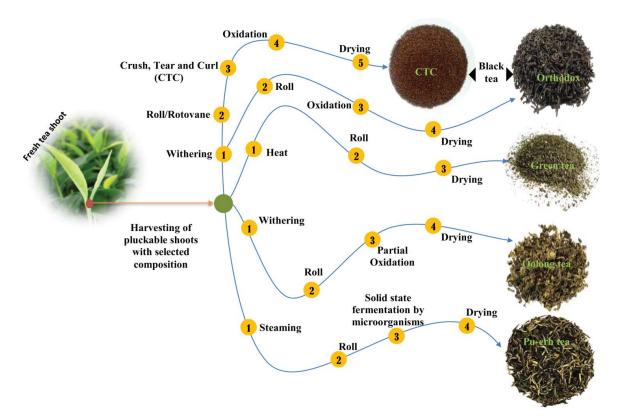


Figure 1. Flow sheet diagram for the preparation of widely consumed major class of made tea from fresh shoot of Camellia sinensis L. (Assam and China variety).

China (35.62) > India (20.77) > Kenya (7.67) > Sri Lanka (6.85) > Turkey (4.67) > Viet Nam (4.51) > Iran (3.28) > Indonesia (3.12) > Argentina (2.08) > Japan (1.78) > Thailand (1.56) > Bangladesh (1.28) > Malawi (1.11) > Uganda (1.06).

A recent comprehensive review by Karak and Bhagat (2010) documented that several nonessential trace elements were available in made tea and tea infusion. However, these elements from tea infusion did not pose any adverse health effects on its consumer. On the contrary, Feng et al. (2003) stated that much higher concentrations of trace metals influence the taste of the tea infusion and do harm to consumers' health. Furthermore, Jain et al. (2013) reported that although tea has several health benefits, analysis of the potential toxicities and possible harmful effects of high dose consumption on human health remains largely ignored. They concluded that the potential toxicity of tea consumption is mainly related to the different biochemical compounds such as flavanoids and xanthenes, present in tea. Therefore, assessment of the potential dietary flavonoid/phenolic induced is of prime importance while considering their beneficial properties. On the contrary, several micronutrients particularly B, Co, Cu, Fe, Mn, Mo, and Zn may be present in made tea and tea infusion, which may have several health benefits (Szymczycha-Madeja et al., 2015). A large number of wideranging reviews about tea components suggest the presence of heavy metals, catechins and other nutrient and antioxidative compounds (Cabrera et al., 2003; Karak and Bhagat, 2010; Welna et al., 2012). The concentration of micronutrients in made tea and tea infusion depends on the quality of soil on which the tea plants are grown (Yemane et al., 2008; Karak et al., 2014b), its origin (Karak and Bhagat, 2010; Szymczycha-Madeja et al., 2012), agricultural practices (Jin et al., 2008; Hunter et al., 2011), pruning status (Kumar et al., 2013), and climatic conditions (Nath, 2013). The

technical procedures adopted for sample preparation for made tea (Fernandez et al., 2002; Gallaher et al., 2006; Pohl and Prusisz, 2007; Szymczycha-Madeja et al., 2012; Welna et al., 2013; Memić et al., 2014 and references there in) and tea infusion (Fernández et al., 2002) also play a great role. Furthermore, the analytical methodologies used for micronutrients measurement in food and beverage like tea is also a crucial one since tea infusion is considered as daily nonalcoholic stimulant in many countries. Hence, the correct measurement of micronutrients in tea infusion is quite important to understand their implications on human health (Mucchino and Musci, 2014). Further, curative properties of tea are reported to have helped to keep a number of diseases at bay (Cooper et al., 2005). Several articles have been published on trace element content in made tea and tea infusion. However, there is a paucity of information regarding the micronutrient status in made tea and tea infusion in review articles despite the numerous data generated in different countries. The determination of micronutrients in made tea and tea infusion is important from two aspects (a) to judge their nutritional value, and (b) to guard against any probable ill-effects due to high concentrations (Karak and Bhagat, 2010). In view of the limited clarity, we attempted to make a systematic review of the recent findings on different micronutrients (B, Co, Cu, Fe, Mn, Mo, and Zn) in made tea and tea infusion based on the available literatures.

Boron

Boron (B) is a beneficial element for humans but its requirement for human body is, however, undervalued due to inadequate proven records of its requirement (Krejčová and Černohorský, 2003; Fageria and Moreira, 2011). It has been reported that B in

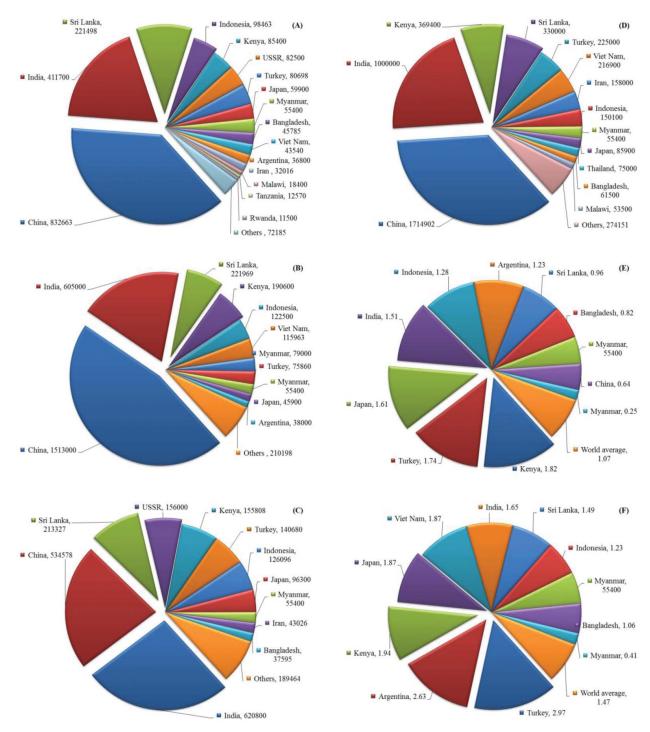


Figure 2. Comparative statistics of global tea cultivation between 1987 and 2012: (A) total cultivated area in ha for the year 1987; (B) total cultivated area in ha for the year 2012; (C) country-wise total production (ton) for the year 1987; (D) country-wise total production (ton) for the year 2012; (E) average yield (t ha⁻¹) for the year 2012 (Source: FAOSTAT, 2014).

human body helps in building and maintaining healthy bones and muscle through Ca absorption, and also beneficial for central nervous system function and inflammatory response (Devirian and Volpe, 2003; Nielsen, 2009; Gupta and Gupta, 2014). Boron stimulates brain function and together with other minerals such as calcium, magnesium, and vitamin D, it also helps to prevent osteoporosis (Krejčová, and Černohorský, 2003). According to WHO guideline, the minimum and maximum values of B in drinking water has been set as 0.2 and 5 mg L⁻¹, respectively, with an average value of 2.4 mg L⁻¹ (WHO, 2009). Total daily

boron intake in human diet ranges from a low of 0.35 to a high of 3.25 mg B d⁻¹ for adults (Nielsen, 2014). Anitha et al. (2013) reported that the average daily amount ingested from tea infusion of 10 g tea is around 0.04–0.10 mg which is about 0.3–1.0% of required amount. Boron is a micronutrient for tea plant and its deficiency afects the growth as well as the quality of made tea (Baruah et al., 2011). However, information on B application in tea cultivation is scarce. Gohain et al. (2000) applied different doses of B for three years to observe the yield and quality of tea in the tea growing region of Assam, India. They concluded that

annual foliar B application of 1 kg ha⁻¹ as boric acid with three to four splits increased tea production by 12% over control treatment. They also concluded that B application had little effect on the quality of made tea. Boron concentration measured in 10 surface soil samples collected from some tea garden belts of Golaghat district, Assam, India was below the detection limit (Baruah et al., 2011). Hajiboland et al. (2013a) reported that tea plant is highly tolerant towards B deficiency while cell wall tightening as well as accumulation of oxidized phenolics is not established for inhibiting growth under B deficiency conditions.

Boron in made tea

In spite of a large amount of works on trace element accumulation in made tea and tea infusion, research works on B content in made tea and tea infusion is very much limited. Table 1 shows the amount of B in made tea reported by few researchers. From Table 1, it has been observed that the range of B content in Chinese and Indian black teas ranges from 3.04 to 58.44 and 9.40 to 22.00 $\mu g g^{-1}$, respectively. The overall average of B content in black tea for all the countries mentioned in Table 1 ranges from 3.04 to 58.44 μg g⁻¹. For green tea, the ranges are $3.42-5.67 \ \mu g \ g^{-1}$, $16.3-19.9 \ \mu g \ g^{-1}$, and $11.7-22.3 \ \mu g \ g^{-1}$ B for China, India and Japan respectively. The overall average of B content in green, Oolong and white teas for all the countries mentioned in Table 1 ranges from 3.42 to 26.73, 13.7 to 35.6, and 9.9 to 33.4 μ g g⁻¹, respectively. Zhu et al. (2011) reported that B content in green tea samples collected from Shizuoka Prefecture of Japan was ranged between 19.6 \pm 0.4 and 20.0 \pm $0.4 \ \mu g \ g^{-1}$. Malik et al. (2008) analyzed B content in 18 types of pure mixture of tea leaves (four black teas, four green teas,

four Oolong teas, and six white teas) available in the market from Czech Republic and found a wide range from 0.8 ± 0.9 to $31.5 \pm 1.9 \ \mu g \ g^{-1}$. They reported that the levels of B did not differ significantly among tea types. Hajiboland et al. (2013b) concluded that B content and concentration in leaves and shoots were lower in B deficient compared to B sufficient plants. In their study, they reported that B in young tea shoots with adequate B supply (46 μM H₃BO₃) and low B supply were $4.90\pm0.81~\mu\mathrm{g}~\mathrm{g}^{-1}$ and $3.29\pm0.13~\mu\mathrm{g}~\mathrm{g}^{-1}$, respectively. Boron content of green, black, and Oolong teas that originated from different countries were from 3.50 to 52.4 μ g g⁻¹, 3.10 to 57.8 μ g g⁻¹, and 14.0 to 56.0 μ g g⁻¹, respectively, as reported by Krejčová and Černohorský (2003), Szymczycha-Madeja et al. (2012) and Özcan et al. (2008). Boron contents of five Turkish teas after microwave digestion ranged from 9.50 to $16.36 \ \mu g \ g^{-1}$ (Piskin et al., 2013). The content of B in made tea produced from eight tea cultivars (e.g., Jinfeng tea, Meizhan tea, Longjing tea, Zhenghe dabai tea, Yingshuang tea, Maoxie tea, Jiukengzhong tea, and Biyun tea) in China ranged from 32.78 to 57.81 $\mu g g^{-1}$ (Chen et al., 2009). These authors reported that, the eight cultivars were grown in the same site, and subjected to the same conditions of soil and environment. Therefore, the differences in B content of tea leaves belong to the eight cultivars were mainly attributed to cultivar variation largely because of their heterogeneity due to cross-pollination.

Boron in tea infusion

Reported data of B content in tea infusion is very much limited. Table 2 represents the B amount based on available literature. From Table 2, it has been observed that the range of B content

Table 1. Boron in black, green, Oolong and white tea.

Type of made tea	Country of origin	Instrument(s) used for analysis [†]	Concentration (μ g g ⁻¹)	Reference(s) [‡]
Black	China	a	$3.21 \pm 0.17 – 57.81 \pm 0.63$	1,2,3
	Range(for Chinese black tea)		3.04-58.44	
	India	a	9.9 ± 0.5 – 21.1 ± 0.9	3
	Range(for Indian black tea)		9.40-22.00	
	Indonesia	a	16.5-18.2	3
	Kenya	a	8.3-12.2	3
	Sri Lanka	a	8.9–16.5	3
	Turkey	a	14.1 ± 1.90	4
Over all range (for black tea)			3.04-58.44	
Green	China	a	$3.54 \pm 0.12 – 5.52 \pm 0.15$	1
	Range(for Chinese green tea)		3.42-5.67	
	India	b	17.5 \pm 1.2–18.9 \pm 1.0	5
	Range(for Indian green tea)		16.3–19.9	
	Japan	b, c, d, e	11.9 ± 0.2 – 22.1 ± 0.2	5,6,7
	Range (for Japanese green tea)		11.7–22.3	
	Turkey	a	24.6 ± 2.13	4
Over all range (for green tea)			3.42–26.73	
Oolong	China	b	15.1 v 1.4–25.6 \pm 10.0	5
	Range(for Chinese Oolong tea)		13.7–35.6	
	Indonesia	b	14.0 ± 0.3	5
Over all range (for Oolong tea)			13.7–35.6	
White	China	b	10.8 ± 0.9	5
	India	b	$12.8 \pm 1.8 – 31.5 \pm 1.9$	5
	Range(for Indian white tea)		11.0–33.4	
Over all range (for white tea)			9.9–33.4	

[†]In the column, the following letters denote the following points: a, ICP-AES (inductively coupled plasma atomic emission spectrometry); b, FAAS (flame atomic absorption spectrometry); c, HR-ICP-MS (high-resolution inductively coupled mass spectrometry); d, ICP-OES (inductively coupled plasma optical emission spectrometry); e, GFAAS (graphite-furnace atomic-absorption spectrometry).

[‡]In the column, the following digits denote the following references: 1, Krejčová and Černohorský, 2003; 2, Chen et al., 2009; 3, Zioła-Frankowska et al., 2014; 4, Özcan et al., 2008; 5, Malik et al., 2008; 6, Islam and Ebihara, 2012; 7, Zhu et al., 2011.



Table 2. Concentration of B in black, green, Oolong, and white tea infusion.

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L^{-1})	Reference [‡]
Black	China	1	a	1.45–7.25	1
		2	a	3.21 ± 0.17 – 9.25 ± 0.42	2
	Range (for Chines	se black tea)		1.45-9.67	
	India	1	a	$2.18 \pm 0.10 – 3.09 \pm 0.10$	1
		3	b	$0.197 \pm 0.047 – 0.403 \pm 0.012$	3
	Range (for Indian	black tea)		0.15-3.19	
	Indonesia	1	a	3.27 ± 0.15	1
	Kenya	1	a	2.63 ± 0.35	1
	South Africa	1	a	2.95 ± 0.17	1
	Sri Lanka	1	a	$2.23 \pm 0.07 – 2.68 \pm 0.14$	1
	Range (for Sri Lar	nkan black tea)		2.16-2.82	
	Turkey	4	a	0. 35	4
Over all range (for blace	:k tea)			0.15-9.67	
Green	China	2	a	$3.54 \pm 0.12 – 5.52 \pm 0.15$	2
	India	3	b	$0.165 \pm 0.037 – 0.259 \pm 0.009$	3
	Japan	3	b	0.512 ± 0.071	3
	Turkey	4	a	0.78	3
Over all range (for gree	en tea)			0.13-5.67	
Oolong	China	3	b	$0.123 \pm 0.019 – 0.307 \pm 0.053$	3
•	Indonesia	3	b	0.214 ± 0.054	3
Over all range (for Oole	ong tea)			0.10-0.36	
White	China	3	b	0.204 ± 0.027	3
	India	3	b	$0.134 \pm 0.017 – 0.412 \pm 0.058$	3
Over all range (for white	te tea)			0.12-0.47	

^{*}In the column, the following bold digits denote the following points: 1, 10 g of black tea brewed in 100 mL double distilled for one hour at 80°C; 2, 1 g tea extracted with 50 mL boiling demineralized water for five minutes and volume was made up to 100 mL; 3, 1 g of tea was carefully weighed out into standardized glass beakers and 50 mL of boiled distilled water was poured into the glass beakers and infused for 15 minutes; 4, 2 g tea was added in 98 mL distilled water and heated in a steel kettle, and was allowed to stay for five minutes after boiling.

in Chinese, Indian, and Sri Lankan black tea infusion ranges from 1.45 to 9.67, 0.15 to 3.19, and 2.16 to 2.82 mg L^{-1} , respectively. The overall average of B content in black tea infusion for all the countries mentioned in Table 2 ranges from 0.15 to 9.67 mg L^{-1} . The overall average of B content in green, Oolong, and white tea infusions ranges from 0.13 to 5.67 mg L^{-1} , 0.10 to 0.36 mg L^{-1} , and $0.12 \text{ to } 0.47 \text{ mg L}^{-1}$, respectively. Boron content in tea infusions produced from 15 tea samples originated from different tea growing countries of Asia ranged from 0.123 \pm 0.019 to 0.512 \pm 0.071 mg L⁻¹ (Malik et al., 2008). Krejčová and Černohorský (2002) investigated the B content in tea produced in China and reported that the mean extracted boron content are ranged from 3.21 \pm 0.17 to 9.25 \pm 0.421 $\mu g g^{-1}$ and from 3.5 \pm 0.12 to 5.52 \pm 0.15 μ g g⁻¹ in black and green tea, respectively. Boron concentrations measured in infusion of black and green tea samples obtained from the local market of Istanbul, Turkey were 0.728 and 0.544 mg L⁻¹, respectively (Derun et al., 2010). However, B contents of five different Turkish tea infusions ranged from 0.081 to 2.023 mg L⁻¹; and correspond to extraction rate of 0.5-18.5% (Piskin et al., 2013). Kronborg (2013) prepared tea infusions from 75 loose tea and tea bag samples collected from Poland and Norway and reported 8.24-44.5 μg B L $^{-1}$ (mean: 19 \pm 10.8 μg L $^{-1}$) and 5.71–91.9 $\mu g \ B \ L^{-1}$ (mean: 31.9 \pm 19.4 $\mu g \ L^{-1}$), respectively. However, comparatively higher B content was extracted from the tea bags, which ranged from 28.9 to 95.8 μ g L⁻¹ (mean: $48.9 \pm 24.7 \ \mu g \ L^{-1}$) and 24.5 to 141 $\mu g \ L^{-1}$ (mean: 75.2 \pm 27.9 μ g L⁻¹) in Poland and Norway samples, respectively. Moreover, the B content determined by Özcan et al. (2008), in black tea, infusion originated from Turkey were much higher

and amounted to $0.35~{\rm mg~L}^{-1}$. Zioła-Frankowska et al. (2014) also reported that the degree of extraction of B from black tea marketed in Poland ranged from 8 to 27%.

Cobalt

Cobalt (Co) is a transition element found throughout the environment as well as in common foods (Unice et al., 2012). Gál et al. (2008) reported that Co is an essential element required in trace amounts by humans and other mammals. In general, more routinely consumed plant parts like fruit, grain, and seeds contain low amount of Co as plants can accumulate only small amount of Co (ATSDR, 2004; Gál et al., 2008). Cobalt uptake by plants from soil and its accumulation is species dependent, while its availability depends on soil pH and presence of Mn oxides in soil (Gál et al., 2008). As tea plant grows in MnO₂rich soils, Co is strongly sorbed and co-precipitated with MnO₂ promoting its availability to tea plants. Hokin et al. (2004) reported that typical international total dietary Co intake ranges from 11 to $45\mu g d^{-1}$. However, dietary Co intake is estimated to range from 5 to 40 μ g d⁻¹ in the general Chinese population (Kim et al., 2006). In human body, Co requirement for vitamin B12 synthesis is approximately 0.1 μ g d⁻¹ and it serves as a cofactor in the synthesis of methionine and the metabolism of folates and purines (Varela-Moreiras et al., 2009; Unice et al., 2012). According to ATSDR (2004), the allowance of vitamin B12 for adults is 2.4 μ g d⁻¹, which corresponds to 0.1 μ g d⁻¹ of Co. However, some purveyors of dietary supplements recommend daily intake of cobalt chloride (CoCl₂) formulations ranging from 200 to 1000 μ g Co d⁻¹ (Unice et al., 2012). Intake

[†]In the column, the following letters denote the following points: a, ICP-AES (inductively coupled plasma atomic emission spectrometry); b, FAAS (flame atomic absorption spectrometry).

in the column, the following lightface digits denote the following references: 1, Zioła-Frankowska et al., 2014; 2, Krejčová and Černohorský, 2003; 3, Malik et al., 2008; 4, Özcan et al., 2008.

of CoCl₂ at a sufficient oral dose has long been known to stimulate red blood cell production, and is a potent erythropoietin transcription inducer (Jelkmann and Lundby, 2011). An amount of 0.16-1.0 mg Co kg⁻¹ of body weight is recommended as a treatment for anemia including pregnant women as it helps producing red blood cells (ATSDR, 2004).

Cobalt in made tea

Like B, information related to Co contents in made tea and tea infusion is scanty. The concentration of Co in five commercial tea samples collected from Shillong in the state of Meghalaya, India ranged from 1.26 to 1.32 $\mu g g^{-1}$ with a mean value of $1.28 \pm 0.02~\mu \mathrm{g}~\mathrm{g}^{-1}$ (Marbaniang et al., 2011). Shen and Chen (2008) reported very low average concentration of Co (0.06-0.40 μ g g⁻¹, mean: 0.20 μ g g⁻¹) in black tea samples compared to green tea (0.4–1.2 μ g g⁻¹, mean: 0.7 μ g g⁻¹) and Oolong tea (0.09–0.6 μ g g⁻¹, mean: 0.3 μ g g⁻¹) samples from Taiwan. However, higher concentrations of Co, that is, 11.0 \pm 3.4 and $21.8 \pm 1.1 \ \mu g \ g^{-1}$ have been reported by Waqar and Mian (2008) and Narin et al. (2004), for Saudi Arabian and Turkish black tea samples respectively. Nookabkaew et al. (2006) reported that Co content in black tea samples in Thailand ranged from 0.119 to 0.765 μg g⁻¹. Zhu et al. (2011) reported that Co content in green tea samples collected from Shizuoka Prefecture of Japan were between 0.255 \pm 0.002 and 0.260 \pm $0.003 \ \mu g \ g^{-1}$. Cobalt content in tea samples from Ethiopia were found to vary between 7.54 and 21.2 μ g g⁻¹ as reported by Yemane et al. (2008). Comparatively low amounts of Co in four commercially available teas purchased from local market of Poland were found and the range was 0.18 \pm 0.02 to 0.57 \pm $0.05~\mu \mathrm{g}~\mathrm{g}^{-1}$ (Pękal et al., 2013). A total of 48 commercial tea samples (15 green tea, 15 black tea, and 18 Oolong tea) were collected from six markets in Hsinchu, Taiwan and Co in green tea, black tea, and Oolong tea were found from 0.4 to 1.2 μg g^{-1} (mean: 0.7 $\mu g g^{-1}$), 0.06 to 0.40 $\mu g g^{-1}$ (mean: 0.2 $\mu g g^{-1}$), and 0.09 to 0.60 μ g g⁻¹ (mean: 0.3 μ g g⁻¹), respectively (Shen and Chen, 2008). Cobalt content in six marked brands of black tea and one type of green tea commonly consumed in Egypt ranged between below detectable limit (BDL) and 0.13 μ g g⁻¹ (Lasheen et al., 2008). Al-Othman et al. (2012) analyzed 10 black tea samples available from Jazan and Jeddah region of Saudi Arabia and Co concentration varied from BDL to 3.1 μ g g^{-1} . The concentrations of Co in black tea samples reported by Ashraf and Mian (2008) were in the range from 23.7 to 122.4 μ g g⁻¹. Cobalt concentrations of tea samples from Saudi Arabia were reported to be in the range from 0.05 to 2.35 μ g g⁻¹ (AL-Oud, 2003). Islam and Ebihara (2012) reported that Co content in six green tea samples from Japan varied between $0.089 \pm 0.015 \ \mu g \ g^{-1}$ and $0.202 \pm 0.023 \ \mu g \ g^{-1}$. Cobalt in lowand high-quality instant black teas from Turkey were 0.7 and $0.6 \mu g g^{-1}$ respectively (Alasalvar et al., 2013). Dobrowolski and Otto (2012) determined Co in certified reference material sample of tea leaves was $0.41 \pm 0.02 \ \mu g \ g^{-1}$. Seven different grades of black teas (three high-quality black tea samples and four low-quality black tea samples), of Turkey were analyzed by Serpen et al. (2012) and Co contents were ranged from 0.04 \pm 0.01 to 0.05 \pm 0.00 mg per 100 g of tea and 0.03 \pm 0.00 to 0.07 ± 0.01 mg per 100 g of tea for high-quality black tea and

low-quality black tea respectively. Out of different trace elements analyzed in 18 black tea samples available in Italy Co ranged from <1 to 1.3 $\mu g g^{-1}$ as reported by Desideri et al. (2011). However comparatively lower amount of Co, that is, from 0.11 to 0.35 μ g g⁻¹ in eight black tea samples from China had been reported by Chen et al. (2009) and 0.65 \pm 0.01 μg g⁻¹ in black tea sample from Turkey by Narin and Soylak (2003). Average Co contents in 10 black bagged teas and leaf teas each originated from Sri Lanka, China, and India were found 0.21 \pm 0.15 and 0.26 \pm 0.12 μg g⁻¹, respectively (Polechońska et al., 2015). Cobalt contents in six black tea samples and one green tea sample consumed in Egypt originated from Sri Lanka were relatively low and found in the range BDL to $0.13 \ \mu g \ g^{-1}$ and BDL, respectively (Lasheen et al., 2008).

Cobalt in tea infusion

Mean Co contents in black tea infusion in Turkey ranged from 0.01 to 1.58 μ g L⁻¹, which was below the Turkish nonalcoholic beverage limit (Sofuoglu and Kavcar, 2008). The mean Co contents in green tea, black tea, and Oolong tea infusions (from Taiwan were 0.7, 0.1, and 0.1 μ g per 100 mL, each representing 59.3, 25.3, and 30.5% of total Co, respectively (Shen and Chen, 2008). Islam and Ebihara (2012) reported that Co extraction efficiencies in infusion of green tea produced in Japan was about 51% of total Co in tea. Cobalt content in loose tea and tea bag infusions of 40 samples obtained from Norwegian supermarkets ranged from 0.19 to 2.92 $\mu g L^{-1}$ (mean: 0.68 \pm $0.57~\mu {\rm g~L^{-1}})$ and $0.59~{\rm to}~3.11~\mu {\rm g~L^{-1}}$ (mean: $1.67~\pm~0.72~\mu {\rm g}$ L^{-1}), respectively. However, for 30 tea infusions obtained from Polish supermarket, Co content ranged from 0.12 to 1.15 μg L^{-1} (mean: 0.46 \pm 0.27 μ g L^{-1}) and 0.14 to 1.07 μ g L^{-1} (mean: 0.54 \pm 0.34 μ g L^{-1}) for loose tea and tea bag, respectively (Kronborg, 2013). Polechońska et al. (2015) concluded that Co contents in most of tea infusions produced from twenty black tea samples were BDL when detection limit of Co was 0.066 mg kg⁻¹ except black tea produced from China and Sri Lanka $(0.14-0.23 \mu g L^{-1})$. Paz-Rodríguez et al. (2015) reported that Co contents in black tea and green tea infusions from China were 8.2 and 7.9 to 21.8 μ g L⁻¹, respectively.

Copper

Copper (Cu) is an essential mineral for human health but consumption of excess amount may be harmful (Araya et al., 2007; Gupta and Gupta, 2014). This element is associated with alterations in cholesterol metabolism, bone health, cardiovascular risk, immune function, and frequency of disease infections (Araya et al., 2007; Gupta and Gupta, 2014). Its deficiency may lead to decreased Fe levels in some human tissues (Gupta and Gupta, 2014). WHO (2003a), b sets the guideline value of Cu for drinking water at 2.0 mg L⁻¹, while the tolerable upper intake level of Cu is 10 mg d⁻¹ (FNB, 2001). Several plant foods like bean, black pepper, Brazilian nut, cashew, cereal, chick pea, cocoa, prune, and sunflower seed are good sources of Cu; and a balanced diet can provide up to 50% of the required Cu intake for human (Gupta and Gupta, 2014). Jin et al. (2008) reported that both edaphic and nonedaphic factors can contribute to the final Cu accumulation in tea leaves and may serve as an

important Cu source for tea consumer. Data on Cu content of made and tea infusion had been reported in our previously published article (Karak and Bhagat, 2010); however, for the late readers, we have documented selected recent data in relation to Cu in made and tea infusion in this section.

Copper in made tea

Table 3 presented Cu concentrations in different types of made tea available in existing literatures. From Table 3, it has been observed that the range of Cu contents in Chinese, Ethiopian, Indian, Indonesian, Iranian, Kenyan, Saudi Arabian, Sri Lankan, Turkish, and unknown origins' black teas range from 0.05 to 602.5, BDL to 27.7, 7.2 to 377.6, 24 to 582.2, 6.07 to 211, 4.03 to 38.1, 5.2 to 382.1, 10.1 to 413.1, 0.05 to 405.8, and 10.88 to 119.61 μ g g⁻¹, respectively. The overall average of Cu content in black tea for all the countries mentioned in Table 3 ranges from BDL to 602.5 μ g g⁻¹. For green tea, the ranges are from 0.14 to 270, 10.4 to 37.6, 12.6 to 29, 11.3 to 25.8, 15.6 to 26.7, 0.2 to 119.6, and 9.1 to 38 $\mu g g^{-1}$ Cu for Chinese, Indian, Indonesian, Japanese, Sri Lankan, Turkish, and unknown origins' green teas respectively. The overall average of Cu content in green tea for all the countries mentioned in Table 3 ranges from 0.14 to 270 μ g g⁻¹. For Oolong tea, the ranges are from 4 to 104.8 and 0.70 to 30 μ g g⁻¹ Cu for Chinese and unknown origins' Oolong teas respectively. The overall average of Cu content in Oolong tea for all the countries mentioned in Table 3 ranges from 0.70 to 104.8 $\mu \mathrm{g}~\mathrm{g}^{-1}$. The overall average of Cu content in Pu-erh and white teas for all the countries mentioned in Table 3 ranges from 12 to 43 and 10 to 26 μ g g⁻¹, respectively. Copper contents in Indian and US tea brands ranged from 1.60 to 35.0 and 4.4 to 17.3 μ g g⁻¹, with mean values of 14.8 \pm 8.2 and 12.3 \pm 4.8 μ g g⁻¹, respectively, although, relatively varied over wide ranges. However, Cu content in Indian tea brands had a much wider range compared to that in US brands (Kumar et al., 2005). Jin et al. (2008) reported that Cu accumulation in tea leaves produced in twenty tea gardens of Yuyao County in China was below 60 μ g g⁻¹, a permissible level given by the Chinese Ministry of Health. Approximately 15% of the samples were higher than 15 μ g g⁻¹, which is the permissible limit in "green food" as defined by the Chinese Ministry of Agriculture. These observations suggest that Cu concentrations in tea leaves from different tea producing areas are within acceptable limit, although some degree of concern still remains. The authors concluded that Cu concentrations in made tea might have been greatly increased by the machinery processing in factories that used copper boards at the twisting stage. Among 13 black tea samples from different factories, the Cu levels ranged from 8.89 \pm 0.38 to 39.66 \pm 2.69 $\mu g g^{-1}$ (Jin et al., 2008). This study further suggests that both edaphic and nonedaphic factors can contribute to the final Cu accumulation in tea leaves used by consumers. Among the several samples analyzed from Turkey, the concentration of Cu varied between 6.4 and 13.1 μ g g⁻¹ (Soylak et al., 2007). Again, Cur level in tea samples from Turkey and United Kingdom were reported as 16.5 $\mu g g^{-1}$ (Colak et al., 2005) and 21.0 $\mu g g^{-1}$ (Mehra and Baker, 2007), respectively. Eight made tea samples from a local supermarket of United Kingdom imported from different parts of the world were analyzed and total Cu concentration in the

samples ranged from 15.7 to 34.0 μ g g⁻¹ (Mehra and Baker, 2007). However, Chen (1984) reported that Cu content of all analyzed tea samples from China were below the upper limits imposed by various countries: Australia (150 μ g g⁻¹), China (60 μ g g⁻¹), Germany (40 μ g g⁻¹), Japan (100 μ g g⁻¹), United Kingdom (150 μ g g⁻¹), and the United States (150 μ g g⁻¹). Copper content in five different black tea samples from Mumbai, India ranged from 0.6293 \pm 0.36 to 4.2635 \pm 1.59 $\mu {
m g}~{
m g}^{-1}$ (Ambadekar et al., 2012). The Cu concentration in four types of commercially produced tea samples from Zhejiang province at the eastern part of China was found to range from 14.43 to 24.05 μ g kg⁻¹ (Xiong et al., 2012). Copper in 10 tea samples available in the supermarkets from Kayseri and Tokat, Turkey varied from 6.4 to 13.1 μ g g⁻¹ (Soylak et al., 2007).

Experimental results published by Seenivasan et al. (2008) on the analysis of Cu content in 86 black tea samples collected from South India indicated that Cu contents were in between 15.9 and 32.2 $\mu g g^{-1}$ (mean: 24.07 \pm 2.25 $\mu g g^{-1}$). Copper contents of five commercial tea samples collected from Shillong in the state of Meghalaya, India ranged from 7.2 to 10.5 μ g g⁻¹ with a mean value of 9.0 \pm 1.3 μg g⁻¹ (Marbaniang et al., 2011). In 57 commercial tea samples purchased from the local market in Beijing, China, Cu contents varied from 1.74 to 21.15 μ g g⁻¹ (average, 12.63 μ g g⁻¹) for green tea; 12.10 to 48.19 μ g g⁻¹ (average, 22.02 μ g g⁻¹) for Oolong tea; and 8.72 to 28.78 μ g g⁻¹ (average, 23.88 μ g g⁻¹) for black tea (Qin and Chen, 2007). A report on Cu content in African and Asian teas revealed a range of 12.4-19.34 $\mu g g^{-1}$ (Moreda-Piñeiro et al., 2003). Tautkus et al. (2004) revealed that Cu concentration ranged from 19.8 to 33.9 μ g g⁻¹ in five different tea samples, viz. Brooke Bond, Georgian, Dilmah, Ceylon, and Chinese green teas. On analyzing 18 tea samples (4 samples produced in Thailand and 14 samples imported from China, Japan, Sri Lanka, India, and Indonesia), Cu contents were found to range from 3.08 to 22.42 μ g g⁻¹ with the mean value of 15.20 μ g g⁻¹ (Nookabkaew et al., 2006). Narin et al. (2004) reported a mean Cu concentration of 24.8 \pm 1.4 μ g g⁻¹ in black tea samples from Turkey. Zhu et al. (2011) reported that Cu content in green tea samples collected from Shizuoka Prefecture of Japan varied between 18.93 \pm 0.12 and 19.66 \pm 0.24 $\mu g g^{-1}$. Ahmad et al. (2012) analyzed Cu in 12 black tea samples commonly consumed in Pakistan and Cu contents were between 0.01 and $2.40 \ \mu g \ g^{-1}$ with the mean of $0.82 \ \mu g \ g^{-1}$.

The total Cu contents in 16 tea samples (10 were of Ethiopian origin, 5 tea samples were of Sri Lankan origin, and 1 from the United Kingdom) procured from nearby supermarkets in Addis Ababa, Ethiopia ranged from 4.7 to 12.9 μ g g⁻¹ (Ashenef, 2014). Yemane et al. (2008) reported 2.88-25.6 μ g g⁻¹ of Cu for Ethiopian black tea collected from a farm site. Additional investigations of three Ethiopian commercial tea brands, namely, Wushwush, Gumero, and Black Lion, done by Woldegebriel et al. (2012) reported 9.1 to 10.3 μ g g⁻¹ of Cu. The contents of Cu in six green tea samples (three green teas from India, China and Japan, and three kinds at the local markets in Ravenna, Italy) available in Italy varied between 9.6 \pm 0.2 and 23.8 \pm 0.4 $\mu g g^{-1}$ (Melucci et al., 2013). Among 48 commercial tea samples in China comprising of 15 green tea, 15 black tea, and 18 Oolong tea, measured Cu contents were 0.2-0.9 μ g g⁻¹ (mean: 0.4 μ g g⁻¹), 0.05-0.7 μ g g⁻¹ (mean:

Table 3. Copper content in black, green, Oolong, Pu-erh, and white tea.

ype of made tea	Country of origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference
Black	Africa (Burundi, Ethiopia, Kenya, Malawi, Tanzania, and Zimbabwe)	a	19.14 ± 8.80	1
	Argentina	b	377.5-602.5	2
	Asia (India and Sri Lanka)	a	21.25 ± 8.79	1
	Bangladesh	a	0.05-0.07	3
	China	С	28-65.1	4
		b	140.3-270.0	2
		b	8.1-33.5	5
		С	12–35	6
		C	16.6–23	7
		d	8.72-28.78	8
		e	22.61-33.45	9
		b	10.26-37.41	10
	Range (for Chinese black tea)		0.05-602.5	
	England	f	BDL-27.7	11
	Ethiopia	d	4.7–12.9	12
	21110 111	b	$9.1 \pm 0.14 – 11.5 \pm 0.1$	13
		b	BDL-19.15	14
	Range (for Ethiopian black tea)	D .	BDL-27.7	
	India	С	26–31.5	4
	IIIuid			2
		b	377.6	
		g	$12.3 \pm 4.8 - 17.6 \pm 0.8$	15
		b	7.2–10.5	16
		C	18.2–20.9	7
		C .	19.84–26.64	17
		d	12.1–44.1	18
		b	23.3–33.9	19
	Range (for Indian black tea)		7.2–377.6	
	Indonesia	h	25.3 ± 1.3	20
		b	385.9–582.2	2
	Range (for Indonesian black tea)		24–582.2	
	Iran	b	23.43-33.60	21
		b	29.3 ± 6.37	22
		b	110.0-211.0	23
		b	6.07-15.24	24
		C	40.19-58.48	17
		d	40.06 ± 2.04	25
	Range (for Iranian black tea)		6.07-211	
	Japan Japan	a	22.1 ± 0.1	26
	Kenya	C	38.1	4
	Kenyu	f	26.6	11
		b	9.0–17.5	27
		b	9.0–17.5 9.0–17.5	27
				28
		b	8.7 ± 4.67	
	Dange (for Konsen black too)	b	9.54–17.13	28
	Range (for Kenyan black tea)	L.	4.03–38.1	2
	Myanmar	b	382.1	2
	Pakistan	b	10.3–12.6	29
		b	18.4–23.9	30
	Russia	C	13.8	4
		b	277.8	2
	Rwanda	b	14.3	27
	Saudi Arabia	b	5.2–21.6	31
		b	22.12-40.66	32
	Range (for Saudi Arabian black tea)		5.2-382.1	
	Singapore	i	23.94	33
	Sri Lanka	С	27.1-36.2	4
		j	18.6	34
		ŕ	12.2-46.7	11
		b	413.1	2
		b	11.78–32.42	35
		C	23–34	7
			35.79–39.57	, 17
		C b		
		b	10.80 ± 0.70	36 10
	Danier (for Call 1 1 1 1 1 1 1 1 1	b	28.9	19
	Range (for Sri Lankan black tea)	•	10.1–413.1	
	Syria	b	10.6–54.4	37
		b	320.0-405.8	2
	Taiwan	a	0.05-0.7	38
	T	b	13.3	27
	Tanzania	b	13.3	_,

(Continued on next page)

Table 3. (Continued)

pe of made tea	Country of origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Referen
	Turkey	С	13.4	4
		b	$12.2 \pm 0.8 – 13.2 \pm 0.3$	40
		a	4.7–9.9	41
		b	16.5	42
		f	$25.83 \pm 0.62 - 120.46 \pm 0.69$	43
		a	13.1 ± 4.3	44
		b	16.0 ± 0.6	45
		b	0.5 ± 0.1	46
		b	10.4–24.8	47
		b		
			$7.24 \pm 0.38 - 13.10 \pm 0.80$	36
		b	6.4–13.1	36
		k	$7.24 \pm 0.38 – 13.10 \pm 0.80$	36
		k	10.80 ± 0.70	36
		b	0.74 ± 0.05	48
		b	12.5 ± 1.10	49
		a, 1	7.73-21.34	50
	Range (for Turkish black tea)	-, .	0.05–405.8	
	Uganda	f	119.21 ± 0.40	43
	Oganua	b	16.0	27
	Huite d Analy Francisco			
	United Arab Emirates	f	14.4	11
	Unknown	С	31.7	4
		b	10.93 ± 0.05	51
		j	15.8–23.5	34
		Č	14–39	52
		b	$38.2 \pm 1.5 – 46.8 \pm 1.1$	53
	Range (for Unknown origins black tea)	2	10.88–119.61	33
orall range (for black too)	nange (for offknown origins black tea)			
erall range (for black tea)	CI :		BDL-602.5	
en	China	C	14.6–28.5	4
		j	$0.16 \pm 0.02 – 0.19 \pm 0.03$	54
		b	19 ± 1.1	55
		C	$9.68 \pm 0.07 – 18.82 \pm 0.22$	56
		f	16	11
		i	21.83	33
		b	270.0	2
		b	$34.50 \pm 1.00 - 63.50 \pm 1.92$	57
		b	$34.50 \pm 1.00 - 63.50 \pm 1.92$	57
		b	$8.89 \pm 0.38 – 39.66 \pm 2.69$	58
		C	11–26	6
		d, m	$23.2 \pm 0.4 – 24.5 \pm 0.5$	59
		d	1.74-21.15	8
		b	17.52 ± 6.25	60
		e	17.01–63.07	9
		b	19.8	19
	D (f Cl)	b	8.99–20.28	10
	Range (for Chinese green tea)		0.14–270	
	India	C	37.6	4
		d, m	$10.5 \pm 0.1 – 10.8 \pm 0.1$	59
	Range (for Indian green tea)		10.4–37.6	
	Indonesia	С	29	4
		h	13.6 ± 1.0	20
			16.6–16.9	34
	Panga (for Indonesian groon toa)	j	12.6–29	34
	Range (for Indonesian green tea)			
	Japan	С	25.8	4
		a	11.4 ± 0.1	26
		d, m	$14.9 \pm 0.3 – 15.9 \pm 0.3$	59
	Range (for Japanese green tea)		11.3–25.8	
	South Africa	f	21.8	11
	South Korea	f	24.3	11
	Sri Lanka	j	15.8	34
	J., Zurina	, f	26.7	11
		b	15.56	35
	Range (for Sri Lankan green tea)		15.6–26.7	
	Taiwan	a	0.2-0.9	38
	Turkey	f	119.21 ± 0.40	43
	•	a	11.1 ± 3.5	44
		b	$6.39 \pm 0.26 - 9.84 \pm 0.67$	36
			0.07 ± 0.07 ± 0.07	36
		L	6 20 1 0 26 0 04 1 0 67	
	Denote (for Toda's	k	$6.39 \pm 0.26 - 9.84 \pm 0.67$	30
	Range (for Turkish green tea)		0.2-119.6	
	Range (for Turkish green tea) Unknown	k c		4
			0.2-119.6	
		С	0.2–119.6 18.2–24	4

(Continued on next page)

Table 3. (Continued)

Type of made tea	Country of origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference [†]
		n	$9.6 \pm 0.2 – 13.1 \pm 0.3$	59
		0	$9.3 \pm 0.2 – 12.7 \pm 0.2$	59
		b	$18.8 \pm 0.4 – 25.2 \pm 0.9$	53
	Range (for unknown origins green tea)		9.1–38	
	Vietnam	C	21.4	4
Overall range (for green tea)			0.14270	
Oolong	China	i	14.21	33
		b	104.8	2
		C	4–12	6
		d	12.10-48.19	8
		e	7.73-20.49	9
		b	8.38-14.29	10
	Range (for Chinese Oolong tea)		4–104.8	
	Japan	a	5.80 ± 0.22	26
	Taiwan	a	0.7–1.5	38
	Unknown	b	7.36 ± 0.03	51
		C	10–30	52
	Range (for unknown origins Oolong tea)		0.70-30	
Overall range (for Oolong tea)	g = (0.70104.8	
Pu-erh	China	C	19	4
		C	16.3-26.3	61
		a	16.3–26.3	61
		C	14.8–19.3	62
		C	15–43	6
		b	12–22	63
		e	21.06-31.35	9
	Range (for Chinese pu-erh tea)		12–43	
	Japan	a	16.1 ± 1.1	26
Overall range (for Pu-erh tea)	1243	_		
White	China	C	19.7	4
		C	10–26	6
Overall range (for white tea		-	1026	-

*In the column, the following letters denote the following points: a, ICP-MS (inductively coupled plasma mass spectrometry); b, FAAS (flame atomic absorption spectrometry); c, ICP-AES (inductively coupled plasma atomic emission spectrometry); d, GFAAS (graphite furnace atomic absorption spectrophotometry); e, HRCS-GFAAS (high-resolution continuum source graphite furnace atomic absorption spectrometry); f, ICP-OES (inductively coupled plasma optical emission spectrometry); g, TNAA (thermal neutron activation analysis); h, ICP-QMS (inductively coupled plasma quadrupole mass spectrometry); i, capillary electrophoresis; j, polarized X-ray fluorescence spectrometry; k, AAS with D₂ background corrector; l, ICP-ES (inductively coupled plasma emission spectrometry); m, SWASV (square wave anodic stripping voltammetry); n, SWASV with gold electrode; o, SWASV with hanging mercury drop electrode.

In the column, the following digits denote the following references: 1, Moreda-Piñeiro et al., 2003; 2, Ferrara et al., 2001; 3, Islam et al., 2013; 4, Street et al., 2006; 5, Jin et al., 2005; 6, McKenzie et al., 2010; 7, Mehra and Baker, 2007; 8, Qin, and Chen, 2007; 9, Zhong et al., 2015; 10, Zheng et al., 2014; 11, Donkora et al., 2015; 12, Ashenef, 2014; 13, Gebretsadik and Chandravanshi, 2010; 14, Yemane et al., 2008; 15, Kumar et al., 2005; 16, Marbaniang et al., 2011; 17, Salahinejad and Aflaki, 2010; 18, Seenivasan et al., 2008; 19, Tautkus et al., 2004; 20, Pękal et al., 2013; 21, Abdolmaleki et al., 2013; 22, Ansari et al., 2009; 23, Falahi and Hedaiati, 2013; 24, Nejatolahi et al., 2014; 25, Ziarati et al., 2013; 26, Han et al., 2014; 27, Moseti et al., 2013; 28, Omwoyo et al., 2013; 29, Jabeen et al., 2015; 30, Soomro et al., 2008; 31, Al-Othman et al., 2012; 32, AL-Oud, 2003; 33, Feng et al., 2003; 34, Desideri et al., 2011; 35, Lasheen et al., 2008; 36, Soylak et al., 2007; 37, Antakli and Al-Check, 2011; 38, Shen and Chen, 2008; 39, Nookabkaew et al., 2006; 40, Aksuner et al., 2012; 41, Alasalvar et al., 2013; 42, Colak et al., 2005; 43, Görür et al., 2011; 44, Kara, 2009; 45, Kendüzler and Türker, 2003; 46, Narin and Soylak, 2003; 47, Narin et al., 2004; 48, Uçar et al., 2014; 49, Wadhwa et al., 2014; 50, Yaylalı-Abanuzand Tüysüz, 2009; 51, Dawodu et al., 2013; 52, Herrador and González, 2001; 53, Memić et al., 2014; 54, Alassane et al., 2013; 65, Brzezicha et al., 2014; 56, Chen et al., 2009; 57, Gallaher et al., 2006; 58, Jin et al., 2008; 59, Melucci et al., 2013; 60, Wong et al., 1998; 61, Cao et al., 2010; 62, Lv et al., 2013; 63, Ning et al., 2011.

0.3 μg g⁻¹), and 0.7–1.5 μg g⁻¹ (mean: 0.9 μg g⁻¹), respectively (Shen and Chen, 2008). Mose et al. (2014) reported that the Cu content in black tea samples collected from three agroecological areas of Murang'a, Meru, and Kisii in Kenya varied between 10 \pm 3 and 16 \pm 1 μg g⁻¹. Copper content ranged from 11.78 to 32.42 μg g⁻¹ when seven tea samples were collected from local market of Egypt (Lasheen et al., 2008). Malik et al. (2008) analyzed Cu in 15 types of pure mixture of tea leaves that originated from different countries and reported that Cu content ranged from 15.1 \pm 0.3 to 66.5 \pm 4.3 μg g⁻¹. The mean level of Cu among 10 black tea samples from Jazan and Jeddah region of Saudi Arabia ranged from 5.2 to 21.6 μg g⁻¹ as reported by Al-Othman et al. (2012). AL-Oud (2003) reported the Cu content of tea and herb leaves from Saudi Arabia in the range of 9.04 to 40.66 μg g⁻¹.

The copper level of black tea samples from South India has been reported as 24.07 $\mu g g^{-1}$ (Seenivasan et al., 2008). The concentration of Cu in tea leaf samples from Syria varied between

10.6 and 54.4 μg g⁻¹ (Antakli and Al-Check, 2011), while a range of 9.1–11.5 μ g g⁻¹ was reported from Ethiopia (Gebretsadik and Chandravanshi, 2010). Among the 43 representative tea samples (including 18 green, 12 Oolong, and 13 black teas) in China, Cu content ranged widely from 8.38 to 37.41 μ g g⁻¹ (median of 17.31 μ g g⁻¹ and mean of 17.75 μ g g⁻¹; Zheng et al., 2014). The average Cu concentrations in each of these three types of tea followed the order black tea > green tea > Oolong tea. In Saudi Arabia, Shaltout et al. (2013) revealed that the total Cu concentration in six marked brands of black tea samples that originated from India, Kenya, Sri Lanka, and unknown sources ranged from 7.55 to 28.30 μ g g⁻¹ (mean: $17.0 \pm 6.8 \ \mu \mathrm{g} \ \mathrm{g}^{-1}$). This result was found to be lower than the values that ranged from 0.12 \pm 0.02 to 0.34 \pm 0.05 $\mu \mathrm{g}~\mathrm{g}^{-1}$ and $0.22 \pm 0.03 \ \mu \mathrm{g}^{-1}$ reported by Achudume and Owoeye (2010) in four black tea samples from Nigeria.

Chand et al. (2011) assessed Cu in made tea produced from more than 100 years old tea plants at the experimental farm of

the Institute of Himalayan Bioresource Technology, Council of Scientific and Industrial Research, Palampur, India and reported that Cu ranged from $0.092-0.57 \mu g g^{-1}$. Copper contents in three type of teas (green, black, and Oolong) available in local markets in Ibadan, Oyo State, Nigeria were 9.16 \pm 0.04, 10.93 ± 0.05 , and $7.36 \pm 0.03~\mu g~g^{-1}$, respectively (Dawodu et al., 2013). Eight commercial tea leaves available in Nigeria were randomly purchased at the local retail outlets in Zaria and Cu contents ranged from BDL to 64.9 \pm 14.3 $\mu g g^{-1}$ (Jonah and Williams, 2000). Memić et al. (2014) reported that total Cu contents in black tea and green tea were 46.8 \pm 1.1 and 25.2 \pm $0.9\mu g g^{-1}$, respectively.. Copper content in twenty nine marked brands of black tea and one type of green tea commonly consumed in Turkey had been reported by Görür et al. (2011) to range from 23.59 \pm 0.48 to 120.46 \pm 0.69 $\mu g g^{-1}$ (mean: $42.07 \pm 0.41 \ \mu g \ g^{-1}$). Salahinejad and Aflaki (2010) analyzed 11 black tea (4 from Iran, 3 from India, and 4 from Sri Lanka) samples available in Iran revealed that Cu content ranged from 19.84 to 58.48 μ g g⁻¹. Cu contents in 18 black tea samples available in Italy ranged from 15.4 to 30.2 μ g g⁻¹ (Desideri et al., 2011). The results of 10 tea samples from the north of Iran revealed that the level of Cu varied between 23.43 \pm 0.47 and 33.60 \pm 0.54 μ g g⁻¹ (Abdolmaleki et al., 2013). Copper contents in made tea produced from eight tea cultivars in China ranged from 9.68 to 18.82 mg kg^{-1} (Chen et al., 2009).

Lv et al. (2013a) analyzed 56 Chinese Pu-erh tea samples collected from four cities in Yunnan province of China (Dali, Lincang, Xishuangbanna, Simao, and Dehong) and reported Cu contents varied between 14.8 and 19.3 μg g⁻¹ with the mean of 16.5 μ g g⁻¹. In this study, the highest level of copper was found in samples collected from Xishuangbanna and the lowest from Simao. However, no statistically significant differences (P > 0.05) were found in copper contents of the five production areas of China. Copper contents in 17 Pu-erh tea samples have been documented by Cao et al. (2010), and Cu contents ranged between 16.3 and 26.3 μ g g⁻¹.

Copper in tea infusion

Copper concentration reported in various tea infusions are shown in Table 4. From Table 4, it has been observed that, the range of Cu contents in Chinese, Indian, Sri Lankan, Turkish, and unknown origins' black tea infusions range from 0.13 to 73.4, 0.072 to 3.72, 0.09 to 20.12, 0.09 to 65.4, and 0.15 to 18 mg L⁻¹, respectively. The overall average of Cu content in black tea infusions for all the countries mentioned in Table 4 ranges from 0.07 to 84.27 mg L^{-1} . From 0.05 to 114, 0.10 to 9.6 and 0.03 to 131.1 mg L⁻¹ Cu were found in Chinese, Turkish, and green tea originated from unknown regions respectively with the overall range from 0.03 to 131.1 mg L^{-1} . The overall average of Cu content in Pu-erh tea infusions ranges from 0.04 to 0.12 mg L^{-1} . Like other elements, Cu concentration in tea infusion depends on the origin of tea, amount present in made tea and the time allowed for the infusion process (Mehra and Baker, 2007; Mucchino and Musci, 2014). Mehra and Baker (2007) reported that the solubility of Cu in the first infusion was significantly (P < 0.01) higher than in the second infusion and that of second infusion was significantly higher than the third infusion (P < 0.01).

Copper concentrations in 56 Chinese Pu-erh tea infusions collected from Yunnan province of China were between 35.14 and 51.29 μ g L⁻¹ with the mean value of 43.18 μ g L⁻¹ (Lv et al., 2013a). The mean Cu dissolving rate of those samples ranged from 21.3 to 30.1% of total Cu with the overall average of 26.2%. Nookabkaew et al. (2006) documented Cu concentration in 18 tea infusions ranged from 1.20 to 8.43 μ g 100 mL⁻¹ with the mean value of 4.15 μ g 100 mL⁻¹. They reported that 12.96% of total Cu was released into tea infusion. The mean Cu contents in green tea (n = 15), black tea (n = 15), and Oolong tea (n = 18) infusions available in Taiwan were 1.8, 1, and 4 μ g L^{-1} , respectively, each representing 22.9, 21.8, and 23.9% of total Cu, respectively (Shen and Chen, 2008).

Copper content in tea infusions prepared from 15 tea samples originated from different tea growing countries ranged from 0.048 ± 0.006 to 0.205 ± 0.020 mg L⁻¹ (Malik et al., 2008). Amount of Cu in four Nigerian black tea infusions were between 2.33 ± 0.71 and $5.69 \pm 0.09~\mu g~L^{-1}$ with the mean $4.03 \pm$ $0.36 \ \mu g \ L^{-1}$ as reported by Achudume and Owoeye (2010).

Fernández et al. (2002) prepared infusion from 20 green teas and 24 black teas available in Spanish market and reported that Cu contents in 3 and 5 minutes' infusion ranged from 0.04 to 0.12 mg L^{-1} (mean: 0.07 mg L^{-1}) and $0.01 \text{ to } 0.12 \text{ mg L}^{-1}$ (mean: 0.06 mg L^{-1}), respectively. While Chand et al. (2011) reported that Cu in tea infusion varied between 0.008 and 0.040 mg L^{-1} and the percentage dissipation of Cu from fresh tea leaves to infusion ranged from 0.2% to 1.8%. Among the five black and green tea samples in Bosnia and Herzegovina, Cu content in tea infusion ranged from 4.28 \pm 0.74 to 4.77 \pm $0.35 \ \mu g \ g^{-1}$ (Memić et al., 2014). The content of Cu measured in tea infusions prepared from a mixture of black tea sample and tea brew from 29 tea samples collected from Turkish market ranged from 3.10 ± 0.24 to 9.17 ± 0.43 mg g⁻¹ (mean: 4.94 \pm 0.39 mg g⁻¹) and from 2.04 \pm 0.14 to 7.93 \pm 2.18 mg g⁻¹ (mean: $4.79 \pm 0.48 \text{ mg g}^{-1}$) respectively (Görür et al., 2011). Average Cu content in 11 tea infusions originated from Iran, India and Sri Lanka was 5.05 μ g g⁻¹ (Salahinejad and Aflaki, 2010). The measured Cu content in loose tea and tea bag infusions of 40 samples from Norwegian supermarkets ranged from 1.4 to 80.7 $\mu \mathrm{g} \ \mathrm{L}^{-1}$ (mean: 28.1 \pm 15.7 $\mu \mathrm{g} \ \mathrm{L}^{-1}$) and 14.9 to 128 μ g L⁻¹ (mean: 70.3 \pm 29.2 μ g L⁻¹), respectively (Kronborg, 2013). However, measured Cu concentrations in 30 tea infusions from Polish supermarket ranged from 12.6 to 32.3 $\mu g L^{-1}$ (mean: 21.5 \pm 5.95 $\mu g L^{-1}$) and 23.9 to 50.3 $\mu g L^{-1}$ (mean: 35.5 \pm 8.83 $\mu g L^{-1}$) for loose tea and tea bag respectively (Kronborg, 2013).

Iron

Iron deficiency is associated with anemia, fatigue, and decreased work capability in older children, adult's prematurity, and perinatal mortality among pregnant women (Ahmed et al., 2014; Anand et al., 2014). According to WHO (2008), Fe deficiency is a global human health problem and accounted for anemia affecting 1.62 billion people worldwide. Recommended daily intake of dietary Fe for normal infants is 1 mg Fe $kg^{-1} d^{-1}$ and 10, 12, and 15 mg Fe kg⁻¹ d⁻¹ for children, male, and female adolescents, respectively. For women during reproductive years, 15 mg Fe kg⁻¹ d⁻¹ is recommended, while adult men and

Table 4. Concentration of copper in black (CTC and orthodox), green, Oolong, and Pu-reh tea infusion.

Type of made tea	Country of origin	Infusion condition*	Analytical instrument [†]	Concentration (μ g L $^{-1)}$	Reference(s) [‡]
Black	China	1	a	14.3–73.4	1
		2	b	0.13-0.22	2
		3	C	0.24-0.64	3
		4	b	0.164–0.26	4
	Range (for Chinese b	·	~	0.13-73.4	•
	Ethiopia	5	d	38–63	5
	India	2	b	0.08-0.18	2
	IIIuia	6	b	2.38–3.72	6
		4	b	2.36–3.72 0.072–0.441	4
	Damma (family diam blo		D		4
	Range (for Indian bla	•	L.	0.072–3.72	,
	Iran	6	b	3.9–9.0	6
	Kenya	4	b	0.159	4
	Russia	4	b	0.155	4
	Sri Lanka	7	d	0.126 ± 0.008	7
		8	d	3.34–20.12	8
		2	b	0.09-0.28	2
		6	b	4.35-8.63	6
		4	b	0.176-0.308	4
	Range (for Sri Lanka	n black tea)		0.09-20.12	
	Thailand	9	e	12.01–84.27	9
	Turkey	7	d	$0.102 \pm 0.009 - 0.143 \pm 0.015$	7
	runcy	10	f	$3.10 \pm 0.24 - 8.17 \pm 0.11$	10
		11		3.57–65.4	11
		4	e b	0.09	4
	D		D		4
	Range ((for Turkish k			0.09–65.4	42
	Unknown	12	e	16.7 ± 1.3	12
		13	d	4.28 ± 0.74	13
		4	b	0.149	4
	Range (for unknown	regions black tea)		0.15–18	
Overall range (for black tea)	0.0784.27				
Green	China	1	a	3–114	1
		14	d	$8.50 \pm 1.00 – 17.50 \pm 1.00$	14
		4	b	0.053-0.186	4
	Range (for Chinese of	reen tea)		0.05-114	
	India	4	b	0.084	4
	Indonesia	4	b	0.191	4
	Japan	4	b	0.173	4
	Sri Lanka	8	ď	6.54	8
	Turkey	7	d	0.108 ± 0.007	7
	Turkey	10	f	9.17 ± 0.007	10
	Damma (fan Tunkisk m		ı		10
	Range (for Turkish g			0.10–9.6	12
	Unknown	12	e	9.5 ± 0.8	12
		13	d	4.77 ± 0.35	13
		4	b	0.033-0.191	4
		15	g	$45 \pm 5 - 94.8 \pm 3.63$	15
	Range (for Unknown	regions green tea)		0.03-131.1	
	Vietnam	4	b	0.08	4
Overall range (for green tea)				0.03131.1	
Pu-erh	China	4	b	0.117	4
	2	16	b	0.035-0.051	16
					10
Overall range (for Pu-erh tea)		10	_	0.040.12	

^{*}In the column, the following bold digits denote the following points: 1, 1 g of tea sample (vegetal material) was placed inside a tea sachet (commercially available tea filter, made of unbleached paper). The sachet was introduced into a glass beaker, 100 mL of boiling ultrapure water were added and the beaker was allowed to stand for five minutes; 2, 5 g tea was added to 200 mL of boiling distilled water and allowed to infuse for two minutes; 3, 500 mL of boiled tap water and the Chinese tea (1 or 2 g) were added into the glass appliances and infused for 10 minutes; 4, 1 g tea was carefully weighed into glass beakers. Fifty milliliters of boiled distilled water was poured on it and covered with watch glasses for five minutes. Infusion was then filtered; 5, 2 q of tea leaves added in to the boiling water and boil for five minutes, then cooled for five minutes and filtered to 100 mL; 6, 45 mL hot distilled water was added to 1 g tea samples and infused for five minutes; 7: 1.00 g of tea infused for five minutes with 50 mL boiled distilled water; 8, 100 mL of hot distilled water was added to 2 g tea and infused for five minutes at room temperature; 9, 100 mL of boiling deionized water was added to one bag or 2.0 g of tea sample and left at room temperature for five minutes; 10, 100 mL of hot distilled water was added to 2 g of black tea sample. The mixture left to cool at room temperature for five minutes; 11, 200 mL of boiling ultrapure water was added in 2 g tea in a beaker and infused for 15 minutes; 12, 1.5 g tea was infused for three minutes in a cup with 200 mL boiling water and kept for three minutes covered with a watch glass; 13, About 80 mL of redistilled water was added to 3 g of made tea sample in glass beaker, and then heat up to boiling temperature for five minutes; 14, 2.5 g of tea +8 fluid oz boiling water, infused for three to six minutes; 15, 3 g of tea was transferred to the pots and 200 mL of boiled tap water was added and infused for 10 minutes; 16, 5.0 g of Pu-erh tea in 250 mL boiled water and infused for five minutes.

h the column, the following letters denote the following points: a, HR-CS FAAS (high resolution-continuum source flame atomic absorption spectrometry); b, ICP-AES (inductively coupled plasma atomic emission spectrometry); c, GFAAS (atomic absorption spectrometry spectrophotometry with graphite furnace); d, FAAS (flame atomic absorption spectrometry); e, ICP-MS (inductively coupled plasma-mass spectrometry); g, HR-SF-ICPMS (high-resolution sector field inductively coupled mass spectrometry.

in the column, the following digits denote the following references: 1, Paz-Rodríguez et al., 2015; 2, Mehra and Baker, 2007; 3, Ni and Li, 2008; 4, Street et al., 2006; 5, Gebretsadik and Chandravanshi, 2010; 6, Salahinejad and Aflaki, 2010; 7, Aksuner et al., 2012; 8, Lasheen et al., 2008; 9, Nookabkaew et al., 2006; 10, Görür et al., 2012; 11, Sofuoglu and Kavcar, 2008; 12, Milani et al., 2015; 13, Memić et al., 2014; 14, Gallaher et al., 2006; 15, Petit et al., 2013; 16, Lv et al., 2013a.

postmenopausal women require only 10 mg Fe kg^{$^{-1}$} d^{$^{-1}$} (Balch and Balch, 2000; Gupta and Gupta, 2014). The guideline value of Fe in drinking water is 0.3 mg L^{$^{-1}$} (WHO, 2003a, b), while tolerable upper intake level of Fe is reported as 45 mg day^{$^{-1}$} (FNB, 2001).

Iron in made tea

Table 5 provides Fe concentration in made tea available in selected literatures. From Table 5, it has been observed that Fe contents in Chinese, Ethiopian, Indian, Indonesian, Iranian, Kenyan, Pakistani, Russian, Saudi Arabian, Sri Lankan, Turkish, and unknown origins' black teas range from 6.5 to 13040, 29.6 to 880, 0.28 to 11672, 247 to 2172, 2.92 to 307.2, 10 to 329, 5.37 to 114, 303 to 6541, 46 to 513, 59.5 to 6463, 24.6 to 369.4, and 186 to 617 $\mu g g^{-1}$, respectively. The overall average of Fe content in black tea for all the countries mentioned in Table 5 ranges from 0.28 to 13,040 $\mu g g^{-1}$. For green tea, the ranges are from 1.49 to 13040, 124 to 321, 48 to 436, 125.4 to 162, 118.8 to 235.4, and 137 to 391 μ g Fe g⁻¹ for Chinese, Indonesian, Japanese, Sri Lankan, Turkish, and unknown origins' green teas respectively. The overall average of Fe content in green tea for all the countries mentioned in Table 5 ranges from 0.4 to 13,040 μg g⁻¹. For Oolong tea, the ranges are from 13.9 to 6751.5 and 0.7 to 202 μ g Fe g⁻¹ for Chinese and Taiwanese respectively. The overall averages of Fe content in Oolong, Puerh, and white teas for all the countries mentioned in Table 5 ranges from 0.70 to 6751.50, 187 to 2171, and 43 to 1129 μ g Fe g⁻¹, respectively. Iron contents of six Japanese green tea samples varied between 57.8 \pm 9.8 and 105 \pm 13 μ g g⁻¹ as reported by Islam and Ebihara (2012). Comparatively higher total Fe concentrations in 12 commercial bagged black teas from local stores in Wrocław, Poland had been reported by Dambiec et al. (2013) which varied between 120 \pm 1 and 765 \pm 10 μ g g⁻¹. This range compared well to Fe ranges of 88.7 to 947 $\mu g g^{-1}$ and 124 to 259 μg g⁻¹ reported by Ashraf and Mian (2008) and AL-Oud (2003), respectively. Among the analyzed eighteen tea samples (four black teas, four green teas, four Oolong teas, and six white teas), Fe contents varied widely and ranged from 52.2 ± 1.4 to $1551 \pm 23 \ \mu g \ g^{-1}$ (Malik et al., 2008). Total Fe contents in three made tea samples collected from a health food store in Gainesville, FL, USA ranged from 326 \pm 10 to $716 \pm 78 \ \mu g \ kg^{-1}$ (Gallaher et al., 2006). Desideri et al. (2011) reported that Fe in several kinds of tea marketed in Italy ranged from 99 to 617 μ g g⁻¹. In this connection, Desideri et al. (2011) concluded that the variation of Fe content among tea samples was mainly due to the differences in the degree of fermentation and the mineral composition of the soil where the plants were cultivated. Other factors responsible for a variation in Fe content are preferential absorbability of the plant, use of fertilizers, irrigation water, and climatic conditions (Desideri et al., 2011).

Iron levels in tea samples available in Kayseri market of Turkey were reported by Tokalıoğlu and Gürbüz (2010) as in the range of 58.6 \pm 1.2 to 276 \pm 5 μg g $^{-1}$. Iron contents in five types of teas, viz. white, green, black, Oolong, and Pu-erh ranged from 37 to 721 μg g $^{-1}$ (mean 176 μg g $^{-1}$), 99 to 647 μg g $^{-1}$ (mean 241 μg g $^{-1}$), 58 to 421 μg g $^{-1}$ (mean 99 μg g $^{-1}$), 187 to 2171 μg g $^{-1}$ (mean 376 μg g $^{-1}$), and 43 to 1129 μg g $^{-1}$ (mean 187 μg g $^{-1}$), respectively (McKenzie et al., 2010). Iron

contents of commercially available Ethiopian black tea (Wushwush, Gumero and Black Lion) were found to vary within 319-467 μ g g⁻¹ (Gebretsadik and Chandravanshi, 2010). Moreda-Piñeiro et al. (2003) reported that iron contents of African and Asian teas ranged from 153 to 178.97 μ g g⁻¹. Levels of Fe in unprocessed tea samples grown and marketed in Kenya were reported to vary widely between 55 and 203 μ g g⁻¹, whereas, Fe contents in black tea samples from the same location varied between 118 and 329 μ g g⁻¹ (Moseti et al., 2013). On the other hand, Fe levels were found to range from 151 to 369 μ g g⁻¹ in the black tea samples from the Mombasa tea auction center (Moseti et al., 2013). Black tea from Uganda, Rwanda, and Tanzania were reported to contain 344, 262, and 195 μ g g⁻¹ Fe, respectively, with differences attributed to variation in soil characteristics in the different tea-growing regions (Moseti et al., 2013). Furthermore, Moseti et al. (2013) concluded that Fe content in Kenyan unprocessed tea was lower compared to that in Kenyan black tea from the same catchment area, an observation that suggested a possible introduction of additional Fe into the black tea during the manufacturing process possibly as iron fillings. Iron contents in three brands of Ethiopian black tea (Wushwush, Gumero, and Black Lion) as reported by Woldegebriel (2007) varied between 319 \pm 25 and 467 \pm 2 μ g g⁻¹. This wide variations of Fe content in black tea depended upon the country or area of origin, soil mineral content, and the age and part of the tea plant analyzed. The concentration of Fe in 30 samples of black tea cultivated in the north of Iran was compared with the results for 30 samples of imported black tea collected in the year 2006 by Ansari, et al. (2007). The results revealed that the measured concentration of Fe in the Iranian black tea varied between 17.2 and 194.0 μ g g⁻¹ (mean: 92.6 μ g g⁻¹), while that of the imported tea samples varied between 17.6 and 322.8 μ g g⁻¹ (mean: 73.0 μ g g⁻¹). Nookabkaew et al. (2006) analyzed 18 tea samples collected from different supermarkets in Bangkok, Thailand (4 samples produced in Thailand and 14 samples imported from China, Japan, Sri Lanka, India, and Indonesia) where Fe content ranged from 20.91 to 318.3 $\mu g g^{-1}$ with the mean of 167.1 $\mu g g^{-1}$. Zhu et al. (2011) reported that Fe contents in green tea samples collected from Japan were 81.6 \pm 1.2 to 82.4 \pm 0.3 μg g⁻¹. Ahmad et al. (2012) analyzed Fe content in 12 black tea samples commonly consumed in Pakistan and reported that it varied between 5.47 and 11.63 μ g g⁻¹ with the mean of 7.55 μ g g⁻¹. Total Fe content in 16 tea samples in Addis Ababa, Ethiopia ranged from 286.4 to 880.9 μ g g⁻¹ (Ashenef, 2014). Yemane et al. (2008) reported a range of 18.8-23.5 μ g g⁻¹ of Fe in Ethiopian tea clones, which was comparatively lower than the three Ethiopian commercial tea brands (319–467 μ g g⁻¹) reported by Woldegebriel et al. (2012). A summary of total Fe contents in black teas tabulated by Wróbel et al. (2000) suggested that Fe in tea ranged from 170 to 200 μg g⁻¹. Among 48 commercial tea samples (15 green tea, 15 black tea, and 18 Oolong tea), Fe contents in green tea, black tea and Oolong tea were 0.4–1.2 μg g⁻¹ (mean: $0.6~\mu g~g^{-1}$), 0.7– $1.2~\mu g~g^{-1}$ (mean: $0.9~\mu g~g^{-1}$), and 0.7– $1.3~\mu g~g^{-1}$ (mean: $0.9~\mu g~g^{-1}$), respectively (Shen and Chen, 2008). Mose et al. (2014) reported that the Fe content in black tea samples collected from three agroecological areas (Murang'a, Meru and Kisii) in Kenya varied between 136 \pm 8

and 320 \pm 5 μ g g⁻¹. Significantly higher amount of Fe in black

Table 5. Iron concentration in different types of made tea.

Type of made tea	Country of origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(s)
Black	Africa (Burundi, Ethiopia, Kenya, Malawi, Tanzania, and Zimbabwe)	a	178.97 ± 147.71	1
	Argentina	b	5801	2
	Argentina/Vietnam	b	267 ± 12	3
	Asia (India and Sri Lanka)	a	102.00 ± 16.33	1
	Bangladesh	b	9.0-10.0	4
	Brazil	b	7.5–8.5	4
	China	a	162–523	5
		b	$120 \pm 1 - 204 \pm 3$	3
		b	4362–11 577	2
		b	4362–13040	2
		a	37–721	6
		b	6.5–13.5	4
		C	139–464	7
	Range (for Chinese black tea)	C	6.5–13040	,
		d		o
	England	d	117.9–182.9	8
	Ethiopia	C	286.4–880	9
		b	$319 \pm 25 – 467 \pm 2$	10
		b	29.6–100	11
	Range (for Ethiopian black tea)		29.6–880	
	India	a	136–368	5
		b	0.275-0.40	12
		b	$135 \pm 7 – 144 \pm 5$	3
		b	2367-11 672	2
		b	124.7-139.0	13
		a	111.5-212.2	14
		b	4.0–10.5	4
		C	243	7
	Range (for Indian black tea)	C	0.28–11672	,
	Indonesia	b	277 ± 30	3
	liuoliesia			15
		e	344 ± 27.5	
	2 (6 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	b	2172	2
	Range (for Indonesian black tea)		247–2172	
	Iran	b	92.6 ± 89.68	16
		b	21.0–42.0	17
		a	105.55-307.2	14
	Range (for Iranian black tea)		2.92-307.2	
	Kenya	a	227	5
		d	231	8
		b	118–329	18
		b	169.3 ± 129.33	19
		b	51.04–116.71	19
		b	10–12	4
	Range (for Kenyan black tea)	2	10–329	•
	Kenya/Malawi	b	122 ± 3	3
	Myanmar	b	6828	2
	Pakistan		$5.6 \pm 0.23 - 29.2 \pm 1.8$	20
	Fakistali	c b		
	Danier (fan Daldstaniklandstan)	D	110.9–114	21
	Range (for Pakistani black tea)		5.37–114	_
	Russia	a	303	5
		b	6541	2
		b	6541	2
	Range (for Russian black tea)		303–6541	
	Rwanda	b	262	18
	Saudi Arabia	b	46-348	22
		b	123.9-513.0	23
	Range (for Saudi Arabian black tea)		46-513	
	Singapore	f	135.5	24
	Sri Lanka	b	198	25
		a	103–309	5
		b	$228 \pm 7 - 765 \pm 10$	3
			99	26
		g d	59.5–2478	8
		b	6463	2
		b	77.48–160.75	27
		a	94.93–310.1	14
		b	103–328	7
	Range (for Sri Lankan black tea)	_	59.5–6463	
	Syria	b	7940–11 250	2
	Taiwan	h	0.7-1.2	28
	Tanzania	b	195	18
			20.91-318.3	29
	Thailand	h	20.91-310.3	23

Table 5. (Continued)

Type of made tea	Country of origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(s)
		a	202	5
		h	24.6–32.3	30
		d	$128.57 \pm 0.55 - 368.85 \pm 0.59$	31
		h	243 ± 4.5	32
	Range (for Turkish black tea)		24.6–369.4	
	Uganda	b	344	18
	United Arab Emirates	d	688.8	8
	Unknown	a	314	5
		b	258.76 ± 2.96	33
		g	186–617	26
	2 (b	$427 \pm 38 - 485 \pm 23$	34
	Range (for unknown origin black tea)		186–617	
verall range (for			0.2813040	_
Green	China	a	140–378	5
		į.	$1.60 \pm 0.11 - 4.52 \pm 0.16$	35
		b	350 ± 14.8	36
		d	229.1	8
		f	74.02	24
		b	13040	2
		b	$326 \pm 10 – 716 \pm 78$	37
		a	99–647	6
	Range (for Chinese green tea)		1.49–13040	
	India	a	342	5
	Indonesia	a	200	5
		e	135 ± 11.0	15
		g	173–321	26
	Range (for Indonesian green tea)		124–321	
	Japan	a	436	5
		j	57.8 \pm 9.8–105 \pm 13	38
	Range (for Japanese green tea)		48-436	
	South Africa	d	1229.5	8
	South Korea	d	346.3	8
	Sri Lanka	g	162	26
		d	125.4	8
		b	156.80	27
	Range (for Sri Lankan green tea)		125.4–162	
	Taiwan	h	0.4–1.2	28
	Turkey	d	119.21 ± 0.40	31
		h	231 ± 4.4	32
	Range (for Turkish green tea)		118.8-235.4	
	Unknown	a	137–391	5
		b	180.38 ± 10.13	33
		b	$252 \pm 30 – 355 \pm 28$	34
	Range (for unknown origin green tea)		137–391	
	Vietnam	a	233	5
	Overall range (for world green tea)		0.413040	
olong	China	f	13.9	24
		b	6751.5	2
		a	58-421	6
	Range (for Chinese Oolong tea)		13.9-6751.5	
	Taiwan	h	0.7-1.3	28
		a	182–202	5
	Range (for Taiwanese Oolong tea)		0.7-202	
	Unknown	b	320.04 ± 11.40	33
overall range (for	world Oolong tea)		0.706751.50	
u-erh	China	a	405	5
		a	187–2171	6
verall range (for	Chinese Pu-erh tea)	u	1872171	Ŭ
Vhite	China	a	210	5
***************************************	Ciliid	a	43–1129	6
		а	コ ンー 1 1 4 2	U

^{*}In the column, the following letters denote the following points: a, ICP-AES (inductively coupled plasma atomic emission spectrometry); b, FAAS (flame atomic absorption spectrometry); c, ETAAS (electrothermal atomic absorption spectrometry); d, ICP-OES (inductively coupled plasma optical emission spectrometry); e, ICP-QMS (inductively coupled plasma, quadrupole mass spectrometry); f, Capillary electrophoresis; g, polarized X-ray fluorescence spectrometry; h, ICP-MS (inductively coupled plasma-mass spectrometry); i, X-ray fluorescence; j, INAA (instrumental neutron activation analysis).

th the column, the following digits denote the following references: 1, Moreda-Piñeiro et al., 2003; 2, Ferrara et al., 2001; 3, Dambiec et al., 2013; 4, Yasmeen et al., 2000; 5, Street et al., 2006; 6, McKenzie et al., 2010; 7, Polechońska et al., 2015; 8, Donkora et al., 2015; 9, Ashenef, 2014; 10, Gebretsadik and Chandravanshi, 2010; 11, Yemane et al., 2008; 12, Chand et al., 2011; 13, Marbaniang et al., 2011; 14, Salahinejad and Aflaki, 2010; 15, Pekal et al., 2013; 16, Ansari et al., 2009; 17, Falahi and Hedaiati, 2013; 18, Moseti et al., 2013; 19, Omwoyo et al., 2013; 20, Jalbani et al., 2007; 21, Jabeen et al., 2015; 22, Al-Othman et al., 2012; 23, AL-Oud, 2003; 24, Feng et al., 2003; 25, Aksuner et al., 2012; 26, Desideri et al., 2011; 27, Lasheen et al., 2008; 28, Shen and Chen, 2008; 29, Nookabkaew et al., 2006; 30, Alasalvar et al., 2013; 31, Görür et al., 2011; 33, Dawodu et al., 2013; 34, Memić et al, 2014; 35, Alassane et al., 2013; 36, Brzezicha et al., 2014; 37, Gallaher et al., 2006; 38, Islam and Ebihara, 2012.

tea collected from Kisii agroecological zone was reported, which was probably due to the higher Fe content in matured leaves. This was attributed to coarse plucking standards and long plucking intervals among other factors, which justified a strong need for strict adherence to good agricultural practices as well as good manufacturing practices in the tea industry.

The content of Fe in seven tea samples collected from Egypt local market ranged from 77.48 to 302 μ g g⁻¹ (Lasheen et al., 2008). Malik et al. (2008) analyzed the Fe content in 15 types of pure mixture of tea leaves that originated from different countries and obtained a range of 52.2 \pm 1.4 to 1551 \pm 23 μ g g⁻¹. Al-Othman et al. (2012) analyzed 10 black tea samples available from Jazan and Jeddah region of Saudi Arabia and found Fe concentration to be the predominant micronutrient in the tea samples analyzed, which varied between 46 and 348 μ g g⁻¹. Earlier, AL-Oud (2003) reported that Fe level ranged from 123.9 to 513.3 μ g g⁻¹ in tea samples from Saudi Arabia. Iron contents in four black tea samples in Nigeria ranged from 0.99 ± 0.10 to $2.39 \pm 0.22 \ \mu g \ g^{-1}$ with the mean of 1.65 \pm $0.16~\mu g~g^{-1}$ (Achudume and Owoeye; 2010). Chand et al. (2011) reported that Fe contents in made tea ranged from $0.275 \text{ to } 0.40 \ \mu\text{g g}^{-1}$.

Dawodu et al. (2013) reported that Fe content in green tea, black tea, and Oolong tea samples available in Nigeria were 180.38 ± 10.13 , 258.76 ± 2.96 , and $320.04 \pm 11.40 \ \mu g \ g^{-1}$, respectively. Görür et al. (2011) reported that Fe content in 29 marked brands of black tea and one type of green tea commonly consumed in Turkey was the second highest element found in the tea samples; and ranged from 119.21 \pm 0.40 to $368.85 \pm 0.59 \text{ mg g}^{-1}$ (mean: $193.69 \pm 0.51 \text{ mg g}^{-1}$). Salahinejad and Aflaki (2010) reported that Fe content in 11 black tea samples available in Iran ranged from 105 to 407 μ g g⁻¹, while Chen et al. (2009) indicated that the Fe content in made tea produced from eight tea cultivars in China ranged from 80.53 to 114.6 μ g g⁻¹. Iron contents in three high-quality black tea and four low-quality black tea grades available in Turkey ranged from 24.26 \pm 1.14 to 31.41 \pm 1.10 mg per 100 g of tea and 16.75 \pm 0.52 to 48.74 \pm 3.50 mg per 100 g of tea, respectively (Serpen et al., 2012). Iron contents in commercially available Oolong tea from Taiwan were 242 \pm 18 and 232 \pm $14 \mu g g^{-1}$, respectively (Mierzwa et al., 1998).

Iron in tea infusion

Iron in tea infusion is presented in Table 6. From Table 6, it is observed that the Fe contents in Chinese, Indian, Iranian, Pakistani, Sri Lankan, Turkish, and unknown origin's black tea infusion ranges from 0.06 to 57, 0.04 to 5.48, 2.86 to 6.41, 1 to 7.4, 0.05 to 122.45, 0.05 to 3.23, and 0.14 to 5.15 mg L^{-1} , respectively. The overall average of Fe content in black tea infusion for all the countries mentioned in Table 6 ranges from 0.04 to 216.2 mg L⁻¹. From 0.02 to 245.8, 0.08 to 83.88, and 0.04 to 9.8 mg L⁻¹ Fe were found in Chinese, Turkish, and green tea originated from unknown regions respectively with the overall range from 0.02 to 245.80 mg L^{-1} . The overall averages of Fe contents in Oolong, Pu-erh, and white tea infusions range from 0.04 to 0.07, 0.101, and 0.037 mg L^{-1} , respectively. Iron contents in 18 tea infusions ranged from 0.005 \pm 0.0023 to 0.100 ± 0.008 mg L⁻¹, when tea infusion was prepared from

1 g made tea with 50 mL boiled distilled water for 15 minutes (Malik et al., 2008). The mean Fe content in infusion of tea available in Kayseri market of Turkey was 2.79 μg g⁻¹ (Tokalıoğlu and Gürbüz, 2010). Kim et al. (2004) reported Fe content in black tea infusion (U.S. Tea Association, Black Tea Research Blend, Thomas J. Lipton Co., Englewood Cliffs, NJ, USA) was $30.7 \pm 3.1 \text{ g kg}^{-1}$ (Kim et al., 2004). The contents of Fe in black tea infusion ranged from 0.98 to 1.56 mg L^{-1} (Gebretsadik and Chandravanshi, 2010). Nookabkaew et al. (2006) reported that the concentration of Fe in 18 black tea infusions ranged from 4.44 to 21.62 µg 100 mL⁻¹ with the mean of 7.99 μ g 100 mL⁻¹, which represented 2.39% of total Fe in the black tea. Wróbel et al. (2000) reported that only 4.3 and 8.1% of total Fe can be infused from black and green tea, respectively despite the high total Fe content in the samples. The mean Fe contents in green tea, black tea, and Oolong tea infusion in Taiwan were 0.13, 0.5, and 0.7 μ g 100 mL⁻¹, respectively, which were equivalent to 10.9, 30.9, and 42.3% of total Fe contents, respectively (Shen and Chen, 2008). Among the seven tea samples from Egypt, Fe content in tea infusion ranged from 67.54 to 122.45 μ g g⁻¹ (Lasheen et al., 2008). Iron content in tea infusion prepared from 1 g of tea infused with 50 mL boiled distilled water and infused from 15 minutes for 15 tea samples originated from different tea growing countries ranged from 0.005 ± 0.002 to 0.100 ± 0.008 mg L⁻¹ (Malik et al., 2008). The amounts of Fe in four Nigerian black tea infusions prepared from 1 g of tea boiled in 50 mL of distilled water for 10 minutes were between 442.95 \pm 8.25 and 1716.6 \pm 186.15 μ g L⁻¹ with mean value of 975.68 \pm 95.16 μ g L⁻¹ as reported by Achudume and Owoeye (2010). Iron levels in nine green tea infusions varied between 0.020 \pm 0.001 and 0.128 \pm 0.007 mg L^{-1} as reported by Reto et al. (2007). Fernández et al. (2002) reported that tea contained only trace amounts of Fe, with a range of 0.02-0.17 mg L⁻¹ for green tea and 0.03-0.23 mg L⁻¹ for black tea. Contrarily, Chand et al. (2011) reported a fairly elevated contents that ranged from 0.068 to $0.085 \text{ mg Fe L}^{-1}$ in black tea. Szymczycha-Madeja et al. (2012) described Fe content in tea infusion as a poorly extractable element representing less that 20% of the total Fe content. Gallaher et al. (2006) also noted that Fe extraction rate in a tea infusion was lower than 10%. However, AL-Oud (2003) reported that Fe concentration in tea beverage ranged from 19.0 to 56.0 mg L^{-1} . The low percentage solubility of Fe revealed that Fe was in the form of the least water-soluble complexes in tea (Soomro et al., 2008).

Among the five black tea and green tea samples in Bosnia and Herzegovina, Fe contents in tea infusion ranged from 4.79 ± 0.36 to $9.51 \pm 0.45~\mu g~g^{-1}$ (Memić et al., 2014). Iron contents in Turkish tea infusion and tea brew ranged from BDL \pm to 3.15 \pm 1.14 mg g⁻¹ (mean: 1.07 \pm 0.70 mg g⁻¹) and from 2.30 \pm 1.18 to 14.78 \pm 0.48 mg g $^{-1}$ (mean: 7.75 \pm 1.0 mg g^{-1}), respectively, as reported by Görür et al. (2011). Average Fe content in 11 tea infusions originated from Iran, India and Sri Lanka was 5.18 μ g g⁻¹ (Salahinejad and Aflaki, 2010). The contents of Fe in loose teas ranged from 2.77 to 15.1 μ g L⁻¹ (mean: 7.34 \pm 4.17 μ g L⁻¹) in the Poland tea samples and from 2.79 to 51.7 μ g L⁻¹ (mean: 13.8 \pm 10.3 μ g L⁻¹) in the Norway tea samples (Kronborg, 2013). However, comparatively higher amount of Fe was extracted from the tea bags than

the loose tea samples for Poland and Norway, which ranged from 3.9 to 32.9 μ g L⁻¹ (mean: 8.53 \pm 0.71 μ g L⁻¹) and 5.73–73 μ g L⁻¹ (mean: 29.8 \pm 21. μ g L⁻¹), respectively (Kronborg, 2013).

Manganese

Notwithstanding the essentiality of manganese (Mn) to human body, its higher dose may result in Mn toxicity (U.S Centers for Disease Control, ATSDR, 2000). However, it plays an important role in bone and cartilage development (Leach, 1988;

Saltman and Strause, 1993); and wound healing through stimulatory and antioxidative activity (Marrotte et al., 2010).

Manganese in made tea

From Table 7, it has been observed that Mn contents in Chinese, Ethiopian, Indian, Indonesian, Iranian, Kenyan, Saudi Arabian, Sri Lankan, Turkish, and unknown origins' black teas range from 160 to 7934, 8.6 to 1425, 0.28 to 1586.6, 231 to 4679, 59.5 to 731, 180 to 2072, 48 to 900, 194.2 to 1441.4, 0.35 to 2121, and 113.39 to 1557 $\mu g g^{-1}$, respectively. The overall average of Mn content in black tea in all the countries mentioned in Table 7

Table 6. Concentration of iron in black, green, Oolong, Pu-erh, and white tea infusion.

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L ⁻¹)	Reference(s
Black	Argentina/Vietnam	1	a	0.425 ± 0.106	1
	China	2	b	25.8-57	2
		3	C	0.055-0.131	3
		4	d	3.03–4.82	4
	Range (for Chinese blac		u	0.06–57	7
			_		-
	Ethiopia	5	a	0.98–1.56	5
	India	6	a	0.068-0.085	6
		1	a	0.200 ± 0.071	1
		7	C	3.34–5.48	7
		3	C	0.038-0.084	3
		4	d	4.02	4
	Range (for Indian black	tea)		0.04-5.48	
	Iran	7	C	5.41–6.41	7
	nan	8	d	3.11 ± 0.25	8
	D(f!t)	0	u		O
	Range (for Iranian tea)	_		2.86–6.41	_
	Kenya	3	C	0.073	3
	Pakistan	9	d	1.1 ± 0.1 – 6.8 ± 0.6	9
	Range (for Pakistani bla	ck tea)		1–7.4	
	Russia	3	С	0.105	3
	Sri Lanka	10	a	0.291 ± 0.013	10
	5.1. Zuu	1	a	1.40 ± 0.071	1
		11		30.56–122.45	11
			a		
		7	С	2.27–5.98	7
		3	С	0.051-0.069	3
		4	d	1.82–7.18	4
	Range (for Sri Lankan b	ack tea)		0.05-122.45	
	Thailand	12	e	44.44-216.2	12
	Turkey	11	a	$0.240 \pm 0.019 – 0.460 \pm 0.020$	10
	· u.n.ey	13	f	2.08 ± 1.15	13
		3	C	0.049	3
	D		C		3
	Range (for Turkish black			0.05–3.23	
	Unknown	14	e	2.2	14
		15	a	4.79 ± 0.36	15
		3	С	0.142	3
	Range (for unknown ori	gins black tea)		0.14-5.15	
Overall range (for blac		,		0.04216.2	
Green	China	16	a	$161 \pm 25.4 - 203.4 \pm 42.4$	16
Siccii	Cima	2	b	21.7-74.4	2
		3		0.045-0.099	3
			С		
		. 17	a	0.020-0.128	17
	Range (for Chinese gree			0.02-245.8	
	India	3	C	0.088	3
	Indonesia	3	С	0.083	3
	Japan	3	C	0.142	3
	Sri Lanka	11	a	83.88	11
	Turkey	10	a	0.270 ± 0.012	10
	Turkey		a f		
	D (6 T 1::	13	I	3.15 ± 1.14	13
	Range (for Turkish gree			0.08-83.88	
	Unknown	14	e	9.5 ± 0.3	14
		15	a	4.77 ± 0.35	15
		3	С	0.038-0.069	3
	Range (for unknown ori	gins green tea)		0.04-9.8	

(Continued on next page)

Table 6. (Continued)

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L^{-1})	Reference(s) [‡]
Overall range (for gree	n tea)			0.02245.80	
Oolong	Taiwan	3	С	0.037-0.074	3
		18	С	0.035-0.062	3
		19	С	0.048-0.054	3
Overall range (for Oolo	ng tea)			0.040.07	
Pu-erh	China	3	С	0.101	3
White	China	3	c	0.037	3

*In the column, the following bold digits denote the following points: 1, 2.0 g of tea leaves +80 mL of deionized boiling water, covered and steeped for exactly for five minutes; 2, 1 g of tea sample was placed inside a tea sachet (commercially available tea filter, made of unbleached paper). The sachet was introduced into a glass beaker, 100 mL of boiling ultrapure water was added and the beaker was allowed to stand for five minutes; 3, 1 g tea samples were carefully weighed into glass beakers. Fifty milliliters of boiled distilled water was poured on it and covered with watch glasses for five minutes. Infusion was then filtered; 4, 80 cm³ of boiling deionized water to 2.0 g of broken tea leaves and infused for five minutes; 5, 2 g of tea leaves added in to the boiling water and boil for five minutes, then cooled for five minutes and filtered to 100 mL; 6, not available; 7, 45 mL hot distilled water to 1 g tea sample and five minutes infusion; 8, 50 mL of boiling deionized water was added to 5 g tea and kept at 80°C on a water bath for 15 minutes. At the end of the infusion period, the extract was filtered; 9, boiling 100 mL of deionized water and pouring the boiling water over duplicate (2.0 g) tea sample; 10, 1.00 g of tea infused for five minutes with 50 mL boiled distilled water; 11, 100 mL of hot distilled water was added to 2 g tea and infused for five minutes at room temperature; 12, 100 mL of boiling deionized water was added to one bag or 2.0 g of tea sample and left at room temperature for five minutes; 13, 100 mL of hot distilled water was added to 2 g of black tea sample. The mixture left to cool at room temperature for five minutes; 14, 1.5 g tea was infused for three minutes in a cup with 200 mL boiling water and kept for three minutes covered with a watch glass; 15, 80 mL of redistilled water was added to 3 g of made tea sample in the beaker. The sample was heated for more five minutes and filtered after cooling; 16, 2.5 g tea +8 fluid oz boiling water, steeping time three to six minutes; 17, 1.

th the column, the following letters denote the following points: a, FAAS (flame atomic absorption spectrometry); b, HR-CS FAAS (high-resolution-continuum source flame atomic absorption spectrometry); c, ICP-AES (inductively coupled plasma atomic emission spectrometry); d, ETAAS (electrothermal atomic absorption spectrometry); e, ICP-MS (inductively coupled plasma-mass spectrometry); f, ICP-OES (inductively coupled plasma optical emission spectrometry).

in the column, the following digits denote the following references: 1, Dambiec et al., 2013; 2, Paz-Rodríguez et al., 2015; 3, Street et al., 2006; 4, Polechońska et al., 2015; 5, Gebretsadik and Chandravanshi, 2010; 6, Chand et al., 2011; 7, Salahinejad and Aflaki, 2010; 8, Ziarati et al., 2013; 9, Jalbani et al., 2007; 10, Aksuner et al., 2012; 11, Lasheen et al., 2008; 12, Nookabkaew et al., 2006; 13, Görür et al., 2012; 14, Milani et al., 2015; 15, Memić et al., 2014; 16, Gallaher et al., 2006; 17, Reto et al., 2007.

ranges from 0.28 to 7934 $\,\mu\mathrm{g}$ $\mathrm{g}^{-1}.$ For green tea, the ranges are from 7.08 to 3358.26, 0.55 to 808, 542 to 918, 517 to 983, 151.3 to 405, 73.3 to 1120.91, and 104.94 to 1584 μ g Mn g⁻¹ for Chinese, Indian, Indonesian, Japanese, Sri Lankan, Turkish, and unknown origins', respectively. The overall average of Mn content in green tea for all the countries mentioned in Table 7 ranges from 0.55 to 3358.26 μ g g⁻¹. For Oolong tea, the ranges are from 132.7 to 2368 and 101.95 to 465 μ g Mn g⁻¹ for Chinese and unknown origins', respectively. The overall average of Mn content in Oolong tea for all the countries mentioned in Table 7 ranges from 101.95 to 2368 $\mu g g^{-1}$. The overall averages of Mn content in Pu-erh and white teas for all the countries mentioned in Table 7 range from 615 to 1268 and 337 to 1463 μ g g⁻¹, respectively. Kronborg (2013) reported that tea leaves contained 350–900 μ g Mn g⁻¹, which is an essential element for plants, microorganisms and higher animals including man. Mn contains in a total of 45 samples of different teas commercialized in Spain have been ranged from 76.1 to 987.6 μ g g⁻¹ (Cabrera et al., 2003). Manganese concentration in the tea leaves of US brands that were derived from India, Sri Lanka, and China, including herbal infusions with several flavoring additives, were found to vary widely with the lowest (79 μ g g⁻¹) and highest (768 μ g g⁻¹) values being for Refresh and Zen brands, respectively (Kronborg, 2013). On the other hand, Mn content in various Indian tea brands is in a much narrower range of 371–758 μg g^{-1} with a mean value of 575 \pm 96 μg g^{-1} (Kumar et al., 2005). Han et al. (2007) established an on-line sample digestion method for tea samples with higher heating efficiency and heating rate by employing electromagnetic heating technique by furnace atomic absorption spectrometry (FAAS). They concluded that the proposed method was in good agreement with the results obtained by using off-line digestion. In this technique, the concentrations of Mn in tea leaf samples were found to vary between 725 and

1249 μ g g⁻¹ (Han et al., 2007). Data of Mn in 29 marked brands of black tea and one type of green tea commonly consumed in Turkey has been reported by Görür et al. (2011) and the authors found that levels of Mn were in the range of 292.65-1850.75 mg g^{-1} (mean:1286.35 \pm 0.58 mg g^{-1}). Yasmeen et al. (2000) have reported 175 mg g⁻¹ Mn content in tea samples from Pakistan. The average level of Mn in black tea samples from Saudi Arabia was 750.9 mg g⁻¹ (Ashraf and Atiq, 2008). Manganese content was the most abundant nutritive metal among other analyzed elements in 30 samples of black tea cultivated in the north Iran and 30 imported black tea samples with 155.2-214.2 and 96.7-332.9 μ g g⁻¹, respectively (Ansari et al., 2007). In all the cases, the average contents of detectable heavy metals were significantly (P < 0.05) higher in Iranian black tea. The wide variations Mn content might be due to the different agroclimatic regions as well. Mn were found in five tea leave samples through off-line digestion and online electromagnetic heating technique 757 \pm 10 to 1233 \pm 18 μ g g⁻¹ and 740 \pm 15 to 1225 \pm 24 μ g g⁻¹, respectively (Han et al., 2007).

Amongst 18 tea samples, namely, 4 samples produced in Thailand and in 14 samples imported from China, Japan, Sri Lanka, India, and Indonesia, Mn concentration were ranged from 229.4–1512 $\mu g g^{-1}$ with the mean of 813.6 $\mu g g^{-1}$ (Nookabkaew et al., 2006). Similarly, Mn in five commercial tea samples collected from Shillong, Meghalaya, India were ranged from 111.4 to 143.8 $\mu g g^{-1}$ with a mean value of 129.5 \pm 14.0 $\mu g g^{-1}$ (Marbaniang et al., 2011). Yasmeen et al. (2000) have reported 175 $\mu g g^{-1}$ Mn concentrations in black tea samples from Pakistan. Naithani and Kakkar (2005) in their study of metallic content in black tea samples in South India reported a mean Mn concentration of 140 \pm 5.29 $\mu g g^{-1}$. Zhu et al. (2011) reported that Mn content in Japanese green teas ranged from 760 \pm 2 to 761 \pm 8 $\mu g g^{-1}$. Ahmad et al. (2012) analyzed

Mn in 12 black tea samples commonly consumed in Pakistan and reported concentration between 1.27 and 3.72 $\mu g \ g^{-1}$ with the mean of 2.06 $\mu g \ g^{-1}$ when FAAS was used. Measured Mn contents in 16 tea samples from Addis Ababa, Ethiopia ranged from 81.7–962.2 $\mu g \ g^{-1}$ (Ashenef, 2014). Yemane et al. (2008) reported a range of 1030–1470 $\mu g \ g^{-1}$ of Mn for Ethiopian tea clones collected at a farm site while Woldegebriel et al. (2012) similarly reported a range of 1242–1421 $\mu g \ g^{-1}$ of Mn in three Ethiopian tea brands.

The content of Mn measured in seven tea samples collected from local market of Egypt ranged from 405–715 $\mu g g^{-1}$ (Lasheen et al., 2008). Malik et al. (2008) analyzed Mn in 15 types of pure mixture of tea leaves that was originated from different countries and reported that it ranged from 211 \pm 15 to 1198 \pm 189 $\mu g g^{-1}$. Al-Othman et al. (2012) reported that Mn is the

second most abundant element in the analyzed 10 black tea samples obtained from Jazan and Jeddah region of Saudi Arabia with a range of 48–859 μg g⁻¹. Yemane et al. (2008) reported that Mn levels in tea leaf samples obtained from Ethiopia were in the range of 501–1281 μg g⁻¹. The Mn level in tea samples from the Kingdom of Saudi Arabia were varied from 390 to 900 μg g⁻¹ (AL-Oud, 2003). Gebretsadik and Chandravanshi (2010) reported a Mn concentration range in black tea samples from Ethiopia as 1242–1421 μg g⁻¹. Korkmaz et al. (2011) considered Mn content as an important element in tea leaves and reported a range of 292.65–1850.75 μg g⁻¹.

Among 43 representative tea products including 18 green, 12 Oolong, and 13 black teas, detected Mn level were ranged from 132.70 to 2399.64 μ g g⁻¹ in all the tested samples, with the range of 181.95 to 2399.64 μ g g⁻¹ in green teas, 132.70 to

Table 7. Manganese in different types of made teas produced the leaf of Camellia sinensis L.

Type of made tea	Country of origin/consumed/marketed	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(
	Africa (Burundi, Ethiopia, Kenya, Malawi, Tanzania, and Zimbabwe)	a	562.61 ± 316.14	1
	Argentina	b	5685	2
A A	Argentina/Vietnam	b	652 ± 69	3
	Asia (India and Sri Lanka)	a	367.26 ± 138.15	1
	Bangladesh	b	132–135	4
	Brazil	b	296-290	4
	China	b	184.69-1182.15	5
		a	572-864	6
		b	$479 \pm 8 – 746 \pm 26$	3
		b	7512–7934	2
		a	190–1570	7
		a	582–1141	8
		b	160–208	4
			565–1169	9
	Danne (for Chinese black too)	С		9
	Range (for Chinese black tea)		160–7934	10
	England	d	206.6–628.8	10
	Ethiopia	C	8.6–198.3	11
		b	$1242 \pm 12 - 1421 \pm 4$	12
		b	501–1281	13
	Range (for Ethiopian black tea)		8.6-1425	
	India	a	630–974	6
		b	$627 \pm 25 – 935 \pm 36$	3
		b	1265.8-1586.6	2
		e	331(226)-565(140)	14
		b	371–758	14
		b	111.4–143.8	15
		a	300–549	8
		a	641(331)	8
			436.2–549.9	16
		a b		17
			709.0 (14.18)	
		b	176–205	4
		C	648	9
		b	0.286 (0.008)-1.29 (0.01)	18
	Range (for Indian black tea)		0.28-1586.6	
	Indonesia	b	924 ± 39	3
		f	251 ± 20	19
		b	4679	2
	Range (for Indonesian black tea)		231–4679	
	Iran	b	682–731	20
		b	182.9 ± 123.4	21
		a	439.9-674.3	16
	Range (for Iranian black tea)		59.5-731	
	Japan	g	767 ± 6	22
	Kenya	a	2072	6
	····· <i>y</i>	d	408.2	10
		b	666.3 ± 121.67	23
		b	630.13-704.17	23
		b	185–180	4
			770 ± 30	
	Dan ma (fau Manuan Inlant taa)	b		3
	Range (for Kenyan black tea)	L	180–2072	2
	Myanmar	b	1146.4	2

 Table 7. (Continued)

pe of made tea	Country of origin/consumed/marketed	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(
	Pakistan	b	165–185	24
	Russia	a	1675	6
	Russia	b	1527.7	2
	Saudi Arabia	b	48-859	25
		b	390-900	26
	Range (for Saudi Arabian black tea)		48-900	
	Sri Lanka	a	614–919	6
		b	1042	27
		b	$408 \pm 8 - 647 \pm 79$	3
		h	213	28
		d	194.2–866.2	10
		b	1441.4	2
		b	354–715	29
		a	409–640	8
			333.9–655.4	16
		a		9
	Danne (for Cri Lankon blask too)	C	313–891	9
	Range (for Sri Lankan black tea)		194.2–1441.4	20
	Thailand	g	229.4–1512	30
	Turkey	a	1273	6
		b	563.9–1081.6	31
		b	$1640 \pm 43 - 1674 \pm 37$	27
		g	1510–1660	32
		d	$292.65 \pm 0.58 – 1850.75 \pm 0.79$	33
		d	580 ± 6.5	34
		b	0.37 ± 0.02	35
		i, g	658–2121	36
	Range (for Turkish black tea)		0.35-2121	
	United Arab Emirates	d	785.2	10
	Unknown	a	795	6
		b	117.85 ± 4.46	37
		h	213–1228	28
		a	342–1528	38
		b	$1502 \pm 23 - 1528 \pm 29$	39
	Range (for black tea from unknown origins)	b	113.39–1557	39
er all range (for	hange (for black tea from unknown origins)			
		L.	0.287934	_
en	China	b	181.95–2399.64	5
		a	511–2086	6
		h	$7.30 \pm 0.22 – 11.94 \pm 0.32$	40
		b	960 ± 19.8	41
		d	795.1	10
		b	$2206.0 \pm 34.16 – 3276.0 \pm 82.26$	42
		a	385–2081	7
		j	414.14-623.44	43
		b	418.02-625.54	43
	Range (for Chinese green tea)		7.08-3358.26	
	India	a	808	6
		b	$0.618 \pm 0.066 - 0.833 \pm 0.047$	18
	Range (for Indian green tea)	-	0.55-808	. •
	Indonesia	a	918	6
		f	589 ± 47	19
		h	752–790	28
	Range (for Indonesian green tea)	11	542–918	20
	Japan	a	979	6
	Jupan		810 ± 39	22
		g k		22 44
	Panga (for Jananese green tos)	К	$528 \pm 11 - 965 \pm 18$	44
	Range (for Japanese green tea)		517–983	10
	South Africa	d	701.5	10
	South Korea	d	900.8	10
	Sri Lanka	h	214	28
		d	151.3	10
		b	405	29
			151.3–405	
	Range (for Sri Lankan green tea)		1120.50 ± 0.41	33
	Range (for Sri Lankan green tea) Turkey	d		
		d d	786 ± 4.7	34
	Turkey			34
			786 ± 4.7 $73.3-1120.91$	34 6
	Turkey Range (for Turkish green tea)	d a	786 ± 4.7 $73.3 - 1120.91$ $535 - 1584$	6
	Turkey Range (for Turkish green tea)	d a b	786 ± 4.7 73.3 -1120.91 535 -1584 114.64 ± 9.70	6 37
	Turkey Range (for Turkish green tea)	d a b a	786 ± 4.7 73.3 -1120.91 535 -1584 114.64 ± 9.70 439 -1256	6 37 38
	Turkey Range (for Turkish green tea) Unknown	d a b	786 ± 4.7 $73.3-1120.91$ $535-1584$ 114.64 ± 9.70 $439-1256$ $1027 \pm 19-1078 \pm 33$	6 37
	Turkey Range (for Turkish green tea)	d a b a	786 ± 4.7 73.3 -1120.91 535 -1584 114.64 ± 9.70 439 -1256	6 37 38

Table 7. (Continued)

Type of made	tea Country of origin/consumed/marketed	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(s) [†]	
Oolong	China	b	132.70–1011.08	5	
_		a	449-2368	7	
	Range (for Chinese Oolong tea)		132.7-2368		
	Japan	q	915 \pm 11	22	
	Unknown	b	104.78 ± 2.83	37	
		a	436-465	38	
	Range (for Oolong tea from unknown origins)		101.95-465		
Overall range	(for Oolong tea)		101.952368		
Pu-erh	China	a	858	6	
		a	615-1268	7	
	Range (for Chinese Pu-erh tea)		615-1268		
	Japan	g	1020 ± 200	22	
Overall range	(for Pu-erh tea)	3	6151268		
White	China	a	1026	6	
		a	337-1463	7	
Overall range	(for white tea)		3371463		

*In the column, the following letters denote the following points: a, ICP-AES (inductively coupled plasma atomic emission spectrometry); b, FAAS (flame atomic absorption spectrometry); c, ETAAS (electrothermal atomic absorption spectrometry); d, ICP-OES (inductively coupled plasma optical emission spectrometry); e, TNAA (thermal neutron activation analysis); f, ICP-QMS (inductively coupled plasma, quadrupole mass spectrometry); g, ICP-MS (inductively coupled plasma-mass spectrometry); h, polarized X-ray fluorescence spectrometry; i, ICP-ES (inductively coupled plasma emission spectrometry); j, NH₂-MIL-125 (Ti)-modified carbon paste electrode; k, INAA (instrumental neutron activation analysis).

†In the column, the following digits denote the following references: 1, Moreda-Piñeiro et al., 2003; 2, Ferrara et al., 2001; 3, Dambiec et al., 2013; 4, Yasmeen et al., 2000; 5, Zheng et al., 2014; 6, Street et al., 2006; 7, McKenzie et al., 2010; 8, Mehra and Baker, 2007; 9, Polechońska et al., 2015; 10, Donkora et al., 2015; 11, Ashenef, 2014; 12, Gebretsadik and Chandravanshi, 2010; 13, Yemane et al., 2008; 14, Kumar et al., 2005; 15, Marbaniang et al., 2011; 16, Salahinejad and Aflaki, 2010; 17, Srividhya et al., 2011; 18, Pohl and Prusisz, 2007; 19, Pekal et al., 2013; 20, Abdolmaleki et al., 2013; 21: Ansari et al., 2009; 22, Han et al., 2014; 23, Omwoyo et al., 2013; 24, Jabeen et al., 2015; 25, Al-Othman et al., 2012; 26, Al-Oud, 2003; 27, Aksuner et al., 2012; 28, Desideri et al., 2011; 29, Lasheen et al., 2008; 30, Nookabkaew et al., 2006; 31, Narin et al., 2004; 32, Alasalvar et al., 2013; 33, Görür et al., 2011; 34, Kara, 2009; 35, Uçar et al., 2014; 36, Yaylalı-Abanuz and Tüysüz, 2009; 37, Dawodu et al., 2013; 38, Herrador and González, 2001; 39, Memić et al., 2014; 40, Alassane et al., 2013; 41, Brzezicha et al., 2014; 42, Gallaher et al., 2006; 43, Xu et al., 2014; 44, Islam and Ebihara, 2012.

 $1011.08 \ \mu g \ g^{-1}$ in Oolong teas, and 184.69 to 1182.15 $\mu g \ g^{-1}$ in black teas (Zheng et al., 2014). The Mn content in processed made tea was between 1.49 and 2.05 μ g g⁻¹ (Chand et al., 2011). The concentration of Mn ranged from 104.78 \pm 2.83 to 117.85 \pm 4.46 μ g g⁻¹ when three types of teas available in Nigeria (specify the three types) were analyzed for metal contents where Oolong tea had the lowest concentration, while black tea had the highest concentration (Dawodu et al., 2013). In another study, Mn content measured in eight tea samples available in Nigeria varied between 286.1 \pm 2.3 and 801.6 \pm $6.4\mu g g^{-1}$ (Jonah and Williams, 2000). According to Reilly (2004), British dry tea leaves may contain up to 90 μ g g⁻¹. Memić et al. (2014) concluded that the digestion procedure of tea samples significantly varied the metal contents in tea samples and the highest concentration of total Mn was obtained after acid digestion in an autoclave. They reported that Mn in five black tea samples available in the market of Sarajevo in Bosnia and Herzegovina were $1528 \pm 29 \mu g g^{-1}$ when samples were digested by acid digestion in an autoclave. However, for green tea, it was $1078 \pm 33 \ \mu g \ g^{-1}$. In general, the concentration of Mn is the highest of all the analyzed metals like Cu, Fe, and Zn (see later); about 3 times higher than the concentration of Fe and 30-300 times higher than the concentration of Cu and Zn (Memić et al., 2014).

The Mn content in 29 marked brands of black tea and one type of green tea commonly consumed in Turkey was found to be highest relative to other trace metal contents; and ranged from 292.65 \pm 0.58 to 1850.75 \pm 0.79 mg g $^{-1}$ (mean: 1286.35 \pm 0.58 mg g $^{-1}$) Görür et al. (2011). Salahinejad and Aflaki (2010) analyzed 11 black tea (four from Iran, three from India, and four from Sri Lanka) samples available in Iran and reported that Mn content ranged from 362 to 674 μg g $^{-1}$. Manganese contents in three high-quality black tea and four

low-quality black tea grades available in Turkey was ranged from 106 ± 61 to 123 ± 8 mg per 100 g of tea and 112 ± 10 to 124 ± 49 mg per 100 g of tea, respectively (Serpen et al., 2012). The Mn contents in 18 black tea samples available in Italy were ranged from 213 to $1228~\mu g~g^{-1}$ which was reported to be the highest concentrations amongst the essential micronutrients (Desideri et al., 2011). Twelve bagged black tea samples available in Poland were analyzed for Mn and it was found to range from 408 ± 8 to $1090 \pm 27~\mu g~g^{-1}$ (Dambiec et al., 2013). The results of 10 tea samples from the north Iran revealed that the level of Mn contents observed in the range 682 ± 7.51 and $731 \pm 6.56~\mu g~g^{-1}$ (Abdolmaleki et al., 2013). The Mn content in made tea produced from eight tea cultivars in China was ranged from 950.10 to 1224.20 $\mu g~g^{-1}$ (Chen et al., 2009).

Manganese in tea infusion

Table 8 represents Mn contents in tea infusion based on available literatures. From Table 8, it has been observed that the range of Mn content in Chinese, Indian, Sri Lankan, Turkish, and unknown origin's black tea infusion ranges from 0.06 to 1905, 0.11 to 318, 0.08 to 249.4, 0.19 to 387.99, and 1.58 to 340 mg $\rm L^{-1}$, respectively. The overall average Mn content in black tea infusion in all the countries mentioned in Table 8 ranges from 0.06 to 7417 mg $\rm L^{-1}$. From 0.52 to 1276.1, 5.20 to 252.44, and 1.15 to 1765 mg $\rm L^{-1}$ Mn were found in Chinese, Turkish, and green tea originated from unknown regions respectively with the overall range from 0.52 to 1765 mg $\rm L^{-1}$. The overall average Mn content in white tea infusions are ranges from 2.27 to 220 mg $\rm L^{-1}$.

Pohl and Prusisz (2007) reported the fractionation pattern of Mn in black tea and green tea infusion using Amberlite XAD7 and Dowex 50Wx4 sorbents connected in a series, after

the thorough examination of the sorption and desorption characteristics of resins applied. From their findings, they concluded that Mn in tea infusions is predominantly present in the form of cationic species (up to 80% or more, especially in the green teas) and Mn is attached to the polyphenolic compounds, and contributed 31–46% (39% on average) in case of the black teas and from 13–20% (16% on average) in case of the green teas. This diference between the tea species can presumably be attributed to transformation during the manufacturing processes of black teas in which the flavanoids present in the leaves polymerize into more complex and condensed polyphenols of a

much higher molecular weight (Pohl and Prusisz, 2007). About 1.0– $6.0~{\rm mg~L^{-1}}$ of Mn was extracted from tea leaves infusion during a typical six-minute brewing process with 5-g quantities of the tea leaves in 400 mL of boiling doubly distilled water (Pohl and Prusisz, 2007). Pohl and Prusisz (2007) concluded that the total Mn content in prepared black and green tea infusion is a rich source of Mn and is safe to drink when a cup (about 250–300 mL) is served as it may contribute 10–43% (medium 26%) of daily dietary intake.

According to Mehra and Baker (2007), Mn content in tea infusions of eight tea samples produced in different parts of the

Table 8. Concentration of manganese in black, green, Oolong, Pu-reh, and white tea infusion.

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L^{-1})	Reference(s)
Black	Argentina/Vietnam	1	a	230 ± 15	1
	China	2	b	553.7-1905	2
		1	a	$117 \pm 9 – 128 \pm 13$	1
		3	С	3.29-5.78	3
		4	C	0.96–1.92	4
		5	d	0.060-0.166	5
		6	a	5.1 ± 0.11	6
	Range (for Chinese blac		u	0.06–1905	· ·
	Ethiopia	7	a	8.5–13.4	7
	India	8	C	0.106-0.318	8
	IIIdia	1		$120 \pm 8 - 282 \pm 36$	1
			a		
		3	С	1.98–6.24	3
		9	С	125.8–162.9	9
		4	C .	0.91–1.46	4
		5	d	0.108	5
	Range (for Indian black	tea)		0.11–318	
	Indonesia	1	a	138 ± 40	1
	Iran	9	С	136–164	9
	Kenya	4	С	7.59	4
	Kenya/Malawi	1	a	214 ± 18	1
	Russia	4	С	2.07	4
	Sri Lanka	10	a	7.08 ± 0.35	10
		1	a	$59 \pm 12 - 231 \pm 8$	1
		11	a	130.77-220.00	11
		3	C	1.76–6.01	3
		9		88.7–249.4	9
			C		
		4	C	0.7–2.7	4
	D // C:1 1 1	5	d	0.077-0.258	5
	Range (for Sri Lankan b			0.08–249.4	
	Thailand	12	e	1375–7417	12
	Turkey	10	a	$6.49 \pm 0.41 – 8.17 \pm 0.58$	10
		13	f	79.24 \pm 0.25–386.57 \pm 1.42	13
		14	e	0.19–2.11	14
		14	С	0.572 ± 0.0561	14
		4	С	0.56	4
	Range (for Turkish black	c tea)		0.19-387.99	
	Unknown	15	e	175 ± 9	15
		16	d	110–250	16
		17	C	334 ± 6	17
		4	C	1.58	4
	Range (black tea for un		C	1.58–340	7
Overall range (for black		Kilowii regions/		0.067417	
		3	L.		2
Green	China	2	b	287.7–1276.1	2
		18	a	$1.86 \pm 0.83 - 2.48 \pm 0.11$	18
		4	С	0.78–3.89	4
		19	a	0.52–1.9	19
	Range (for Chinese gree	en tea)		0.52-1276.1	
	India	4	С	1.31	4
	Indonesia	4	c	1.22	4
	Japan	4	С	2.74	4
	Sri Lanka	11	a	140.67	11
	Turkey	10	a	5.41 ± 0.21	10
	· ,	13	f	251.69 ± 0.75	13
	Range (for Turkish gree		•	5.20-252.44	13
	nange nor rurkish diee	ii icu/		J.20 ZJZ. TT	
	Unknown	15	e	1619 ± 146	15

Table 8. (Continued)

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L^{-1})	Reference(s) [‡]
		17	С	286 (10)	17
		20	С	3.129 (0.067)-4.217 (0.643)	20
		21	a	1.53 (0.04)-2.01 (0.12)	21
		4	С	1.15-2.53	4
	Range (green tea fo	r unknown regions)		1.15–1765	
	Vietnam	4	С	3.94	4
Overall range (for gree	en tea)			0.521765	
Pu-erh	China	4	С	1.01	4
White	China	4	С	2.27	4
	Unknown	16	d	190–220	16
Overall range (for whi	te tea)			2.27–220	

*In the column, the following bold digits denote the following points: 1, 2.0 g of tea leaves +80 mL of deionized boiling water, covered and steeped for exactly five minutes; 2, 1 g of tea sample (vegetal material) was placed inside a tea sachet (commercially available tea filter, made of unbleached paper). The sachet was introduced into a glass beaker, 100 mL of boiling ultrapure water were added and the beaker was allowed to stand for five minutes; 3, 5 g tea was added to 200 mL of boiling distilled water and allowed to infuse for two minutes; 4, 1 g tea samples were carefully weighed into glass beakers. Fifty milliliters of boiled distilled water was poured on it and covered with watch glasses for five minutes. Infusion was then filtered; 5, 80 cm³ of boiling deionized water to 2.0 g of broken tea leaves and infused for five minutes; 6, 250 mL of boiling tap water were added to one bag of tea in a clean mug for one minute. The bag was then removed and the infusion allowed to cool before analysis; 7, 2 g of tea leaves added in to the boiling water and boil for five minutes, then cooled for five minutes and filtered to 100 mL; 8, not available; 9, 45 mL hot distilled water to 1 g of tea and five minutes infusion; 10, 1.00 g of tea infused for five minutes with 50 mL boiled distilled water; 11, 100 mL of hot distilled water was added to 2 g tea and infused for five minutes at room temperature; 12, 100 mL of boiling deionized water was added to one bag or 2.0 g of tea sample and left at room temperature for five minutes; 13, 100 mL of hot distilled water was added to 2 g of black tea sample. The mixture left to cool at room temperature for five minutes; 14, 2 g of tea sample was weighed in a beaker and 200 mL of boiling ultrapure water was added on it and infused for 15 minutes; 15, 1.5 g tea was infused for three minutes in a cup with 200 mL boiling water and kept for three minutes covered with a watch glass; 16, 2 g of tea leaves/piece of bark was extracted by 100 mL of distilled water (94°C) for 15 minutes; 17, 3 g of made tea sample was transferred into the glass baker. About 80 mL of redistilled water was added to it. Then the baker was placed on a hot plate. The samples were heated up to the boiling temperature for five minutes then cooled and filtered; 18, 2.5 g tea +8 fluid oz boiling water, steeping time three to six minutes; 19, 1.5 g of tea leaf was infused in 250 mL boiling ultrapure water for 10 minutes; 20, 3 g of tea leaf was transferred to the pots and 200 mL of boiled tap water was added and infused for 10 minutes; 21, 5 g of tea sample was infused with 400 mL of boiling doubly distilled water for about six minutes. In the column, the following letters denote the following points: a, FAAS (flame atomic absorption spectrometry); b, HR-CS FAAS (high-resolution-continuum source flame atomic absorption spectrometry); c, ICP-AES (inductively coupled plasma atomic emission spectrometry); d, ETAAS (electrothermal atomic absorption spectrometry); e, ICP-MS (inductively coupled plasma-mass spectrometry); f, ICP-OES (inductively coupled plasma optical emission spectrometry). th the column, the following digits denote the following points: 1, Dambiec et al., 2013; 2, Paz-Rodríguez et al., 2015; 3, Mehra and Baker, 2007; 4, Street et al., 2006; 5, Polechońska et al., 2015; 6, Hope et al., 2006; 7, Gebretsadik and Chandravanshi, 2010; 8, Chand et al., 2011; 9, Salahinejad and Aflaki, 2010; 10, Aksuner et al., 2012; 11, Lasheen et al., 2008; 12, Nookabkaew et al., 2006; 13, Görür et al., 2012; 14, Sofuoglu and Kavcar, 2008; 15, Milani et al., 2015; 16, Jeszka-Skowron et al., 2015; 17, Memić et al, 2014; 18, Gallaher et al., 2006; 19, Reto et al., 2007; 20, Petit et al., 2013; 21, Pohl and Prusisz, 2007.

world, decreased with increasing sequential infusion times like 2, 5, and 10 minutes with boiling water. The Mn concentrations in two-minute tea infusions obtained from commercial tea samples from around the world varied between 1.76 and 11.61 mg L^{-1} , while the percent transfers in the first, second and third infusions (2, 5, and 10 minutes) being 29.5, 12.6, and 4.3%, respectively (Mehra and Baker, 2007). Wróbel et al. (2000) reported that among several micronutrients, Mn was leached at a higher rate (33.6% of total Mn for black tea and 44.6% of total Mn for green tea); suggesting that tea drinking could be a good dietary source of Mn. Wróbel et al. (2000) also concluded that micronutrient contents in tea infusion is related to preparation process such as sample mass to volume of water, temperature, time, agitation, repetition of infusion etc. Nookabkaew et al. (2006) reported that the concentration of Mn in 18 tea infusions that were each prepared from 2.0 g or one tea bag in 100 mL were ranged from 1.38 to 7.42 μ g L⁻¹ with the mean of 4.10 μ g L⁻¹; and contained 25.52% of the total Mn released through tea infusion. Sofuoglu and Kavcar (2008) reported that 188-2105 μ g L⁻¹ Mn was infused from black tea samples available in Turkey.

Among the seven tea samples from Egypt, Mn content in tea infusion are ranged from 130.77 to 220.0 $\mu g \, g^{-1}$ when tea infusion was produced from 2 g tea with 100 mL of hot distilled water for five minutes (Lasheen et al., 2008). The Mn content in tea infusion prepared using 1 g each for 15 tea samples that originated from different tea growing countries infused with 50 mL boiled distilled water for 15 minutes ranged from 1.50 \pm 0.10 to 10.9 \pm 0.7 mg L⁻¹ (Malik et al., 2008). Therefore, the

average Mn portion in the infusions represented 31% of the total element content in raw tea leaves material, which was comparable to the results of Mehra and Baker (2007). The Mn content in infusions of nine green teas marketed in Portugal prepared from 1.5 g of each tea added into 250 mL of boiling ultra-pure water infused for 10 minutes varied between 0.52 \pm 0.1 and 1.9 \pm 0.2 mg L^{-1} (Reto et al., 2007).

Fernández et al. (2002) prepared infusion from 20 green and 24 black teas available in Spanish market and found that Mn contents in three and five minutes of green tea infusions were ranged from 1.3-3.5 mg L^{-1} (mean: 2.1 mg L^{-1}) and 1.5-3.8 mg L^{-1} (mean: 2.6 mg L^{-1}), respectively. However, the study showed that Mn contents in the black tea infusions brewed for three and five minutes did not differ significantly but varied between 1 and 6 mg $\rm L^{-1}$ (mean: 3 mg $\rm L^{-1}$). The Mn content measured in tea infusions obtained from 30 tea samples (13 green tea samples, 13 black tea samples, 2 semifermented, and 2 white tea samples) of different origins imported to the Czech Republic were ranged from $0.56-7.9 \mu g g^{-1}$ after five minutes extraction time (Street et al., 2006). The proportion of Mn contents in the infusion related to the respective total contents in leaves was $18 \pm 10\%$ thereby confirming that tea infusion can be an important dietary source of Mn. Street et al. (2006) further concluded that Mn content increased with time and differed statistically across the four major tea groups studied. The Mn content was higher in the five minutes green tea infusions than in the five minutes black tea infusion, and statistically higher in black tea in the 60-minute and 24-hour

infusions. Furthermore, they concluded that Mn was the only element found in significant dietary amounts in the tea infusions amongst all the analyzed micronutrients. Tea infusion prepared from three black tea samples in India showed that the dissipation of manganese from made tea to its brew ranged from 0.106 to 0.424 μ g g⁻¹ (Chand et al., 2011). Comparatively high amount of Mn in made tea and its subsequent higher leaching to tea infusion could be due to the soil applied P as it plays a significant role in the Mn uptake by tea plant. Among five black and five green tea samples found in Bosnia and Herzegovina, Mn content in tea infusion ranged from 286 \pm 10 to $334 \pm 6 \,\mu\mathrm{g}\,\mathrm{g}^{-1}$ (Memić et al., 2014). Average Mn content in 11 tea infusion that originated from Iran, India, and Sri Lanka was 57.63 $\mu g g^{-1}$ (Salahinejad and Aflaki, 2010). In another related study, measured Mn contents in 12 tea infusions ranged from 59 ± 12 to $224 \pm 12~\mu g~g^{-1}$ (Dambiec et al., 2013). The Mn content measured in infusions of 40 samples of loose teas and tea bags from Norwegian supermarkets ranged from 273- $5140~\mu {\rm g}~{\rm L}^{-1}$ (mean: $1100\pm863~\mu {\rm g}~{\rm L}^{-1}$) and $744-5710~\mu {\rm g}~{\rm L}^{-1}$ (mean: 2990 \pm 1220 μ g L⁻¹) respectively. However, Mn contents in 30 tea infusions from Polish supermarket ranged from 279 to 1780 $\mu g L^{-1}$ (mean: 797 \pm 441 $\mu g L^{-1}$) and 291 to 2930 $\mu \mathrm{g \ L^{-1}}$ (mean: 1800 \pm 789 $\mu \mathrm{g \ L^{-1}}$) for loose tea and tea bag, respectively (Kronborg, 2013).

Molybdenum

Molybdenum (Mo) is an essential trace element for human beings and animals. Although Mo is essential, it might create adverse effects if exposure is excessive to humans through food and beverage. It is an essential element required by human in a minute amount for the function of bounding of several molybdo enzymes. Molybdenum has been reported to reduce the incidence and severity of dental caries (Burguera and Burguera, 2007). Also, it has been reported to be beneficial to various groups of individuals, including those with sulfite sensitivity, asthmatics and people with elevated urinary ratios. The WHO (1993) estimated a daily requirement for molybdenum to vary between 0.1 and 0.3 mg d⁻¹ for adults. Very little research has been done on Mo and its role for tea plant and therefore, Mo content in made tea and tea infusion is scanty.

Molybdenum in made tea and tea infusion

As indicated earlier, works on Mo content in made tea and tea infusion is very limited. Ferrara et al. (2001) analyzed the Mo contents in 10 different commercial teas obtained from Syria, Russia, and China using AAS and reported no Mo in 4 samples, viz. Black Syria tea, Red Syrian long leaf tea, Red Syrian tea, and Russian tea. Similarly, no Mo content was found in seven Chinese tea samples namely Tè toucha Yunnan, Kwong sang tea, Green tea china, Tiamu king ding, Tikuan yin, Long-ching, and Oolong china except in Tè toucha Yunnan black tea where 161.9 μ g g⁻¹ was obtained (Ferrara et al., 2001). Nine tea samples which were commonly available in commercial market in Italy, namely, Compagnia del tè, Lipton tea light quality, Star tea detanninated, Te Ati (det.), Tè star classic blend, The lyons ceylon tea, Twinning earl grey tea, Twinning English tea, and Twinning Prince of Wales tea were analyzed for the estimation

of Mo content through AAS by acid digestion methodology (Ferrara et al., 2001). Among these samples, only one sample (Twinning Prince of Wales tea) showed Mo content of 570.8 μ g g⁻¹. A multielement determination of major-to-trace elements in black tea leaves and their tea infusions was carried out by Matsuura et al. (2001) using ICP-AES and ICP-MS and Mo content in black tea was found to be $0.041 \pm 0.001 \ \mu g \ g^{-1}$ with 3% relative standard deviation. The poor precision for Mo may be ascribed to its extremely low concentration, close to the analytical detection limit obtained by the ICP-MS instrument. Matsuura et al. (2001) reported that Mo is a poorly extractable element with only 0.12 ± 0.05 mg L⁻¹ extracted (<20% of total Mo) in a tea infusion prepared from 2 g of a powdered black tea leaves added to 100 mL of boiling water and infused for five minutes. Molybdenum concentrations in black tea leaves, green tea leaves, and tea leaves standard reference material (NIES No. 7) determined by ICP-AES and ICP-MS were 0.041 \pm 0.001 and 0.031 \pm 0.012 μ g g⁻¹, respectively (Matsuura et al., 2001). Molybdenum contents in three high-quality black tea and four low-quality black tea grades available in Turkey ranged from 0.10 ± 0.03 – 0.04 ± 0.00 mg per 100 g of tea and 0.01 ± 0.00 – 0.02 ± 0.00 mg per 100 g of tea, respectively (Serpen et al., 2012). Gürkan et al. (2013) reported that Mo contents in green tea sample could be $3.63 \pm 0.15 \ \mu g L^{-1}$. Mo in low- and highquality instant black teas available in Turkey were 4.2 and 2.1 μ g g⁻¹, respectively (Alasalvar et al., 2013).

Zinc

Zinc (Zn) deficiency in human was first recognized in 1963 and is known as a growth factor for human being (Prasad, 2012). Zinc is an essential micronutrients for the crystallization of insulin in pancreatic β -cells and is required by thousands of proteins for catalytic, structural, or transcriptional functions (Zhu and Jing, 2013; Park et al., 2014). Zinc deficiency may also increases the tendency in human being to catch cold (Fischer et al., 2011), resulting retardation in growth, male hypogonadism, gonadal failure, intercurrent infections, premature death and impaired cognitive functions, and anorexia (Prasad, 2012). Zinc deficiency can also adversely affect reproduction in both male and female, diarrhea and pneumonia (Nriagu, 2011). Presently 2 billion people in the world is suffering for Zn deficiency (Prasad, 2012). The concentrations of Zn measured in various black and green tea samples from different countries are contained in Table 9.

Zinc in made tea

From Table 9, it has been observed that the range of Zn contents in Chinese, Ethiopian, Indian, Indonesian, Iranian, Kenyan, Pakistani, Saudi Arabian, Sri Lankan, Syrian, Turkish, and unknown origins' black tea ranges from 15 to 1000, 8.6 to 330.5, 0.08 to 874.8, 22.94 to 490.7, 22.2 to 127.22, 17.1 to 60, 1.3 to 59.4, 6.6 to 120, 11.2 to 645.4, 18 to 730.5, 6.71 to 147.5, and 21 to 84.6 μ g g⁻¹, respectively. The overall average Zn content in black tea in all the countries mentioned in Table 9 ranges from 0.08 to 1000 μ g g⁻¹. For green tea, the ranges are from 0.11 to 630, 25.47 to 53.8, 20.1 to 30.1, 13.8 to 65.2, 24.2 to 70.94, 7.97 to 30.57, and 19.64 to 82.2 μ g g⁻¹

 Table 9. The concentration of zinc in black, green, Oolong, Pu-reh, and white tea samples obtained from various sources.

Type of made tea	Origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(s) [†]
Black	Africa (Burundi, Ethiopia, Kenya, Malawi, Tanzania, and Zimbabwe)	a	25.06 ± 9.76	1
	Argentina	b	621.3	2
	Argentina/Vietnam	b	26.8 ± 2.76	3
	Asia (India and Sri Lanka)	a	24.39 ± 7.39	1
	Bangladesh	C	0.45-1.00	4
	China	b	27.76–65.32	5
		a	42.4–71.3	6
		b	$22.6 \pm 0.919 - 22.9 \pm 2.47$	3
		b	431.3–1000	2
		a	15–38	7
		b d	27.76–65.32 23.6–33.5	5 8
	Range (for Chinese black tea)	ď	25.0–55.5 15–1000	0
	England	e	21.4–33.8	9
	Ethiopia	d	8.6–198.3	10
	Linopia	b	$20.2 \pm 0.1 - 21.6 \pm 0.3$	11
		b	57.9–330.5	12
	Range (for Ethiopian black tea)	b	8.6–330.5	12
	India	a	33–54.9	6
	maia	b	0.080-0.10	13
		b	$20.9 \pm 0.283 - 22.5 \pm 1.34$	3
		b	523.2-874.8	2
		b	9.1–15.1	14
		a	21.20–33.12	15
		b	25.39 ± 0.59	16
		d	27.9	8
	Range (for Indian black tea)	ď	0.08-874.8	O
	Indonesia	b	24.0 ± 1.06	3
	madricsia	b	490.7	2
		f	025.3 ± 1.3	17
	Range (for Indonesian black tea)	•	22.94–490.7	.,
	Iran	b	40.3 ± 13.9	18
		b	50.7	19
		b	55.35–127.22	20
		a	22.20–26.88	15
		d	58.28 ± 4.25	21
	Range (for Iranian black tea)	<u>.</u>	22.2–127.22	
	Kenya	a	51.1	6
		e	34.1	9
		b	18.8–44.9	22
		b	17.1–38.9	22
		b	25–40.5	23
		b	46.5 ± 13.5	24
		b	32.13-42.63	24
	Range (for Kenyan black tea)		17.1–60	
	Malawi	b	20.3 ± 1.34	3
	Myanmar	b	586.7	2
	Pakistan	d	$2.1 \pm 0.8 – 12.7 \pm 0.2$	25
		b	7.66-59.40	26
		b	23.7-25.6	27
	Range (for Pakistani black tea)		1.3-59.4	
	Russia	a	30	6
	Russia	b	565.8	2
	Rwanda	b	18.4–27.9	22
	Saudi Arabia	b	6.6–120	28
		b	26.69-53.89	29
	Range (for Saudi Arabian black tea)		6.6-120	
	Singapore	g	28.42	30
	Sri Lanka	a	32.2-47.5	6
		b	$18.6 \pm 1.98 – 27.0 \pm 0.919$	3
		h	25.3	31
		e	11.2–71.7	9
		a	21.2–33.12	32
		b	645.4	2
		b	60.70-90.65	33
		a	23.37–32.34	15
		b	16.50 ± 1.40	34
		d	23.2–37.3	8
	Range (for Sri Lankan black tea)	<u>.</u>	11.2-645.4	
	Syria	b	18.0-44.2	35
		b	440.0–730.5	2
	Range (for Syrian black tea)		18–730.5	

(Continued on next page)

 Table 9. (Continued)

ype of made tea	Origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(
	Taiwan	С	0.9–1.5	36
	Tanzania	b	20.2-27.8	22
	Thailand	C	10.13-55.40	37
	Turkey	a	21.5	6
	rancy	b	$18.5 \pm 1.4 - 19.2 \pm 1.9$	38
		С	17.8–28.5	39
		e	$13.41 \pm 0.08 - 48.27 \pm 0.41$	40
		С	18.6 ± 6.3	41
		b	112.7–147.5	42
		b	$7.01 \pm 0.30 – 10.50 \pm 0.71$	34
		b	$7.01 \pm 0.30 – 16.50 \pm 1.40$	34
	Range (for Turkish black tea)		6.71–147.5	
	Uganda	b	20.2–31.7	22
	United Arab Emirates		30.3	9
		e		
	Unknown	a	51.8	6
		b	30.66 ± 1.53	43
		h	84.6-27.8	31
		a	21–47	44
		b	$27.9 \pm 2.5 - 41.0 \pm 2.4$	45
	Dange (black too from unknown origins)	Ь		73
	Range (black tea from unknown origins)		21–84.6	
erall range (for b		_	0.081000	
een	China	b	24.11–54.93	5
		a	33.2-68.7	6
		i	$0.13 \pm 0.02 – 0.15 \pm 0.02$	46
		b	37 ± 2.9	47
			$24.19 \pm 0.12 - 31.86 \pm 1.21$	48
		a		
		e	28	9
		g	48.77	30
		b	630.0	2
		b	$58.00 \pm 2.82 - 81.00 \pm 11.38$	49
		a	14–39	7
			$67.9 \pm 0.8 - 69.6 \pm 0.9$	50
		d, j		
		b	24.11–54.93	5
	Range (for Chinese green tea)		0.11–630	
	India	a	46.6	6
		d, j	$51.5 \pm 0.9 – 53.0 \pm 0.8$	50
		b	26.39 ± 0.92	16
	Range (for Indian green tea)	ь	25.47-53.8	10
				_
	Indonesia	a	30.1	6
		f	22.0 ± 1.9	17
		h	22.7–23.5	31
	Range (for Indonesian green tea)		20.1–30.1	
	Japan	a	65.2	6
		k	$14.7 \pm 0.9 – 26.4 \pm 1.2$	51
		d, j	$47.2 \pm 0.9 - 49.7 \pm 0.9$	50
	D (f +)	u, j		30
	Range (for Japanese green tea)		13.8–65.2	_
	South Africa	e	31.6	9
	South Korea	e	51.2	9
	Sri Lanka	h	24.2	31
		e	31.3	9
		b	70.94	33
	Range (for Sri Lankan green tea)	5	24.2–70.94	33
		_		20
	Taiwan	С	4.8–9.7	36
	Turkey	e	22.50 ± 0.58	40
		С	21.4 ± 9.17	41
		b	$8.52 \pm 0.55 – 14.80 \pm 1.31$	34
		b	$8.52 \pm 0.55 – 14.80 \pm 1.31$	34
	Range (for Turkish green tea)	~	7.97–30.57	٥.
	Unknown	2		,
	UIIMIUWII	a	32.1–82.2	6
		b	21.17 ± 1.53	43
		a	23–55	44
		b	$21.8 \pm 0.8 – 30.8 \pm 1.2$	45
		d	23.9 ± 0.8 – $29.9 \text{ v } 0.7$	52
		j	$25.3 \pm 0.7 - 27.5 \pm 0.8$	52
	Panga (groon toa from unknown arisina)	J		<i>J</i> ∠
	Range (green tea from unknown origins)		19.64–82.2	_
	Vietnam	a	40.3	6
erall range (for g	reen tea)		0.11630	
long	China	b	18.04-40.56	5
9			24.59	30
		g h		
		b	523.9	2
		a	10–26	7
		a	18.04–40.56	,

Table 9. (Continued)

Type of made tea	Origin	Instrument(s) used for analysis*	Concentration (μ g g ⁻¹)	Reference(s) [†]
	Range (for Chinese Oolong tea)		10–523.9	
	Taiwan	С	2.5-4.6	36
	Unknown	b	40.00 ± 3.13	43
		a	29-38	44
	Range (Oolong tea from unknown origins)		29-43.13	
Overall range (for			2.5523.9	
Pu-erh	China	a	45.4	6
		С	37.5-61.3	53
		a	37.5-61.3	53
		a	26–52	7
Overall range (for	Pu-erh tea)		2661.3	
White	China	a	75.2	6
		a	20-44	7
Overall range (for	white tea)		2075.2	

*In the column, the following letters denote the following points: a, ICP-AES (inductively coupled plasma atomic emission spectrometry); b, FAAS (flame atomic absorption spectrometry); c, ICP-MS (inductively coupled plasma-mass spectrometry); d, ETAAS (electrothermal atomic absorption spectrometry); e, ICP-OES (inductively coupled plasma optical emission spectrometry); f, ICP-QMS (inductively coupled plasma, quadrupole mass spectrometry); g, capillary electrophoresis; h, polarized X-ray fluorescence spectrometry; i, X-ray fluorescence; j, SWASV (square wave anodic stripping voltammetry); k, INAA (instrumental neutron activation analysis).

În the column, the following digits denote the following references: 1, Moreda-Piñeiro et al., 2003; 2, Ferrara et al., 2001; 3, Dambiec et al., 2013; 4, Islam et al., 2013; 5, Zheng et al., 2014; 6, Street et al., 2006; 7, McKenzie et al., 2010; 8, Polechońska et al., 2015; 9, Donkora et al., 2015; 10, Ashenef, 2014; 11, Gebretsadik and Chandravanshi, 2010; 12, Yemane et al., 2008; 13, Chand et al., 2011; 14, Marbaniang et al., 2011; 15, Salahinejad and Aflaki, 2010; 16, Srividhya et al., 2011; 17, Pekal et al., 2013; 18, Ansari et al., 2009; 19, Moghaddam et al., 2008; 20, Nejatolahi et al., 2014; 21, Ziarati et al., 2013; 22, Moseti et al., 2013; 23, Njogu et al., 2014; 24, Omwoyo et al., 2013; 25, Jalbani et al., 2007; 26, Soomro et al., 2008; 27, Jabeen et al., 2015; 28, Al-Othman et al., 2012; 29, Al-Oud, 2003; 30, Feng et al., 2003; 31, Desideri et al., 2011; 32, Albani et al., 2013; 33, Lasheen et al., 2008; 34, Soylak et al., 2007; 35, Antakli and Al-Check, 2011; 36, Shen and Chen, 2008; 37, Nookabkaew et al., 2016; 38, Aksuner et al., 2012; 39, Alasalvar et al., 2013; 47, Brzezicha et al., 2014; 48, Chen et al., 2009; 49, Gallaher et al., 2006; 50, Melucci et al., et al., 2013; 51, Islam and Ebihara, 2012; 52, Melucci et al., 2013; 53, Cao et al., 2010.

Zn for Chinese, Indian, Indonesian, Japanese, Sri Lankan, Turkish, and unknown origins' green teas, respectively. The overall average of Zn content in green tea in all the countries mentioned in Table 9 is ranged from 0.11 to 630 μ g g⁻¹. For Oolong tea, the ranges are from 10 to 523.9 and 29 to 43.13 $\mu g g^{-1}$ Zn for Chinese and unknown origins' Oolong teas, respectively. The overall average of Zn content in Oolong tea for all the countries mentioned in Table 9 are ranges from 2.5 to 523.9 μ g g⁻¹. The overall average of Zn content in Puerh and white teas in all the countries mentioned in Table 9 are ranges from 26 to 61.3 and 20 to 75.2 μ g g⁻¹, respectively. Görür et al. (2011) reported that the total Zn in made tea collected in Turkey were ranged from 23.59 to 120.46 μ g g⁻¹. On the other hand, a comparative higher Zn level of 147.5 μ g g⁻¹ have been reported by Narin et al. (2004). Matsuura et al. (2001) have reported the mean Zn level in black tea as 36.6 \pm $0.7 \mu g g^{-1}$. A microwave assisted digestion procedure for the determination of Zn in 10 tea samples available in Turkey was established employing flame atomic absorption spectrometry and measured Zn concentration ranged from 7.0 to 16.5 μg g⁻¹ (Soylak et al., 2007). Earlier report on Zn contents in African and Asian tea was also found as 25.06-26.29 μ g g⁻¹ (Moreda-Piñeiro et al., 2003). The Zn levels in different Kenyan unprocessed tea samples varies between 15.4 and 32.6 μg g⁻¹ with the highest content recorded in Cheboswa tea, while the lowest Zn content was recorded in Kitumbe sample (Moseti et al., 2013). These authors also reported that Zn levels among Kenyan black tea samples were varied between 18.8 and 44.9 μg g $^{-1}$ with black tea from Kitumbe having the lowest content (19.4 μ g g⁻¹), while that from Chemomi having the highest (40.8 μ g g⁻¹).

Study by AL-Oud (2003) suggested that Zn concentration in Chinese and Indian teas are ranged between 26.7 and 53.9 μ g

g⁻¹. The level of Zn in 17 black tea samples consumed in Saudi Arabia, analyzed by ICP-AES revealed that it varied between 23.7 and 122 μ g g⁻¹ (Ashraf and Mian, 2008). Dambiec et al. (2013) analyzed 12 commercial bagged black teas from different origin marketed at the local stores in Wrocław, Poland and the highest Zn concentration in tea leaves was noted in tea from Sri Lanka (27.0 \pm 0.919 $\mu \mathrm{g}~\mathrm{g}^{-1}$) and the lowest (17.9 \pm $0.849 \ \mu g \ g^{-1}$) in one of the blended teas of unspecified origin. Herrador and González (2001) analyzed 48 commercial tea samples comprising of 26 black, 18 green, and 4 Oolong teas marketed in Spain were analyzed for Zn using ICP-AES. Measured content of Zn in black, green, and Oolong teas ranged from 24 \pm 3–47 \pm 1 μg g^{-1} , 28 \pm 4–55 \pm 5 μg g^{-1} , and 31 \pm $3-38 \pm 3 \mu g g^{-1}$, respectively. Moreda-Piñeiro et al. (2003) analyzed Zn in 54 tea samples originating from various Asian and African countries using ICP-AES and ICP-MS and reported that the average values of Zn in African (n = 18) and Asian (n=36) countries were 25.06 \pm 9.76 and 26.29 \pm 11.33 μ g g⁻¹, respectively. In this study, average values of Zn concentrations in tea samples from China (n = 13) and India + Sri Lanka (n = 13) were 27.85 \pm 12.37 and 24.39 \pm 7.39 μ g g^{-1} , respectively. From this data, Moreda-Piñeiro et al. (2003) concluded that the differentiation and classification of tea samples from Africa and Asia is made possible using the Zn profile in the samples and application of multidimensional chemometric techniques.

The Zn content in 31 tea samples consumed in Iranian has been reported to range from 19.80 to 97.80 μg g⁻¹ with the mean of 50.7 μg g⁻¹ (Moghaddam et al., 2008). Han et al. (2007) assessed an on-line sample digestion method of higher heating efficiency and rate using electromagnetic heating technique through FI-FAAS for estimation of Zn in five tea samples. The contents of Zn were found to range from 34.8 \pm 0.8–

53.1 \pm 1.7 and 35.0 \pm 1.1-52.5 \pm 1.8 $\mu g g^{-1}$ in an off-line digestion and the online electromagnetic heating technique, respectively. Among 18 tea samples (4 samples produced in Thailand and 14 samples imported from China, Japan, Sri Lanka, India, and Indonesia) assessed, measured Zn content ranged from 10.13 to 55.40 $\mu g g^{-1}$ with the mean of 32.17 μg g⁻¹ (Nookabkaew et al., 2006). The Zn content in five commercial tea samples collected from Shillong Meghalaya, India ranged from 9.1 to 15.1 $\mu g g^{-1}$ with a mean value of 12.2 \pm $2.2 \mu g g^{-1}$ (Marbaniang et al., 2011). Shen and Chen, (2008) reported that mean Zn concentration of 1.2 $\mu g g^{-1}$ in Chinese black tea leaves. Zhu et al. (2011) reported that Zn content in green tea samples collected from Shizuoka Prefecture of Japan were 22.58 ± 0.29 to $22.86 \pm 0.07 \ \mu g \ g^{-1}$.

Ahmad et al. (2012) analyzed 12 black tea samples commonly consumed in Pakistan using FAAS and revealed that Zn content varied between 1.05 and 3.21 μ g g⁻¹ with the mean of 2.06 μ g g⁻¹. The Zn contents in 16 tea samples from Addis Ababa, Ethiopia ranged from 8.6–198.3 μ g g⁻¹ (Ashenef, 2014) while Zn contents range of 43.9-71.3and 20.2-21.2 μ g g⁻¹ were also reported by Yemane et al. (2008) and Woldegebriel et al. (2012), respectively. Zinc content in 5 green tea samples (three green teas from India, China, and Japan, and three kinds at the local markets in Ravenna, Italy) available in Italy was determined by square wave anodic stripping voltammetry was found to vary between 25.3 \pm 0.7 and 69.6 \pm 0.9 μ g g⁻¹ (Melucci et al., 2013). Zinc content measurement using ETAAS ranged from 23.9 \pm 0.8–67.9 \pm 0.8 μ g g⁻¹. Zinc contents in 30 black tea samples cultivated in Iran varied between 34.1 and 47.4 μ g g⁻¹ with the mean of 40.3 μ g g⁻¹ while the content in 30 black tea samples imported to Iran ranged from 21.6 to 46.7 μ g g⁻¹ with the mean of 35.8 μ g g⁻¹ (Ansari et al., 2007). Among 48 commercial tea samples comprising of 15 green tea, 15 black tea, and 18 Oolong tea, the content of Zn measured were 4.8–9.7 μg g⁻¹ (mean: 6.3 μg g⁻¹), 0.9–1.5 μg g⁻¹ (mean: 1.2 μg g⁻¹), and 2.5–4.6 μg g⁻¹ (mean: 3.4 μg g⁻¹) in green tea, black tea, and Oolong tea, respectively (Shen and Chen, 2008). Zinc contents in three black and three green tea samples available in Poland ranged from 0.0262 \pm 0.0011 to 0.0223 \pm $0.0050~{\rm mg~g}^{-1}$ and $0.0232~\pm~0.0070~{\rm to}~0.0346~\pm~0.0020~{\rm mg}$ g⁻¹, respectively (Pohl and Prusisz, 2007).

Mose et al. (2014) reported that the Zn content in black tea samples collected from three agroecological areas (Murang'a, Meru, and Kisii) in Kenya ranged between 27 \pm 1 and 39 \pm $7\mu g g^{-1}$. Zn content in 7 tea samples marketed in Egypt ranged from 65.84 to 90.65 μg g⁻¹ as reported by Lasheen et al. (2008). A total of 50 tea samples collected from Gilan Province of Iran used for measurement of Zn content revealed that Zn content in these samples varied between 40.48 \pm 2.86 and $58.28 \pm 4.25 \ \mu g \ g^{-1}$ (Ziarati et al., 2013). Malik et al. (2008) analyzed Zn in 15 types of pure mixture of tea leaves that originated from different countries and reported that zinc content ranged from 15.1 \pm 0.7 to 60.3 \pm 2.5 μ g g⁻¹. The range of Zn concentration in 10 black tea samples available from Jazan and Jeddah region of Saudi Arabia was from 6.6 to 120 μ g g⁻¹ (Al-Othman et al., 2012). Zinc levels in tea leaf samples from Syria have been reported to range from 18.0 to 44.2 μ g g⁻¹ (Antakli and Al-Check, 2011) while the measured Zn concentration in black tea samples from Saudi Arabia ranged from 26.69to 53.89 μ g g⁻¹ (AL-Oud, 2003). Gebretsadik and Chandravanshi (2010) reported that the range of zinc in Ethiopian Black tea samples is 20.2–21.6 μ g g⁻¹. Zheng et al. (2014) analyzed the Zn concentrations in 43 representative tea products comprising 18 green, 12 Oolong, and 13 black teas from 7 main tea production provinces in China using FAAAS and reported a range of 24.11–54.93 μg g⁻¹ in green teas, 18.04–40.56 μg g⁻¹ in Oolong teas, and 27.76–65.32 μg g⁻¹ in black teas. The Zn contents measured in four black tea samples in Nigeria ranged from 0.01 \pm 0.00–0.02 \pm 0.01 $\mu \mathrm{g}~\mathrm{g}^{-1}$ with the mean of 0.02 \pm $0.01~\mu g~g^{-1}$ (Achudume and Owoeye, 2010). In processed made tea from India, the total concentration of Zn was found to be ranged from 0.080 to 0.10 μ g g⁻¹ (Chand et al., 2011). Dawodu et al. (2013) reported that Zn content in green tea, black tea and Oolong teas available in Nigeria were 21.17 \pm 1.53, 30.66 \pm 1.53, and 40.00 \pm 3.13 μ g g⁻¹, respectively.

Total Zn contents in black tea after an autoclave, dry ashing in an oven, acid digestion in a closed system and acid digestion in an open system digestion procedures were 41.0 \pm 2.4, 33.6 \pm 1.2, 27.9 \pm 2.5, and 28.2 \pm 2.8 $\mu \mathrm{g} \, \mathrm{g}^{-1}$, respectively, while for green tea sample, it was 30.8 \pm 1.2, 28.1 \pm 1.13, 26.9 \pm 1.9, and $21.8 \pm 0.8 \ \mu g \ g^{-1}$, respectively (Memić et al., 2014). According to these authors, the lower concentrations of Zn metal obtained after wet digestion at normal pressures apparently originate from incomplete digestion process of samples. The measured Zn content in 29 marketed brands of black tea and one type of green tea commonly consumed in Turkey were ranged from 13.41 \pm 0.08 to 48.27 \pm 0.41 mg g^{-1} with a mean value of 22.65 ± 0.55 mg g⁻¹ (Görür et al., 2011). Salahinejad and Aflaki (2010) analyzed 11 black tea samples available in Iran comprising of 4 from Iran, 3 from India and 4 from Sri Lanka and reported Zn content range of 21.2-33.1 μ g g⁻¹. Zinc contents in seven black tea samples that includes three high-quality and four low-quality black tea grades available in Turkey were ranged from 1.65 \pm 0.12 to 1.90 \pm 0.15 mg per 100 g and 1.51 \pm 0.02 to 2.20 \pm 0.11 mg per 100 g of tea, respectively (Serpen et al., 2012). The measured Zn contents in 18 black tea samples available in Italy were ranged from 20.4 to 84.6 μ g g⁻¹ (Desideri et al., 2011). Twelve bagged black tea samples available in Poland were analyzed for Zn and the measured content ranged from 17.9 \pm 0.849 to 27.0 \pm 0.919 μ g g⁻¹ (Dambiec et al., 2013) while Zn content in made tea produced from eight tea cultivars in China were ranged from 24.19 to 31.86 $\mu g g^{-1}$ (Chen et al., 2009).

Zinc in tea infusion

From Table 10, it has been observed that the range of Zn content in Chinese, India, Iranian, Pakistani, Sri Lankan, Turkish, and unknown origin's black tea infusion ranges from 0.181 to 163, 0.098 to 18.03, 2.61 to 11.39, 0.59 to 6.20, 0.15 to 43.50, 0.04 to 18.14, and 0.21 to 34.99 mg L⁻¹, respectively. The overall average Zn content in black tea infusion in all the countries mentioned in Table 10 are ranged from 0.04 to 433.20 mg L^{-1} . From 0.09 to 188.20, 0.14 to 12.82, and 0.13 to 34.98 mg L^{-1} Zn were found in Chinese, Turkish, and green tea originated from unknown regions respectively with the overall range from 0.09 to 188.20 mg L⁻¹. The overall average of Zn content in white tea infusions ranges are from 2.27 to 220 mg L^{-1} .

Görür et al. (2011) reported that Zn contents in tea infusions prepared from 100 mL of hot distilled water added to 2 g of black tea sample ranged from 2.83 to 17.97 mg g $^{-1}$. The same mixtures when left to cool at room temperature for five minutes and then filtered to obtain the clear solution for further processing and the Zn contents measured show a range from 6.74 to 33.39 mg g $^{-1}$ in the brew. The concentration of Zn in tea infusion and per cent transfer to tea infusion in relation to extraction times in 31 tea sample brands in Iran market has been documented by Moghaddam et al. (2008). In the study, concentrations of Zn in the tea infusions in the first (2 minutes), second (5 minutes), and third (10 minutes) infusions ranged from 0.12 to 0.60 mg L $^{-1}$ (mean: 0.32 mg L $^{-1}$), 0.05 to 0.29 mg L $^{-1}$ (mean: 0.14 mg L $^{-1}$), and 0.02 to 0.14 mg L $^{-1}$ (mean:

0.07 mg L⁻¹), respectively. However, the cumulative percent transfer of Zn in the first (2 minutes), second (5 minutes), and third (10 minutes) infusions are ranged from 26.7 to 37.2 (mean: 31.7), 39.8 to 53.4 (mean: 45.43), and 47.2 to 61.3 (mean: 52.75), respectively. The solubility of Zn in the first infusion was significantly (P < 0.01) higher than that of the second infusion while the solubility in the second infusion was also higher significantly (P < 0.05) than the third infusion. Similar findings have been reported by Wong et al. (1998), where the solubility of Zn in the first infusion was significantly higher than the second infusion. Nookabkaew et al. (2006) reported that the concentration of Zn in 18 tea infusion prepared from 2.0 g or one bag of tea in 100 mL are ranged from 5.947 to 43.32 μ g 100 mL⁻¹ with the mean of 21.94 μ g

Table 10. Concentration of zinc in black (CTC and orthodox), green, Pu-reh, and white tea infusion.

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L^{-1})	Reference(s)
Black	Argentina/Vietnam	1	a	6.25 ± 0.424	1
	China	2	b	21.2–163	2
		1	a	$4.85 \pm 0.354 – 8.00 \pm 0.707$	1
		3	С	0.181-0.244	3
		4	d	5.45-12.14	4
	Range (for Chinese blac	:k tea)		0.181-163	
	Ethiopia	5	a	0.098-0.100 ¹	5
	India	6	a	0.098-0.523	6
		1	a	$5.2 \pm 0.424 – 8.35 \pm 1.06$	1
		7	С	6.24-18.03	7
		3	С	0.1210.202	3
		4	d	7.21	4
	Range (for India black to	ea)		0.098-18.03	
	Indonesia	1	a	3.35 ± 0.919	1
	Iran	7	С	5.86-11.39	7
		8	d	2.85 (0.24)	8
	Range (for Iranian black	(tea)		2.61–11.39	
	Kenya	3	С	0.32	3
	Kenya/Malawi	1	a	7.43 ± 0.601	1
	Pakistan	9	d	$0.6 \pm 0.01 - 5.4 \pm 0.8$	9
	Range (for Pakistani bla		~	0.59-6.20	-
	Russia	3	С	0.113	3
	Sri Lanka	10	a	0.113 0.011	10
	311 Latika	1	a	$4.93 \pm 0.742 - 7.88 \pm 0.318$	1
		11	a	30.0-43.5	11
		7	a C	6.23–12.63	7
		3	C	0.23-12.03	3
		4	d	5.98–11.14	4
	Range (for Sri Lankan b	•	u	0.15–43.50	4
	Thailand	12	•	0.059-0.433	12
		10	e		
	Turkey		a	$0.130 \pm 0.009 - 0.165 \pm 0.013$	10
		13	f	$2.83 \pm 0.70 - 17.97 \pm 0.17$	13
		14	e	0.039-0.253	14
	Dan on Wan Tankish black	3	С	0.057	3
	Range (for Turkish black			0.04–18.14	4.5
	Unknown	15	e	34.98	15
		16	a	9.69 ± 0.72	16
		3	С	0.211	3
	Range (black tea from u	inknown origins)		0.21–34.99	
Overall range (for blac		_		0.04433.20	_
Green	China	2	b	17.5–188.2	2
		17	a	$0.091 \pm 0.004 – 0.138 \pm 0.004$	17
		3	С	0.149–0.579	3
	Range (for Chinese gree			0.09–188.20	
	India	3	С	0.135	3
	Indonesia	3	С	0.209	3
	Japan	3	С	0.588	3
	Sri Lanka	11	a	37.68	11
	Turkey	10	a	0.152 ± 0.013	10
		13	f	12.50 ± 0.32	13
	Range (for Turkish gree	n tea)		0.14-12.82	

(Continued on next page)

Table 10. (Continued)

Type of made tea	Origin	Infusion condition*	Analytical instrument [†]	Concentration (mg L ⁻¹)	Reference(s) [‡]
	Unknown	15	e	34.98	15
		16	a	9.69 ± 0.60	16
		18	g	$0.237 \pm 0.03 – 2.602 – 0.431$	18
		3	c	0.13-0.25	3
	Range (green tea fro	om unknown origins)		0.13-34.98	
	Vietnam	3	С	0.245	3
Overall range (for gre	en tea)			0.09188.20	
Pu-erh	China	3	С	0.124	3
White	China	3	С	0.3	3

*In the column, the following bold digts denote the following points: 1, 2.0 g of tea leaves +80 mL of deionized boiling water, covered and steeped for exactly five minutes; 2, 1 g of tea sample (vegetal material) was placed inside a tea sachet (commercially available tea filter, made of unbleached paper). The sachet was introduced into a glass beaker, 100 mL of boiling ultrapure water were added and the beaker was allowed to stand for five minutes; 3, 1 g tea samples were carefully weighed into glass beakers. Fifty milliliters of boiled distilled water was poured on it and covered with watch glasses for five minutes. Infusion was then filtered; 4, 80 cm³ of boiling deionized water to 2.0 g of broken tea leaves and infused for five minutes; 5, 2 g of tea leaves added in to the boiling water and boil for five minutes, then cooled for five minutes and filtered to 100 mL; 6, not available; 7, 45 mL hot distilled water to 1 g tea and five minutes infusion; 8, 50 mL of boiling deionized water was added to 5 g tea sample and kept at 80°C on a water bath for 15 minutes. At the end of the infusion period, the extract was filtered; 9, boiling 100 mL of deionized water and pouring the boiling water over duplicate (2.0 g) tea sample; 10, 1.00 g of tea infused for five minutes with 50 mL boiled distilled water; 11, 100 mL of hot distilled water was added to 2 g tea and infused for five minutes at room temperature; 12, 100 mL of boiling deionized water was added to one bag or 2.0 g of tea sample and left at room temperature for five minutes; 13, 100 mL of hot distilled water was added to 2 g of black tea sample. The mixture left to cool at room temperature for five minutes; 14, 200 mL of boiling ultrapure water 2 g black tea sample in a beaker, and infused for 15 minutes; 15, 1.5 g tea was infused for three minutes in a cup with 200 mL boiling water and kept for three minutes covered with a watch glass; 16, 3 g of made tea sample was transferred into the glass baker. About 80 mL of redistilled water was added to it. Then th

†In the column, the following letters denote the following points: a, FAAS (flame atomic absorption spectrometry); b, HR-CS FAAS (high-resolution-continuum source flame atomic absorption spectrometry); c, ICP-AES (inductively coupled plasma atomic emission spectrometry); d, ETAAS (electrothermal atomic absorption spectrometry); e, ICP-MS (inductively coupled plasma-mass spectrometry); f, ICP-OES (inductively coupled plasma optical emission spectrometry); g, HR-SF-ICPMS (high-resolution sector field inductively coupled mass spectrometry).

[‡]In the column, the following digits denote the following references: 1, Dambiec et al., 2013; 2, Paz-Rodríguez et al., 2015; 3, Street et al., 2006; 4, Polechońska et al., 2015; 5, Gebretsadik and Chandravanshi, 2010; 6, Chand et al., 2011; 7, Salahinejad and Aflaki, 2010; 8, Ziarati et al., 2013; 9, Jalbani et al., 2007; 10, Aksuner et al., 2012; 11, Lasheen et al., 2008; 12, Nookabkaew et al., 2006; 13, Görür et al., 2012; 14, Sofuoglu and Kavcar, 2008; 15, Milani et al., 2015; 16, Memić et al., 2014; 17, Gallaher et al., 2006; 18, Petit et al., 2013.

 $100~\mathrm{mL}^{-1}$. They reported that 32.15% of total Zn was released through tea infusion. Sofuoglu and Kavcar (2008) reported that 39.2–253 $\mu\mathrm{g}~\mathrm{L}^{-1}$ Zn was infused from black tea samples available in Turkey. The mean Zn contents in green tea, black tea and Oolong tea infusions available in Taiwan prepared from 2 g tea leaves was infused with 100 mL boiling water for 10 minutes were 7.6, 0.2, and 1.8 $\mu\mathrm{g}~100~\mathrm{mL}^{-1}$, respectively, which is equal to 60.7, 9.6, and 27.5% of total Zn, respectively (Shen and Chen, 2008).

Measured Zn content in black tea infusion and brewed tea were ranged from 0.107 ± 0.006 to 0.121 ± 0.011 mg L⁻¹ and 37.7 ± 2.1 to $47.3 \pm 2.0\%$ respectively (Pohl and Prusisz, 2007). However for green tea, measured Zn contents were ranged from 0.121 \pm 0.006 to 0.153 \pm 0.003 mg L⁻¹ and 35.4 \pm 1.8 to 44.1 \pm 0.9%, respectively for tea infusion and brewed tea. Among seven tea samples, the measured Zn content in tea infusion ranged from 30.0 to 43.5 $\mu g g^{-1}$ when tea infusion was produced from 2 g tea with 100 mL of hot distilled water for five minutes (Lasheen et al., 2008). According to Malik et al. (2008), the measured Zn contents in tea infusions prepared from 1 g of tea infused with 50 mL boiled distilled water for 15 minutes using 15 tea samples that originated from different tea growing countries were ranged from 0.070 \pm 0.004-0.441 \pm 0.006 mg L⁻¹. The amount of Zn in four Nigerian black tea infusions prepared from one g of tea boiled in 50 mL of distilled water for 10 minutes were varied between 0.03 \pm 0.04 and 0.80 \pm 0.4 μ g L⁻¹ with a mean of 0.28 \pm 0.23 μ g L⁻¹ (Achudume and Owoeye, 2010). Fernández et al. (2002) prepared infusions

from 20 green and 24 black teas available in Spanish market and found that Zn contents ranged from 0.08 to 0.27 mg L^{-1} (mean: 0.17 mg L^{-1}) and 0.08 to 0.31 mg L^{-1} (mean: 0.19 mg L^{-1}) in three and five minutes green infusion, respectively. For the 24 black teas infusion, measured Zn contents ranged from 0.19 to 0.50 mg L^{-1} (mean: 0.29 mg L^{-1}) and 0.29 to 0.12 mg L^{-1} (mean: 0.20 mg L^{-1}) after three and five minutes of infusion, respectively. Zinc contents in three black tea infusions are in the range of 0.080–0.10 mg L^{-1} , which is equivalent to 1.45–12.24% of total Zn in made tea (Chand et al., 2011). The concentration of Zn in six moderately extracted green tea infusion are ranged from 20 to 55% (Islam and Ebihara, 2012).

Among the five black and green tea samples found in Bosnia and Herzegovina, Zn content did not vary significantly, but the content in tea infusion ranged from 9.69 \pm 0.72-9.69 \pm $0.60 \ \mu g \ g^{-1}$ when 3 g of made tea was infused with 80 mL of warm water for five minutes (Memić et al., 2014). Görür et al. (2011) reported that Zn content in tea infusion and tea brew ranged from 2.83 \pm 0.70 to 17.97 \pm 0.17 mg g⁻¹ (mean: 6.36 \pm 0.41 mg g $^{-1})$ and from 6.74 \pm 0.80 to 33.39 \pm 0.34 mg g $^{-1}$ (mean: $12.07 \pm 0.39 \text{ mg g}^{-1}$), respectively. Average Zn content in 11 tea infusions that originated from Iran, India and Sri Lanka was 6.30 μ g g⁻¹ (Salahinejad and Aflaki, 2010). Gebretsadik and Chandravanshi (2010) reported that Zn transfer into its infusion from a range of teas from the world were ranged from 0.057 to 0.288 mg L⁻¹. Measured Zn concentrations that ranged from 3.35 \pm 0.919 to 8.35 \pm 1.06 μ g g⁻¹ found in tea infusions when prepared from 2 g tea leaves and steeped for



Table 11. Contribution of tea consumption to micronutrients contents in human body.

Elements	Average daily requirement per person (mg)*	Average daily amount (mg) consumed from tea infusion (10 g tea with 100 mL water) for three times)	Proportion of the required amounts (%)
Boron	10–28 ^a	0.10-24.9	1.0-88.9
Cobalt	0.1 μg ^b 2 – 5 ^b	0.01–0.06 $\mu { m g}$	10–60
Copper	2 – 5 ^b	0.04-4.89	2.0-97.8
Iron	12-15 ^b	8.0-13.49	67.8-89.9
Manganese	3-9 ^b	2.13-7.87	71.0-87.4
Molybdenum	0.15-0.5 ^b	0.02-0.17	13.3–34
Zinc	10-15 ^b	3.49-12.46	34.9-83

^{*}aMurray and Schlekat (2004).

exactly five minutes with 80 mL hot water (Dambiec et al., 2013). Kronborg (2013) prepared tea infusions from loose tea and tea bag using 1 g tea infused for five minutes with 100 mL ultrapure boiling water from 75 tea samples comprising of 40 samples from Norwegian supermarkets and 30 from the local tea of Polish supermarket. A comparatively higher amount of Zn was extracted from tea bags with measured Zn range of $45.3-120~\mu {\rm g~L}^{-1}$ (mean: $74.8 \pm 27.2~\mu {\rm g~L}^{-1}$) and $57.2-262\mu {\rm g}$ L^{-1} (mean: 132 \pm 45 μ g L^{-1}) for Poland and Norway, respectively. Measured Zn content in loose teas from Poland and Norway ranged from 34.3 to 90.3 $\mu g L^{-1}$ (mean: 59.6 \pm 17.5 $\mu g L^{-1}$) and 23.2 to 147 $\mu g L^{-1}$ (mean: 67.4 \pm 33.2 μg L^{-1}), respectively.

Health-related issues

Since tea plant has a strong potential to uptake and accumulate several micronutrients from soil, tea infusion may serve as dietary source of different micronutrients for human. However, strong debates are also there regarding safety of tea drinking by taking into consideration of such essential micronutrients accumulation in human body. Although micronutrients are essential for plant growth, they may be phytotoxic at high concentrations (Brun et al., 2001). Karak and Bhagat (2010) noted that over consumption of micronutrients from beverages is detrimental to human health. Therefore, micronutrient content in tea infusion must be traced and intake should be controlled on daily basis (Nookabkaew et al., 2006). For this reason, we calculated the percent daily micronutrients intake requirement of human based on the available data on micronutrient contents in tea infusion considering it as a serious health related issue. Calculation was done on the basis of Indian standard procedure. To generate the data, micronutrients in tea infusion was calculated from the available literatures, where 10 g made tea was infused with 100 mL distilled water for five minutes with intermittent stirring. Contribution of tea consumption (single time infusion of 10 g tea with 100 mL water and three times consumption per day) for micronutrient contents in human body has been tabulated in Table 11. The proportion of the required amounts of micronutrients for human from different types of teas were found as 1.0-88.9% for B, 10-60% for Co, 2.0–97.8% for Cu, 67.8–89.9% for Fe, 71.0–87.4% for Mn, 13.3-34% for Mo, and 34.9-83% for Zn (Table 11). Furthermore, Jin et al. (2008) concluded that people who consume large amounts of tea (e.g., 10 g of tea leaves per day) may not necessarily be at any risk since the daily intake of Cu from such tea drinking is less than 0.4 mg. This is true because, the

recommended daily consumption of Cu for adults as issued by the US Air Force, is 1.5-3.0 mg (US AF, 1990). Welna et al. (2012) reported that when total concentrations of elements in tea infusions are measured and considered, it appears that the intake of 1 L of tea per day may contribute 2.0-6.4% of Cu, 89.5-290% of Mn, and 1.3-2.1% of Zn to the respective RDAs of these elements. However, the actual uptake and absorption of these elements, established on the basis of in vitro bioavailability assay under simulated gastrointestinal digestion conditions in the human body and expressed as percentages of RDA values, are much lower, that is, 0.9-2.9% Cu, 45.8-116% Mn, and 0.4-0.7% Zn (Mehra and Baker, 2007; Moghaddam et al., 2008; Welna et al., 2012). Ashraf and Mian (2008) reported that a single serving of tea can be regarded as a rich absorbable source of Mn only. In the case of Cu and Zn, it cannot be recognized as such (Fernandez et al., 2002; Gallaher et al., 2006; Welna et al., 2012). Polechońska et al., (2015) suggested that the amount of trace elements extracted into the tea infusions depends principally on whether the compound is strongly bound to the matrix or more soluble in the solution used. For example, B and Mo, were poorly soluble, therefore, the mean concentrations of these micronutrients in infusions were very less. Szymczycha-Madeja et al. (2012) classified elements into three groups based on the extraction efficiencies assessed for different types of tea. Among the micronutrients, Szymczycha-Madeja et al. (2012) reported that Co is highly extractable (>55% of total Co contents in made tea), Cu, Mn, and Zn are moderately extractable (20-55% of individual micronutrients in made tea) and Fe as well as Mo are poorly extractable (<20% of individual micronutrients in made tea). Gallaher et al. (2006) reported that Cu, Fe, Mn, or Zn content in Chinese green tea were relatively low in the tea infusions when deionized water was used for their study to ensure that water was not contributing to the mineral content of the infusions. However, as consumers are not likely to use deionized water to make any hot beverage, those concerned with personal intake of the various minerals would do well to know the mineral content of their local tap water (Gallaher et al., 2006). Therefore, the various water sources have the potential to increase the mineral content present in tea infusions because the mineral content is not wholly dependent on extraction from tea leaf and water from various sources containing different elemental concentrations and this may affect elemental concentrations in the tea infusion (Mehra and Baker, 2007).

According to Seenivasan et al. (2008), a database is essential to fix the tolerance limit in Indian tea as there is only the PFA limit for Cu is there in black tea (150 mg kg⁻¹). Falahi and

^bFAO/WHO (2001).

Hedaiati (2013) reported that Cu concentration in Iranian black teas were well below the upper limits stated by various countries and seems to be comparable with the values reported from different countries (Karak and Bhagat, 2010). The authors concluded that Cu contamination in tea could mainly originate from the rolling machine and fungicides. According to Jin et al. (2008), the contribution of tea from drinking three cups daily to total exposure only in the case of Mn is substantial. The values of calculated daily intake of Mn from three cups of tea provide 62.2% of the daily human requirements for bagged tea and 33.9% for leaf tea Because of the homeostatic control that humans maintain over Mn, it is generally not considered to be very toxic when ingested with the diet. The authors also stated that Although food is regarded as the major source of exposure to Fe for the general population, the calculated daily intake of Fe from three cups of tea provide only about 0.22% of the daily human requirement for this metal. Similarly, the calculated daily intake of Zn from three cups of tea was very low and provided only 0.60% of the daily requirement. Aksuner et al. (2012) reported that the elements in the tea infusions can be arranged in the following decreasing order: Mn, Fe, Zn, and Cu and Ni. On their study, Mn level was found to be higher in black tea infusions when compared to the green tea infusions. According to Karak and Bhagat (2010), Mn is the only element with a significant dietary amount in tea, especially in black tea and found 40% of Mn content in bioavailable form under simulated intestinal conditions. Therefore, considering consumption of a few cups of tea per day, the daily dietary intake of 2-5 mg of Mn may be provided (Mehra and Baker, 2007; Aksuner et al., 2012). The speciation of Mn forms in tea infusions showed that 95% was in the form of Mn(II), and the remaining 5% was totally organic bound Mn (Özcan et al., 2008; Welna et al., 2012). According to Mehra and Baker (2007), loose tea provides 35.5% of "available" manganese, as a percentage of the average daily dietary intake. Hence, tea drinking may be regarded as a major source of essential dietary manganese. However, 115.5% "available" manganese from the Tetley tea bag sample is of some concern as high manganese in the diet can lead to long-term toxicity. They further concluded that as their finding is based on only one tea bag sample, further studies are needed to confirm the finding and its related consequences. Mehra and Baker (2007) reported that "available" Cu from drinking 1 g of tea per day provides 2.88% (loose tea) and 2.39% (tea bag tea) of the average daily dietary intake. Compared to Mn, contribution of tea toward the daily dietary intake of Cu is low, which may be attributed to low concentrations of Cu in tea. It is also important to note that unlike Mn, the difference in percentage "available" Cu between loose and tea bag tea is minimal. Dambiec et al. (2013) calculated daily intakes of Cu and Zn in black tea samples consumed in Poland and these values provide 0.43% of the daily requirements of human body for Zn and 53.5% for Mn. Therefore, the contribution of tea to total exposure only in case of Mn is significant. Cao et al. (2010) reported that concentrations of Zn and Cu in both Pu-erh fermented tea and raw tea were found to be far below the safety concentration limits of China and exposure to these elements in Pu-erh tea poses no noncarcinogenic risks (HQ<<1) to the inhabitants. However, it should be noted that the hazard index (HI) method is based on the additive assumption and the

results of cumulative health risk would be revised along with the clarification of the joint act mechanism of these metals in further studies. According to Sofuoglu and Kavcar (2008), 95th percentile HQ values for Mn in tea infusion was 0.11. Therefore, it may be concluded that according to population noncarcinogenic risk distributions, the risk levels were not significant for Mn. Memić et al. (2014) reported that, in general, the total content of all the determined micronutrients was higher in black tea than in green tea. Therefore, consuming a hot beverage of the analyzed black or green tea, humans can carry out their needs for essential elements and at the same time not expose themselves to the risks of excessive concentrations of toxic metals. In their study, the concentration of Mn is the highest of all the analyzed metals; about three times higher than the concentration of iron and about 30-300 times higher than the concentration of copper and zinc (Memić et al., 2014). Shen and Chen (2008) also reported that the HI of daily drinking of green tea, Oolong tea, and black tea was low and within the bounds of safety (<1). Mihara et al. (2014) examined the influence of tea consumption on human health and adopted the estimated daily dietary intake (EDDI) and calculated the EDDI values for the micronutrients by assuming the consumption of 15 g tea leaves in a day. All micronutrients had EDDI values that were lower than their respective upper tolerable intake or adequate intake except Mn. They concluded that in contrast, Mn is beneficial for hypertensive patients and, therefore, tea could be an important source of Mn.

Street et al. (2006) compared easily hot water soluble concentrations of Cu, Fe, Mn, and Zn and for 5-minute, 60-minute, and 24-hour infusions were prepared. The extractability of the elements were in the order of Cu > Zn > Mn > Fe. The proportions of the element contents in the infusion related to the respective total contents in leaves were 30 \pm 16% Cu, 26 \pm 10% Zn, 18 \pm 10% Mn, and 1.5 \pm 0.8% Fe respectively. The results confirmed that tea infusion can be an important dietary source of Mn. According to them, the total contents of metals in tea leaves differ according to the type of tea (green or black) and are probably influenced by many other factors, for example, soil properties, location, rainfall, altitude, genetic properties of the plant, age of the leaves, etc. The total content of Mn was much higher compared to the total contents of Cu, Fe, and Zn resulting in a relatively high concentration of Mn in infusions. Tea infusions cannot be seen as a dietary source of essential trace metals, except for Mn. Tea infusions could be a good dietary source of essential trace metals for humans. For many people, tea drinking may be a major source of dietary Mn (Wrobel et al., 2000), as it is evident also in comparison with other beverages such as hibiscus and maté. However, the percentage of elements leached into the infusion is strongly related to the tannin content in the beverage; with lower tannin levels, better leaching was observed. Lv et al. (2013a) reported that the dietary intake of Cu was below the safe limits recommended by various authorities based on a 70 kg individual consuming 15 g of Pu-erh tea daily or 105 g Pu-erh tea weekly. This suggested that under the current dietary intake, there are no possible health risks to Pu-erh tea drinking consumers with respect to Cu. According to Mehra and Baker (2007), drinking of 1 L tea infusion per day contributes 2.88% (from loose tea) and 2.39% (from teabag tea) of the average daily dietary intake. However,



tea for consumers may be prepared with water from various sources containing diferent elemental concentrations and this may afect elemental concentrations in the tea infusion. In the present study, tea infusions were prepared using distilled water (Mehra and Baker, 2007). Lv et al. (2013a) also reported that Pu-erh loose tea and compressed tea had similar mean Cu level. The authors further concluded that 105 g Chinese Pu-erh tea does not pose any health risks to humans when health risks from consumption of Chinese Pu-erh was estimated through weekly dietary intake.

Conclusions

A wide range of concentrations of selected micronutrient in made tea was found in different literatures. The difference in micronutrient contents of made tea could be attributed to the tea being produced in different tea growing areas of the world having high variation in micronutrient contents as well as its application strategies, which resulted in differential micronutrient uptake by tea plants. In fact, analysis report of micronutrients in tea infusions is not sufficiently available. Micronutrient status in made tea is rather inappropriate to predict the possible nutritional effects of habitual drinking of this beverage. Therefore, micronutrient status in tea infusions is of paramount importance over that of total micronutrient content in made tea. Furthermore, speciation of micronutrients in tea infusion is rather more perfect one to predict the possible nutritional effects of habitual drinking of this beverage as it reflects the uptake of elements in the gastrointestinal tract. Actually, these species may regulate the actual uptake of elements by the human body; however, this is an issue not firmly established in the available literatures. More studies are very much essential for B, Co, and Mo in made tea as well as tea infusion. Furthermore, different methods of processing as well as storage of made tea may also be an influencing factor of micronutrient contents in tea infusion, which is scanty. From the results it can also be concluded that consumption of three cups of tea infusion per day does not have any adverse effect on human health with respect to the referred micronutrients rather got beneficial effects to human. It has been further observed that even though tea plant grows in more than 40 countries' existing literatures mostly focus on selected countries like China, India, Japan, Sri Lanka, and Turkey. Despite of having significant amount of research findings, more research is required to comprehend the science of accumulation of micronutrients by tea plants, specification of uptake of micronutrients and the effect of micronutrients on human health from regular consumption of tea in all the tea growing countries.

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