



## Review

## Trace elements in tea leaves, made tea and tea infusion: A review

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## ABSTRACT

Tea (*Camellia sinensis* L) is one of the most widely popular nonalcoholic beverage, consumed by over two-thirds of the world's population due to its medicinal, refreshing and mild stimulant effects. Mainly four types of made tea viz., black or red, oolong, green and white are used for tea infusion (water extract from fermented tea leaf) worldwide. Tea plays a major role in terms of the intake of a number of nutritional trace elements in humans. Besides essential macro- and microelements, experimental studies have demonstrated that the accumulation of significant amount of excess nonessential trace elements in tea leaves may eventually increase the metal body burden in humans. Different literatures have indicated that among different trace elements, levels of aluminium (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), fluoride (F) manganese (Mn), and nickel (Ni) in different tea infusions were 0.06–16.82 mg L<sup>-1</sup>, trace–1.53 µg L<sup>-1</sup>, trace–0.79 µg L<sup>-1</sup>, below detectable limit–43.2 µg L<sup>-1</sup>, 0.02–40.0 mg L<sup>-1</sup>, 0.2–4.54 mg L<sup>-1</sup>, 0.1–250 mg L<sup>-1</sup> and BDL–0.16 mg L<sup>-1</sup>, respectively. It has also been reported that in some of the tea infusions, toxic metals exceed the maximum permissible limits stipulated for different countries. In the present review, an attempt has been made to update and evaluate the knowledge of the presence of some selected trace elements in tea leaves, made tea and tea infusion, based on the available literature. Existing reports suggested that the presence of trace elements in green tea is lower than the black tea in most cases. However, the available literatures still appear to have some limits and may need more detailed investigations before reaching the conclusions.

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## 1. Introduction

Tea plant is a perennial evergreen plant with three races viz. *Camellia sinensis*, *Camellia assamica* and *Cambodiensis* (Wight, 1962). Tea plant is grown in more than 45 countries spread over all the continents except North America within the latitudinal range of 45° N to 34° S (Barua, 2008). Tea plant grows best in tropical and subtropical areas having adequate rainfall, good drainage and acid soils (Barua, 2008). Leaf tea signifies two leaves and the terminal apical bud of a growing shoot of a tea bush. Made teas are classified into four main types like black tea, green tea, oolong tea and brick tea based on their respective manufacturing techniques.

Originating from China, tea has gained the world's taste in the past 2000 years as one of the most popular nonalcoholic beverage. The economic and social interest of tea is easily understood from the fact that about 18 to 20 billion cups of tea are consumed daily in the world (Marcos, Fischer, Rea, & Hill, 1998).

Tea drinking is associated with the reduction of serum cholesterol, prevention of low density lipoprotein oxidation, decreased risk of cardiovascular disease and cancer (Chung, Schwartz, Herzog, & Yang, 2003). Polyphenols are the most biologically active group of tea components which have antioxidative, antimutagenic and anticarcinogenic effects (Yao et al., 2004). In tea, there are other compounds which are beneficial to human health like fluoride, caffeine and essential minerals (Cabrera, Gimenez, & Lopez, 2003). A recent study has demonstrated that the presence of trace elements (metal with a high relative atomic mass) in tea is due to tea plants being normally grown in highly acidic soils, where trace elements are potentially more bioavailable for root uptake (Han, Zhao, Shi, Ma, & Ruan, 2006). Trace element contents of tea may have both beneficial and adverse effects on human health. The regular consumption of tea can contribute to the daily dietary requirements of some of these elements (Deng, Tao, Li, He, & Chen, 1998; Xie, von Bohlen, Klockenkämper, Jian, & Günther, 1998). Owing to the importance of minerals present in tea, many studies were carried out to determine their levels in tea leaves and their infusions (Fernández, Pablos, Martín, & González, 2002; Fung, Zhang, Wong, & Wong, 1999; Matsuura, Hokura, Katsuki, Itoh, & Haraguchi, 2001). Trace elements content may have adverse effect on human health, e.g. accumulation of aluminium (Al) in tea infusion is associated with Alzheimer's disease (Koch, Pougnet, de Villiers, & Monteagudo, 1988). Besides Al, copper (Cu) contamination in tea leaves remains a concern and practices should be used to ensure food safety from excessive Cu contamination. In similar way other trace element (viz. arsenic, As; cadmium, Cd; chromium, Cr; fluoride, F; lead, Pb; manganese, Mn; and nickel, Ni) contents of tea have been investigated in many South Asian countries and the countries where tea is a traditional drink (Powell,

Greenfield, Parkes, Nicholson, & Thompson, 1993). Determination of trace elements in tea is important from two aspects (a) to judge their nutritional value and (b) to guard against any probable ill-effects. Even though drinking tea is a popular refreshment in many countries, the potential nonessential trace element intake from drinking tea has not been included in the dietary intake assessments.

Keeping in view the growing awareness and concerns about trace elements in tea, we attempted to make a systematic review of the recent findings on different trace elements (Al, As, Cd, Cr, Cu, F, Mn, Ni and Pb) in tea leaves, made tea and tea infusion based on the available literatures.

## 2. Trace elements in tea

A selected summarized data on discussed trace elements (Al, As, Cd, Cr, Cu, F, Mn, Ni and Pb) in fresh tea leaves, made tea (black, green, and oolong tea) and tea infusion with infusion conditions are presented in Tables 1–3 respectively.

### 2.1. Aluminium

It has been observed that Alzheimer's disease (AD) is associated with the presence of aluminium (Al) in barely mostly, derived from Al containing food consumption over a prolonged period of time (McLachlan, 1995; Deng et al., 2000) and therefore, the high Al content in tea may be a matter of concern (Flaten, 2002). Aluminium salts present in tea garden soils dissociate at pH < 5.5 and produce Al<sup>3+</sup> in soil solution and form complexes with phosphate (AIP<sub>x</sub>) at the root surface of tea (Wong, Fung, & Carr, 2003). AIP<sub>x</sub> complexes have less charge, or even are neutral molecules (with or without organic acids) which are absorbed or transported passively into the tea plant. Malic acid would chelate Al<sup>3+</sup> at the root and transport it to the young leaves while releasing P for plant absorption. Some Al<sup>3+</sup> stays in the root, while excess Al<sup>3+</sup> in the young leaves would form strong complexes with organic acids or polyphenols and transport these to the older leaves (Hayacibara, Queiroz, Tabchoury, & Cury, 2004).

#### 2.1.1. Aluminium in tea leaves

Chenery (1955) was the first who reported the accumulation of unusually large quantities of Al (5000 to 16,000 mg kg<sup>-1</sup>) by tea plant. It was also reported that the older tea leaves contained much more aluminium than young leaves (Chenery, 1955). Matsumoto, Hirasawa, Morimura, and Takahashi (1976) reported that tea is one of the few plants accumulating aluminium, making tea a major source of dietary aluminium intake. Heavy drinking of tea may make more than double of

**Table 1**  
Trace elements in fresh tea leaves.

| Country of origin       | Sample analyzed     | Number of samples | Concentration range (mg kg <sup>-1</sup> ) | Reference                              |
|-------------------------|---------------------|-------------------|--|--|
| <i>Aluminium (Al)</i>   |                     |                   |  |  |
| Ceylon                  | Flush leaves        | 8                 | 66–209                                     | Chenery (1955)                         |
| Ceylon                  | Mature leaves       | 8                 | 3480–11,300                                | Chenery (1955)                         |
| China                   | All leaves          | 5                 | 2650–11,500                                | Chenery (1955)                         |
| India                   | All leaves          | 6                 | 8360–10,000                                | Chenery (1955)                         |
| Kenya                   | Young leaves        | 13                | 115–1250                                   | Chenery (1955)                         |
| Tanzania                | Young leaves        | 20                | 13–10,800                                  | Chenery (1955)                         |
| China                   | Fresh tea leaves    | –                 | 300–1600                                   | Chen (1984)                            |
| India                   | All leaves          | 40                | 400–5813                                   | Natesan and Ranganathan (1990)         |
| Japan                   | All leaves          | 7                 | 72–8290                                    | Nagata, Hayatsu and Kosuge (1992)      |
| China                   | Mature leaves       | 13                | 873–3637                                   | Dong, Xie, Du, Liu and Wang (1999)     |
| China                   | Tea leaves          | 13                | 156–4381                                   | Xie et al. (2001)                      |
| China                   | Young tea leaves    | 5                 | 2311–11,981                                | Fung et al. (2003)                     |
| China                   | Bud with two leaves | 5                 | 453–1518                                   | Shu et al. (2003)                      |
| China                   | Tea leaves          | 2                 | 1510–3364                                  | Fung and Wong (2004)                   |
| China                   | Young leaves        | 12                | 370–1526                                   | Xie et al. (2007)                      |
| China                   | Tea leaves          | 8                 | 2034–3322                                  | Chen et al. (2009)                     |
| China                   | Young tea leaves    | 4                 | 381–2152                                   | Fung et al. (2009)                     |
| China                   | Fresh tea leaves    | 8                 | 1080–2020                                  | Cao, Qiao, Zhang and Chen (2010)       |
| <i>Arsenic (As)</i>     |                     |                   |  |  |
| China                   | Young shoots        | 5                 | 0.021–0.073                                | Shi et al. (2008)                      |
| China                   | Fresh tea leaves    | 8                 | 0.024–0.066                                | Cao et al. (2010)                      |
| <i>Cadmium (Cd)</i>     |                     |                   |  |  |
| India                   | All leaves          | 40                | 0.10–0.25                                  | Natesan and Ranganathan (1990)         |
| Turkey                  | Tea leaves          | 3                 | 0.05–1.27                                  | Tokalioglu and Kartal (2004)           |
| China                   | Young shoots        | 5                 | 0.02–0.10                                  | Shi et al. (2008)                      |
| China                   | Tea leaves          | 8                 | 0.03–0.08                                  | Chen et al. (2009)                     |
| China                   | Fresh tea leaves    | 8                 | ND–0.01                                    | Cao et al. (2010)                      |
| <i>Chromium (Cr)</i>    |                     |                   |  |  |
| India                   | All leaves          | 40                | 1.0–2.0                                    | Natesan and Ranganathan (1990)         |
| Turkey                  | Tea leaves          | 3                 | 1.16–3.41                                  | Tokalioglu and Kartal (2004)           |
| Gillan Province of Iran | Tea leaves          | 60                | Nil–0.24                                   | Ebadi, Zare, Mahdavi and Babaee (2005) |
| <i>Copper (Cu)</i>      |                     |                   |  |  |
| India                   | All leaves          | 40                | 66–127                                     | Natesan and Ranganathan (1990)         |
| Turkey                  | Tea leaves          | 3                 | 5.83–10.7                                  | Tokalioglu and Kartal (2004)           |
| China                   | Tea leaves          | 8                 | 9.68–18.82                                 | Chen et al. (2009)                     |
| China                   | Fresh tea leaves    | 8                 | 9.4–13.8                                   | Cao et al. (2010)                      |
| <i>Fluoride (F)</i>     |                     |                   |  |  |
| China                   | Tea leaves          | 19                | 340–808                                    | Fung et al. (1999)                     |
| China                   | Tea leaves          | 2                 | 128–374                                    | Fung and Wong (2004)                   |
| China                   | Young leaves        | 12                | 49–313                                     | Xie et al. (2007)                      |

**Table 1** (continued)

| Country of origin     | Sample analyzed  | Number of samples | Concentration range (mg kg <sup>-1</sup> ) | Reference                                |
|-----------------------|------------------|-------------------|--|--|
| <i>Lead (Pb)</i>      |                  |                   |  |  |
| India                 | All leaves       | 40                | 3.1–14.5                                   | Natesan and Ranganathan (1990)           |
| Iran                  | Tea leaves       | 60                | 0.531–5.3                                  | Ebadi et al. (2005)                      |
| Turkey                | Tea leaves       | 3                 | Not detectable–5.72                        | Tokalioglu and Kartal (2004)             |
| China                 | Fresh tea leaves | 8                 | 0.31–3.42                                  | Cao et al. (2010)                        |
| China                 | Tea leaves       | 51                | 0.59–4.49                                  | Jin, Zheng, He, Zhou and Zhou (2005)     |
| China                 | Tea shoots       | –                 | 0.30–1.90                                  | Han, Shi, Ma, Ruan and Zhao (2007)       |
| Ethiopia              | Tea leaves       | 19                | NIL  | Yemane, Chandravanshi and Wondimu (2008) |
| <i>Manganese (Mn)</i> |                  |                   |  |  |
| India                 | All leaves       | 40                | 174–489                                    | Natesan and Ranganathan (1990)           |
| Kenya                 | Young leaves     | 13                | nd–2300                                    | Chenery (1955)                           |
| Turkey                | Tea leaves       | 3                 | 2617–3154                                  | Tokalioglu and Kartal (2004)             |
| <i>Nickel (Ni)</i>    |                  |                   |  |  |
| India                 | All leaves       | 40                | 13–14                                      | Natesan and Ranganathan (1990)           |
| Turkey                | Tea leaves       | 3                 | 6.60–11.7                                  | Tokalioglu and Kartal (2004)             |

an individual's intake of aluminium (Flaten, 2002). Polyphenolic compounds, especially flavanoids present in tea are important for the binding of metals such as Al, Fe and Cu (Erdemoğlu, Pyrzyniska, & Güçerc, 2000). Wong, Zhang, Wong, and Lan (1998) observed that under acidic conditions, Al is accumulated in tea plants mainly in the leaves, especially in older leaves (average 5600 mg kg<sup>-1</sup>) followed by young leaves (average 997 mg kg<sup>-1</sup>). An aluminium concentration in different tea varieties growing in a tea plantation in Hong Kong was found to be 2500 to 13,000 mg kg<sup>-1</sup> (Wong et al., 2003). It was further observed that the Chinese small leaf variety possessed a significantly higher amount of Al in leaf tissues (4150–11,981 mg kg<sup>-1</sup>) compared to the Chinese large leaf (2311–8362 mg kg<sup>-1</sup>) and Assam varieties (8255 mg kg<sup>-1</sup>). In an extreme case, it has been observed that aluminium contents in old tea leaves exceeded 30,000 mg kg<sup>-1</sup> in Japan (Konishi, Miyamoto, & Taki, 1985; Matsumoto et al., 1976). Jayman and Sivasubramaniam (1980) reported that the accumulation of Al in older tea leaves could be due to a mechanism of its tolerance by tea as older leaves may possess up to ten times the levels of Al found in younger leaves. Recently, it has been demonstrated that young leaves of the Kenyan variety had a significantly high Al level of 2152 mg kg<sup>-1</sup>, and the young leaves of the Chinese variety had the lowest Al of 381 mg kg<sup>-1</sup> when plants were grown in Lantau Peak located on Lantau Island in Hong Kong (Fung, Carr, Poon, & Wong, 2009). Their findings were also in agreement with the previous findings of Chenery (1955).

### 2.1.2. Aluminium in made tea

Marcos et al. (1998) analyzed fifteen types of tea from ten different countries like Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania and Zimbabwe using nitric acid digestion method. From this finding they concluded that Al contents in made tea leaves differed according to the types of tea (green or black) and geological conditions. Mokgalaka, McCrindle, and Botha (2004) evaluated the slurry nebulization method as an alternative method for the analysis of Al in tea samples from different origins (Two Ceylon tea blends and Chinese green tea). They found 52.8 to 1036 mg kg<sup>-1</sup> Al in all tea samples. The total Al contents in made tea was found to be between 144

**Table 2**  
Trace elements in made tea.

| Country of origin  | Sample analyzed               | Number of sample(s)   | Concentration range (mg kg <sup>-1</sup> ) | Reference                                       |
|--|-------------------------------|---|--|---|
| <i>Aluminium (Al)</i>  |                               |   |  |   |
| Not known  | Not known                     | –   | 4.5–6.0                                    | Koch et al. (1988)                              |
| China  | Oolong tea                    | 1   | 797  | Marcos et al. (1998)                            |
| China and Japan  | Green tea                     | 2   | 652–1161                                   | Marcos et al. (1998)                            |
| Not known  | Made tea                      | 4   | 835–1330                                   | Erdemoğlu et al. (2000)                         |
| China, Italy, Russia, and Syria  | Commercial black tea          | 9   | 12,352–27,162                              | Ferrara et al. (2001)                           |
| China, Japan, India, Kenya, and Sri Lanka  | Made tea                      | 46 (21 green, 23 black, and two instant teas)                             | 565.9–2560.4                               | Fernandez-Caceres et al. (2001)                 |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green, and oolong teas | 54  | 753.87–789.81                              | Moreda-Piñeiro et al. (2003)                    |
| China  | Brick tea                     | 4   | 3925–5705                                  | Shu et al. (2003)                               |
| China  | Green tea                     | 11  | 233–1141                                   | Shu et al. (2003)                               |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea                      | 18  | 564.4–2740                                 | Nookabkaew et al. (2006)                        |
| Ceylon, China, India, Indonesia, Japan, Kenya, Sri Lanka, Taiwan, Turkey, and Vietnam                                  | Made tea                      | 29 (13 green tea, 13 black tea, 2 semi-fermented and one white tea)       | 272–3260                                   | Street et al. (2007)                            |
| India, China, Sri Lanka, and other unknown countries   | Black and green teas          | 8   | 458–1307                                   | Mehra and Baker (2007)                          |
| India  | Black tea                     | 4   | 400–1000                                   | Mossion et al. (2008)                           |
| China and Indonesia  | Oolong tea                    | 4   | 280–382                                    | Malik et al. (2008)                             |
| India  | Black tea                     | 4   | 168–288                                    | Malik et al. (2008)                             |
| India and Japan  | Green tea                     | 4   | 211–353                                    | Malik et al. (2008)                             |
| India, China, and Indonesia  | White tea                     | 6   | 123–276                                    | Malik et al. (2008)                             |
| Australia, China, India, Indonesia, Kenya, Papua New Guinea, Rwanda, Sri Lanka, and Tanzania                           | Made tea                      | 75 (55 Chinese tea and 20 samples of Western Black tea and Earl Grey tea) | 224–2633                                   | Fung et al. (2009)                              |
| Ceylon, India, and Iran  | Black tea                     | 11  | 585–1546                                   | Salahinejad and Aflaki (2010)                   |
| Ceylon, China, Georgia, India, and Kenya   | Black tea                     | 36  | 479–2465                                   | McKenzie, Jurado, Fernando and de Pablos (2010) |
| China and Japan  | Green tea                     | 24  | 612–4074                                   | McKenzie et al. (2010)                          |
| China and Japan  | White tea                     | 28  | 140–1736                                   | McKenzie et al. (2010)                          |
| China and Taiwan   | Oolong tea                    | 20  | 273–1853                                   | McKenzie et al. (2010)                          |
| <i>Arsenic (As)</i>  |                               |   |  |   |
| China  | Black tea                     | 148   | Trace–4.43                                 | Han et al. (2005)                               |
| China  | Green tea                     | 536   | Trace–1.66                                 | Han et al. (2005)                               |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea                      | 18  | 0.010–0.238                                | Nookabkaew et al. (2006)                        |
| China  | Not specified                 | 328   | ND (not detectable)–2.00                   | Shi et al. (2008)                               |
| China  | Chinese tea                   | 47  | BDL–4.81                                   | Yuan et al. (2007)                              |
| Iran   | Black tea                     | 5   | 0.07–0.11                                  | Karimi et al. (2008)                            |
| <i>Cadmium (Cd)</i>  |                               |   |  |   |
| Japan  | Green tea                     | 139   | 0.013–0.098                                | Tsushida and Takeo (1977)                       |
| India  | Black tea                     | 5   | 0.27–0.35                                  | Natesan and Ranganathan (1990)                  |
| China and India  | Unknown                       | 12  | 0.04–0.33                                  | Pedersen, Mortensen and Larsen (1994)           |
| China  | Oolong tea                    | 1   | 0.088                                      | Marcos et al. (1998)                            |
| Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania, and Zimbabwe                             | Black tea                     | 12  | 0.024–0.161                                | Marcos et al. (1998)                            |
| China and Japan  | Green tea                     | 2   | 0.051–0.114                                | Marcos et al. (1998)                            |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green and oolong tea   | 54  | 2.22–2.39                                  | Moreda-Piñeiro et al. (2003)                    |
| Turkey   | Black tea                     | 6   | 0.05–1.27                                  | Tokalioglu and Kartal (2004)                    |
| China  | Not specified                 | 328   | ND (not detectable)–1.59                   | Shi et al. (2008)                               |
| Iran   | Black tea                     | 10  | 0.10–1.92                                  | Shokrzadeh et al. (2008)                        |
| Iran   | Black tea                     | 4   | n.d.*–0.76                                 | Salahinejad and Aflaki (2010)                   |
| India  | Black tea                     | 3   | n.d.*–0.77                                 | Salahinejad and Aflaki (2010)                   |
| Ceylon   | Black tea                     | 4   | n.d.*–0.65                                 | Salahinejad and Aflaki (2010)                   |
| <i>Chromium (Cr)</i>   |                               |   |  |   |
| Iran   | Black tea                     | 2   | 0.03–0.06                                  | Kasrai et al. (1977)                            |
| India  | Black tea                     | 5   | 2.0–2.7                                    | Natesan and Ranganathan (1990)                  |
| China and India  | Unknown                       | 12  | 1.4–3.3                                    | Pedersen et al. (1994)                          |
| China  | Oolong tea                    | 1   | 0.623                                      | Marcos et al. (1998)                            |
| Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania, and Zimbabwe                             | Black tea                     | 12  | 0.29–32.87                                 | Marcos et al. (1998)                            |

(continued on next page)

Table 2 (continued)

| Country of origin  | Sample analyzed  | Number of sample(s)                           | Concentration range (mg kg <sup>-1</sup> ) | Reference                                      |
|--|--|---|--|--|
| <i>Chromium (Cr)</i>   |  |   |  |  |
| China and Japan  | Green tea  | 2   | 0.45–0.99                                  | Marcos et al. (1998)                           |
| Turkey   | Black tea  | 6   | 1.16–2.87                                  | Tokalioglu and Kartal (2004)                   |
| Turkey   | Black tea  | 14  | <10–14.8                                   | Narin et al. (2004)                            |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea   | 18  | 0.205–10.54                                | Nookabkaew et al. (2006)                       |
| South India  | Black tea  | 12  | 2.95–7.59                                  | Seenivasan et al. (2008a)                      |
| Iran   | Black tea  | 4   | n.d. <sup>a</sup> –1.94                    | Salahinejad and Aflaki (2010)                  |
| India  | Black tea  | 3   | 0.98–1.83                                  | Salahinejad and Aflaki (2010)                  |
| Ceylon   | Black tea  | 4   | 0.94–2.55                                  | Salahinejad and Aflaki (2010)                  |
| <i>Copper (Cu)</i>   |  |   |  |  |
| Japan  | Green tea  | 139   | 4.7–11.1                                   | Tsushida and Takeo (1977)                      |
| India  | Black tea  | 5   | 130–141                                    | Natesan and Ranganathan (1990)                 |
| Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania, and Zimbabwe                             | Black tea  | 12  | 7.1–28.3                                   | Marcos et al. (1998)                           |
| China and Japan  | Green tea  | 2   | 31.5–32.3                                  | Marcos et al. (1998)                           |
| China  | Oolong tea   | 1   | 20.0                                       | Marcos et al. (1998)                           |
| China, Italy, Russia, and Syria  | Commercial black tea   | 9   | 377.5–602.5                                | Ferrara et al. (2001)                          |
| China, Japan, India, Kenya, and Sri Lanka  | Made tea   | 46 (21 green, 23 black, and two instant teas) | 7.6–37.5                                   | Fernandez-Caceres et al. (2001)                |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green and oolong tea  | 54  | 12.34–19.14                                | Moreda-Piñeiro et al. (2003)                   |
| Turkey   | Black tea  | 6   | 6.68–31.2                                  | Tokalioglu and Kartal (2004)                   |
| India and USA  | Black  | 22  | 4.4–35.5                                   | Kumar et al. (2005)                            |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea   | 18  | 3.08–22.42                                 | Nookabkaew et al. (2006)                       |
| India, China, Sri Lanka, and other unknown countries   | Black and Green tea  | 8   | 15.7–34                                    | Mehra and Baker (2007)                         |
| Turkey   | Black tea and green tea  | 10  | 6.4–13.1                                   | Soylak, Tuzen, Souza, Korn and Ferreira (2007) |
| India  | Black tea  | 4   | 22.1–66.5                                  | Malik et al. (2008)                            |
| India and Japan  | Green tea  | 4   | 23.1–36.5                                  | Malik et al. (2008)                            |
| China and Indonesia  | Oolong tea   | 4   | 15.1–25.8                                  | Malik et al. (2008)                            |
| India, China, and Indonesia  | White tea  | 6   | 17.6–31.6                                  | Malik et al. (2008)                            |
| Ceylon, India, and Iran  | Black tea  | 11  | 19.84–58.48                                | Salahinejad and Aflaki (2010)                  |
| Ceylon, China, Georgia, India, and Kenya   | Black tea  | 36  | 12–35                                      | McKenzie et al. (2010)                         |
| China and Japan  | Green tea  | 24  | 11–26                                      | McKenzie et al. (2010)                         |
| China and Japan  | White tea  | 28  | 10–26                                      | McKenzie et al. (2010)                         |
| China and Taiwan   | Oolong tea   | 20  | 4–12                                       | McKenzie et al. (2010)                         |
| <i>Fluoride (F)</i>  |  |   |  |  |
| China  | Oolong tea   | 3   | 170–423                                    | Fung et al. (1999)                             |
| China  | Green tea  | 3   | 217–344                                    | Fung et al. (1999)                             |
| China  | Green tea  | 11  | 49–104                                     | Shu et al. (2003)                              |
| China  | Brick tea  | 4   | 590–708                                    | Shu et al. (2003)                              |
| Tibet  | Brick tea  | 15  | 541.1–612.3                                | Jin et al. (2000)                              |
| China  | Made tea   | 5   | 170–878                                    | Wong et al. (2003)                             |
| <i>Lead (Pb)</i>   |  |   |  |  |
| Japan  | Green tea  | 139   | 0.11–1.93                                  | Tsushida and Takeo (1977)                      |
| India  | Black tea  | 5   | 6.0–7.6                                    | Natesan and Ranganathan (1990)                 |
| China and India  | Unknown  | 12  | 0.19–1.29                                  | Pedersen et al. (1994)                         |
| China and Japan  | Green tea  | 2   | 0.59–0.74                                  | Marcos et al. (1998)                           |
| Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania, and Zimbabwe                             | Black tea  | 12  | 0.18–2.18                                  | Marcos et al. (1998)                           |
| China  | Oolong tea   | 1   | 1.26                                       | Marcos et al. (1998)                           |
| China  | Green tea (n = 20), oolong (n = 5), black tea (n = 8), white tea (n = 2) | 39  | 0.3–8.3                                    | Xie et al. (1998)                              |
| China, Italy, Russia, and Syria  | Commercial black tea   | 9   | 0.0–240.1                                  | Ferrara et al. (2001)                          |
| Pakistan   | Made tea   | 14  | 14.8                                       | AL-Oud (2003)                                  |
| China  | Made tea   | 85  | 0.15–11.61                                 | Tang et al. (2003)                             |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green and oolong tea  | 54  | 0.18–1.12                                  | Moreda-Piñeiro et al. (2003)                   |
| Turkey   | Black tea  | 6   | 1.46–5.64                                  | Tokalioglu and Kartal (2004)                   |
| China  | Made tea   | 1225  | <0.2–97.9                                  | Han et al. (2006)                              |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea   | 18  | 0.060–53.89                                | Nookabkaew et al. (2006)                       |
| China  | Made tea   | 57  | 0.198–6.345                                | Qin and Chen (2007)                            |
| Saudi Arabia   | Black Tea  | 17  | 0.30–2.2                                   | Ashraf and Mian (2008)                         |

Table 2 (continued)

| Country of origin  | Sample analyzed   | Number of sample(s) | Concentration range (mg kg <sup>-1</sup> ) | Reference                       |
|--|---|---------------------|--|---------------------------------|
| <i>Lead (Pb)</i>   |   |                     |  |                                 |
| Iran   | Black tea   | 5                   | 1.37–2.31                                  | Karimi et al. (2008)            |
| Unknown  | Black tea   | 5                   | 0.664–5.430                                | Shokrzadeh et al. (2008)        |
| Iran   | Black tea   | 5                   | 8.381–15.480                               | Shokrzadeh et al. (2008)        |
| Ceylon, Iran, and India  | Black tea   | 11                  | 0.92–2.92                                  | Salahinejad and Aflaki (2010)   |
| <i>Manganese (Mn)</i>  |   |                     |  |                                 |
| Iran   | Black tea   | 2                   | 132.7–481                                  | Kasrai et al. (1977)            |
| India  | Black tea   | 5                   | 220–324                                    | Natesan and Ranganathan (1990)  |
| China  | Black tea   | 3                   | 930–1064                                   | Wang et al. (1993)              |
| China  | Black tea   | 4                   | 370–510                                    | Liu, Zhang, Liu and Wang (1995) |
| Africa, China, India, Japan, and Sri Lanka   | Black tea and Green tea   | 9                   | 730–880                                    | Lamble and Hill (1995)          |
| Bangladesh, Burundi, China, India, Japan, Kenya, Malawi, Sri Lanka, Tanzania, and Zimbabwe                             | Black tea   | 12                  | 405–1604                                   | Marcos et al. (1998)            |
| China  | Green tea ( <i>n</i> = 20), oolong( <i>n</i> = 5), black tea ( <i>n</i> = 8), white tea ( <i>n</i> = 2) | 39                  | 160–1500                                   | Xie et al. (1998)               |
| China  | Oolong tea  | 1                   | 785  | Marcos et al. (1998)            |
| China and Japan  | Green tea   | 2                   | 654–919                                    | Marcos et al. (1998)            |
| Not known  | Black tea   | –                   | 350–900                                    | Powell et al. (1998)            |
| Turkey   | Black tea   | 5                   | 1107–2205                                  | Ozdemir and Gucer (1998)        |
| China, Italy, Russia, and Syria  | Black tea   | 9                   | 4679–15,866                                | Ferrara et al. (2001)           |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green and oolong tea   | 54                  | 562.61–843.56                              | Moreda-Piñeiro et al. (2003)    |
| Turkey   | Black tea   | 6                   | 912–1354                                   | Tokalioglu and Kartal (2004)    |
| Turkey   | Black tea   | 14                  | 563.9–1081.6                               | Narin et al. (2004)             |
| India and USA  | Black   | 22                  | 79–768                                     | Kumar et al. (2005)             |
| India, China, Sri Lanka, and other unknown countries   | Black and Green tea   | 8                   | 300–1147                                   | Mehra and Baker (2007)          |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand  | Made tea  | 18                  | 229.4–1512                                 | Nookabkaew et al. (2006)        |
| Polish market  | Black tea   | 3                   | 286–1290                                   | Pohl and Prusisz (2007)         |
| Polish market  | Green tea   | 3                   | 618–1440                                   | Pohl and Prusisz (2007)         |
| India  | Black tea   | 4                   | 392–589                                    | Malik et al. (2008)             |
| China, and Indonesia   | Oolong tea  | 4                   | 393–1198                                   | Malik et al. (2008)             |
| India, and Japan   | Green tea   | 4                   | 211–1045                                   | Malik et al. (2008)             |
| India, China, and Indonesia  | White tea   | 6                   | 293–479                                    | Malik et al. (2008)             |
| Ceylon   | Black tea   | 4                   | 333.9–655.4                                | Salahinejad and Aflaki (2010)   |
| Ceylon, China, Georgia, India, and Kenya   | Black tea   | 36                  | 190–1570                                   | McKenzie et al. (2010)          |
| China and Japan  | White tea   | 28                  | 337–1463                                   | McKenzie et al. (2010)          |
| China and Taiwan   | Oolong tea  | 20                  | 449–2368                                   | McKenzie et al. (2010)          |
| China and Japan  | Green tea   | 24                  | 385–2081                                   | McKenzie et al. (2010)          |
| India and Iran   | Black tea   | 7                   | 436.2–674.3                                | Salahinejad and Aflaki (2010)   |
| <i>Nickel (Ni)</i>   |   |                     |  |                                 |
| India  | Black tea   | 5                   | 8–13                                       | Natesan and Ranganathan (1990)  |
| China and India  | Unknown   | 12                  | 4–10                                       | Pedersen et al. (1994)          |
| China  | Green tea ( <i>n</i> = 20), oolong( <i>n</i> = 5), black tea ( <i>n</i> = 8), white tea ( <i>n</i> = 2) | 39                  | 2–23                                       | Xie et al. (1998)               |
| China  | Commercial tea  | 9                   | BDL <sup>#</sup> –136.9                    | Ferrara et al. (2001)           |
| China  | Chinese tea   | 7                   | 77.8–2025                                  | Ferrara et al. (2001)           |
| Bangladesh, Burundi, China, Ethiopia, India, Japan, Kenya, Malawi, Malaysia, Papua New Guinea, Sri Lanka, and Tanzania | Black, green and oolong tea   | 54                  | 4.24–4.76                                  | Moreda-Piñeiro et al. (2003)    |
| Turkey   | Black tea   | 6                   | 5.15–8.32                                  | Tokalioglu and Kartal (2004)    |
| Thailand   | Black Tea   |                     | 2.281–9.194                                | Nookabkaew et al. (2006)        |
| Turkey   | Black tea and green tea   | 10                  | 3.1–5.7                                    | Soylak et al. (2007)            |
| India  | Black Tea   | 100                 | 0.4–9.2                                    | Seenivasan et al. (2008b)       |
| India  | Black tea   | 4                   | 4.02–8.03                                  | Malik et al. (2008)             |
| India and Japan  | Green tea   | 4                   | 2.57–9.35                                  | Malik et al. (2008)             |
| China and Indonesia  | Oolong tea  | 4                   | 1.88–3.33                                  | Malik et al. (2008)             |
| India, China, and Indonesia  | White tea   | 6                   | 4.18–7.71                                  | Malik et al. (2008)             |
| Ceylon, India, and Iran  | Black tea   | 11                  | 4.13–12.40                                 | Salahinejad and Aflaki (2010)   |
| Ceylon, China, Georgia, India, and Kenya   | Black tea   | 36                  | 0–7  | McKenzie et al. (2010)          |
| China and Japan  | Green   | 24                  | 0–9  | McKenzie et al. (2010)          |
| China and Taiwan   | Oolong  | 20                  | 1–5  | McKenzie et al. (2010)          |
| China and Japan  | White   | 28                  | 0–18                                       | McKenzie et al. (2010)          |

\*nd: not detected (below 0.007 mg kg<sup>-1</sup>).

# BDL: below detectable limit.

**Table 3**  
Trace elements in tea infusion.

| Country of origin                                       | Sample analyzed                 | Number of samples | Extraction procedure* | Concentration range | References  |
|---|---------------------------------|-------------------|-----------------------|---------------------|---|
| <i>Aluminium (Al) (in mg L<sup>-1</sup>)</i>            |                                 |                   |                       |                     |   |
| China and India   | Black tea                       | 4                 | 1                     | 2–4                 | Varo and Koivistoinen (1980)                      |
| China   | Black tea                       | –                 |                       | 2.0–6.0             | Chen (1984)                                       |
| Different countries                                     | Not known                       | 12                | 2                     | 2.7–4.9             | Fairweather-Tait, Moore and Fatemi (1987)         |
| Unknown   | Black tea                       | 5                 | 3                     | 2–6.2               | Flaten and Odegard (1988)                         |
| China, India, and Sri Lanka                             | Black tea                       | 13                | 4                     | 2.2–4.5             | Baxter, Burrell and Massey (1990)                 |
| India   | Black tea                       | 5                 | 5                     | 3.72                | Natesan and Ranganathan (1990)                    |
| China   | Green, black, and oolong tea    | 18                | 6                     | 2.3–2.5             | Wong et al. (1998)                                |
| Not known   | Black tea                       | 4                 | 7                     | 11–13               | Erdemoglu et al. (2000)                           |
| Not known   | Green tea                       | 17                | 8                     | 2–6                 | Fernández et al. (2002)                           |
| Ceylon and India  | Black tea                       | 23                | 8                     | 1–9                 | Fernández et al. (2002)                           |
| China   | Green tea and black tea         | 2                 | 9                     | 2.45–6.76           | Fung et al. (2003)                                |
| China   | Black tea                       | Not known         |                       | 1–6                 | Luo et al. (2004)                                 |
| India   | Black tea                       | 5                 | 10                    | 1.2–2.7             | Kralj, Krizaj, Bukovec, Slejko and Milacic (2005) |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Made tea                        | 17                | 11                    | 1.88–16.52          | Nookabkaew et al. (2006)                          |
| China, India, Sri Lanka, and other unknown countries    | Black and green tea             | 8                 | 12                    | 3.0–9.8             | Mehra and Baker (2007)                            |
| China, India, and Indonesia,                            | Black, green, oolong, and white | 18                | 13                    | 0.055–1.734         | Malik et al. (2008)                               |
| India   | Black tea                       | 4                 | 14                    | 0.06–3.2            | Mossion et al. (2008)                             |
| Turky   | Black tea                       | 100               | 15                    | 1.66–5.35           | Sofuoglu and Kavcar (2008)                        |
| China   | Oolong tea                      | 9                 | 16                    | 1.5–5.4             | Fung et al. (2009)                                |
| China   | Green tea                       | 7                 | 16                    | 0.7–5.5             | Fung et al. (2009)                                |
| Iran, India, and Ceylon                                 | Black tea                       | 11                | 17                    | 5.61–16.82          | Salahinejad and Aflaki (2010)                     |
| <i>Arsenic (As) (in µg L<sup>-1</sup>)</i>              |                                 |                   |                       |                     |   |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                       | 17                | 11                    | 0.02–1.53           | Nookabkaew et al. (2006)                          |
| Turky   | Black tea                       | 100               | 15                    | 0.002–0.73          | Sofuoglu and Kavcar (2008)                        |
| <i>Cadmium (Cd) (in µg L<sup>-1</sup>)</i>              |                                 |                   |                       |                     |   |
| India   | Black tea                       | 5                 | 5                     | 1.2–2.0             | Natesan and Ranganathan (1990)                    |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                       | 17                | 11                    | 0.04–0.24           | Nookabkaew et al. (2006)                          |
| China and India   | Unknown                         | 12                | 18                    | <0.08–0.32          | Pedersen et al. (1994)                            |
| Turky   | Black tea                       | 100               | 15                    | 0.02–0.79           | Sofuoglu and Kavcar (2008)                        |
| <i>Chromium (Cr) (in µg L<sup>-1</sup>)</i>             |                                 |                   |                       |                     |   |
| India   | Black tea                       | 5                 | 5                     | 1.7                 | Natesan and Ranganathan (1990)                    |
| China and India   | Unknown                         | 12                | 18                    | 0.7–4.5             | Pedersen et al. (1994)                            |
| Turkey and UK   | Black and green tea             | 10                | 19                    | Nil–0.8             | Tascloglu and Kok (1998)                          |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                       | 17                | 11                    | BDL–6.91            | Nookabkaew et al. (2006)                          |
| India   | Black tea                       | 12                | 20                    | 40–42               | Seenivasan et al. (2008a)                         |
| Turkey  | Black tea                       | 100               | 15                    | 1.58–43.2           | Sofuoglu and Kavcar (2008)                        |
| <i>Copper (Cu) (in mg L<sup>-1</sup>)</i>               |                                 |                   |                       |                     |   |
| India   | Black tea                       | 5                 | 5                     | 0.69                | Natesan and Ranganathan (1990)                    |
| China   | green, black, and oolong tea    | 18                | 6                     | 0.029–0.061         | Wong et al. (1998)                                |
| China   | Green, black and oolong tea     | 39                | 21                    | 0.048–0.640         | Xie et al. (1998)                                 |
| Turkey and UK   | Black and green tea             | 10                | 19                    | 0.019–0.084         | Tascloglu and Kok (1998)                          |
| Ceylon and India  | Black tea                       | 23                | 8                     | 0.03–0.16           | Fernández et al. (2002)                           |
| Not known   | Green tea                       | 17                | 8                     | 0.04–0.12           | Fernández et al. (2002)                           |
| China   | Green tea                       | 3                 | 22                    | 0.04–0.07           | Gallaher et al. (2006)                            |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                       | 17                | 11                    | 0.04–0.24           | Nookabkaew et al. (2006)                          |
| China, India, Kenya, Sri Lanka, and Turkey              | Black tea                       | 13                | 23                    | 0.072–0.441         | Street et al. (2006)                              |
| China, India, Kenya, Sri Lanka, and Turkey              | Green tea                       | 13                | 23                    | 0.033–0.191         | Street et al. (2006)                              |
| China, India, Sri Lanka, and other unknown countries    | Black and green                 | 8                 | 12                    | 0.08–0.27           | Mehra and Baker (2007)                            |
| China, India, and Indonesia                             | White tea                       | 6                 | 13                    | 0.048–0.182         | Malik et al. (2008)                               |
| Turky   | Black tea                       | 100               | 15                    | 3.57–65.4           | Sofuoglu and Kavcar (2008)                        |
| Ceylon, India, and Iran                                 | Black tea                       | 11                | 17                    | 2.38–9.00           | Salahinejad and Aflaki (2010)                     |
| <i>Fluoride (F) (in mg L<sup>-1</sup>)</i>              |                                 |                   |                       |                     |   |
| Unknown origin marketed in UK                           | Black tea                       | 30                | 24                    | 0.25–1.50           | Speirs (1983)                                     |
| India   | Black tea                       | 4                 | 25                    | 0.4–2.8             | Gulati et al. (1993)                              |
| China   | Brick tea                       | 590               | 26                    | 2.58–3.69           | Cao et al. (1997a)                                |
| China   | Green tea                       | 3                 | 27                    | 1.65–1.83           | Fung et al. (1999)                                |

Table 3 (continued)

| Country of origin                                       | Sample analyzed             | Number of samples | Extraction procedure* | Concentration range | References                     |
|---|-----------------------------|-------------------|-----------------------|---------------------|--------------------------------|
| <i>Fluoride (F) (in mg L<sup>-1</sup>)</i>              |                             |                   |                       |                     |                                |
| China   | Oolong tea                  | 3                 | 27                    | 1.22–1.51           | Fung et al. (1999)             |
| China   | Black tea                   | 3                 | 27                    | 1.90–2.41           | Fung et al. (1999)             |
| Turkey  | Black tea                   | 4                 | 28                    | 0.10–0.85           | Tokalioglu et al. (2001)       |
| China   | Green tea and black tea     | 2                 | 9                     | 2.45–6.76           | Fung et al. (2003)             |
| China, Japan, Sri Lanka, and India                      | Black tea                   | 12                | 29                    | 1.08–1.32           | Cao, Luo, Liu and Li (2004)    |
| China, and Taiwan                                       | Oolong tea                  | 2                 | 30                    | 0.87–2.85           | Malinowska et al. (2008)       |
| China, India, Kenya Nepal, and Sri Lanka                | Black tea                   | 20                | 30                    | 0.32–6.87           | Malinowska et al. (2008)       |
| China, Japan, Java, Sri Lanka, and Vietnam              | Green tea                   | 17                | 30                    | 0.59–2.52           | Malinowska et al. (2008)       |
| Turkey  | Black tea                   | 100               | 15                    | 0.34–1.48           | Sofuoglu and Kavcar (2008)     |
| China   | Brick tea                   | 39                | 31                    | 0.77–6.06           | Li et al. (2009)               |
| India, Kenya, Sri Lanka, and Turkey                     | Black tea                   | 26                | 32                    | 0.57–3.72           | Emekli-Alturfan et al. (2009)  |
| Inner Mongolia and China                                | Brick tea                   | 39                | –                     | 0.77–6.06           | Li et al. (2009)               |
| <i>Lead (Pb) (in mg L<sup>-1</sup>)</i>                 |                             |                   |                       |                     |                                |
| India   | Black tea                   | 5                 | 5                     | 0.038               | Natesan and Ranganathan (1990) |
| China   | Green, black and oolong tea | 39                | 21                    | NIL–0.23            | Xie et al. (1998)              |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                   | 17                | 11                    | 0.004–0.032         | Nookabkaew et al. (2006)       |
| Iran  | Black tea                   | 5                 | 33                    | 1.25–3.13           | Karimi et al. (2008)           |
| Iran  | Black tea                   | 5                 | 34                    | 5.82–6.00           | Shokrzadeh et al. (2008)       |
| Unknown   | Black tea                   | 5                 | 34                    | 4.74–5.20           | Shokrzadeh et al. (2008)       |
| <i>Manganese (Mn) (in mg L<sup>-1</sup>)</i>            |                             |                   |                       |                     |                                |
| India   | Black tea                   | 5                 | 5                     | 1.24                | Natesan and Ranganathan (1990) |
| Turkey  | Black tea                   | 2                 | 35                    | 3.66–4.21           | Ozdemir and Gucer (1998)       |
| Ceylon and India  | Black tea                   | 23                | 8                     | 1–6                 | Fernández et al. (2002)        |
| Not known   | Green tea                   | 17                | 8                     | 1.3–3.5             | Fernández et al. (2002)        |
| China   | Green tea                   | 3                 | 36                    | 1.86–2.48           | Gallaher et al. (2006)         |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                   | 17                | 37                    | 1.38–7.42           | Nookabkaew et al. (2006)       |
| China, India, Kenya, Sri Lanka, and Turkey              | Black tea                   | 13                | 38                    | 0.56–7.59           | Street et al. (2006)           |
| China, India, Kenya, Sri Lanka, and Turkey              | Green tea                   | 13                | 38                    | 0.78–3.94           | Street et al. (2006)           |
| China, India, Sri Lanka, and other unknown countries    | Black and green             | 8                 | 12                    | 1.91–11.9           | Mehra and Baker (2007)         |
| China and Indonesia                                     | Oolong tea                  | 4                 | 13                    | 2.20–3.73           | Malik et al. (2008)            |
| China, India, and Indonesia                             | White tea                   | 6                 | 13                    | 1.50–3.38           | Malik et al. (2008)            |
| India   | Black tea                   | 4                 | 13                    | 2.37–3.10           | Malik et al. (2008)            |
| India and Japan   | Green tea                   | 4                 | 13                    | 2.05–10.09          | Malik et al. (2008)            |
| Turkey  | Black tea                   | 100               | 39                    | 0.188–2.105         | Sofuoglu and Kavcar (2008)     |
| Ceylon, India, and Iran                                 | Black tea                   | 11                | 40                    | 125.8–164.0         | Salahinejad and Aflaki (2010)  |
| Ceylon  | Black tea                   | 4                 | 40                    | 88.7–249.4          | Salahinejad and Aflaki (2010)  |
| <i>Nickel (Ni), (in mg L<sup>-1</sup>)</i>              |                             |                   |                       |                     |                                |
| India   | Black tea                   | 5                 | 41                    | 0.118               | Natesan and Ranganathan (1990) |
| China and India   | Unknown                     | 12                | 18                    | 0.013–0.054         | Pedersen et al. (1994)         |
| China   | Green, black and oolong tea | 39                | 42                    | 0.034–0.106         | Xie et al. (1998)              |
| Turkey and UK   | Black and green tea         | 10                | 43                    | 0.007–0.067         | Tascloglu and Kok (1998)       |
| China, India, Indonesia, Japan, Sri Lanka, and Thailand | Green tea                   | 17                | 37                    | 0.04–0.16           | Nookabkaew et al. (2006)       |
| Turkey  | Black tea                   | 100               | 39                    | 0.001–0.049         | Sofuoglu and Kavcar (2008)     |
| India   | Black tea                   | 4                 | 13                    | 0.071–0.152         | Malik et al. (2008)            |
| India and Japan   | Green tea                   | 4                 | 13                    | 0.059–0.269         | Malik et al. (2008)            |
| China and Indonesia                                     | Oolong tea                  | 4                 | 13                    | 0.026–0.089         | Malik et al. (2008)            |
| China, India, and Indonesia                             | White tea                   | 6                 | 13                    | 0.050–0.135         | Malik et al. (2008)            |
| Ceylon, India, and Iran                                 | Black tea                   | 11                | 44                    | 1.86–7.65           | Salahinejad and Aflaki (2010)  |

\* 1: 2 g tea + 100 mL deionized water, simmered for 5 min; 2: 1 g tea + 100 mL water; 3: 1 g tea + 100 mL distilled water, infused for 5 min; 4: 3 g tea + 150 mL boiling deionized distilled water, infused for 4 min; 5: 2 g tea + 100 mL boiling water for 1 min; 6: 2 g tea + 200 mL boiling distilled water, 5 min; 7: 1 g tea + 20 mL of hot deionized water, heated up to boiling point and waited 10 min for cooling; 8: 1.50 g tea + 100 mL of boiling water, kept for adequate time; 9: 1 g tea + 100 mL double-distilled water, infused at 100 °C for 5 min; 10: 2.25 g tea + boiled with 150 mL water for 5 min; 11: 2.0 g tea + 100 mL of boiling deionized water, sample was left at room temperature for 5 min; 12: 5 g tea + 200 mL boiling distilled water, infused for 2 min; 13: 1 g tea + 50 mL boiling distilled water, covered by watch glasses, kept for 15 min; 14: 3.6–12 g tea + 600 mL water, boil at 95 °C up to 3 min; 15: 2 g tea + 200 mL ultra pure water, boiled for 15 min; 16: 2 g of tea + 200 mL double-distilled water, infused at 90 °C for 30 min; 17: 1 g tea + 45 mL hot distilled water, kept for 5 min; 18: 10 g tea + 1 L boiling Millipore water; 19: 10 g tea + 120 mL tap water, kept at 60 °C for 30 min (volume after filtration was 80 mL); 20: 2 g tea + 100 mL boiled water, kept for 6 min; 21: 1 g tea + 50 mL boiling distilled water, infused for 5 min; 22: 2.5 g tea + 236 mL boiling water, infused for 3 to 6 min; 23: 1 g tea + 50 mL of boiling distilled water, kept for 5 min; 24: 2 g tea + 200 mL glass distilled water, boil for 5 min; 25: 2.0 g of tea + 100 mL double-distilled water, 10 min boiling beakers; 26: Pieces broken off the block of brick tea and put into a boiler, after boiling, the tea water was mixed with milk; 27: 1 g tea + 100 mL double-distilled water at 100 °C for 25 min; 28: 10 g tea + 400 mL of boiling deionized water, tea liquor was taken after 5, 10, 20, 30 and 40 min; 29: 2 g tea + 200 mL deionized water, infused for 10 min at 100 °C; 30: 2 g tea + 100 mL of drinking water, boil, brewing time 5, 10 and 30 min; 31: 2 g tea + 100 mL water, infused at 95 °C for 10 min; 32: One tea bag kept in 100 mL boiled water; 33: 2 g tea + 100 mL boiling distilled water, incubated in 80 °C, left to cool at room temperature for 10 min; 34: 5 g tea + 250 mL boiling distilled water, infused for 15–20 min; 35: 5 g tea + 500 mL deionized water, 95 °C for 15 min; 36: 2.5 g tea + 236 mL boiling water, infused for 3 to 6 min; 37: 2.0 g tea + 100 mL of boiling deionized water, sample was left at room temperature for 5 min; 38: 1 g tea + 50 mL of boiling distilled water, kept for 5 min; 39: 2 g tea + 200 mL ultra pure water, boiled for 15 min; 40: 1 g tea + 45 mL hot distilled water, kept for 5 min; 41: 2 g tea + 100 mL boiling water for 1 min; 42: 1 g tea + 50 mL boiling distilled water, infused for 5 min; 43: 10 g tea + 120 mL tap water, kept at 60 °C for 30 min (volume after filtration was 80 mL); 44: 1 g tea + 45 mL hot distilled water, kept for 5 min.

and 382 mg kg<sup>-1</sup> (Malik, Szakova, Drabek, Balik, & Kokoska, 2008). Sofuoglu and Kavcar (2008) analyzed 100 made tea samples marketed in Turkey and reported that about 48 tea samples had Al concentrations higher than the Turkish nonalcoholic beverage limit of 2 mg L<sup>-1</sup>. Wróbel, Wróbel, and Urbina (2000) reported that Al content in green tea (919 ± 29 mg kg<sup>-1</sup>) was higher than in black tea (759 ± 31 mg kg<sup>-1</sup>). Also total acid digestable Al in nine oolong tea and seven green tea purchased from Hong Kong were 578–1859 mg kg<sup>-1</sup> and 224–2633 mg kg<sup>-1</sup> respectively (Fung et al., 2009).

### 2.1.3. Aluminium in tea infusion

Generally, drinking water provides 0.1 mg of Al (1.5% of total daily dietary Al intake), but in countries where Al from other sources is relatively small and tea consumption relatively large, tea may contribute up to 50% of total daily Al intake (UKMAFF, 1993). In a defined diet study conducted in Australia to estimate oral Al absorption, instant tea provided 1.8 mg Al L<sup>-1</sup>, >50% of the 3.2 mg Al consumed daily (Stauber, Florence, Davies, Adams, & Buchanan, 1999). In general, tea infusions typically contain 2 to 4 mg Al L<sup>-1</sup> (Sepe, Costantini, Ciaralli, Ciprotti, & Giordano, 2001). However, Flaten and Odegard (1988) concluded from their review of the literature that concentrations of Al in brewed teas were commonly in the range of 2 to 6 mg L<sup>-1</sup>, whereas the levels of 40 to 100 mg Al L<sup>-1</sup> was reported by Coriat and Gillard (1986), which were typical and exceptionally in high amounts. Findings for 10-min infusions of 2 g tea in 200 mL with a hard tap water were in the range of 1.8 to 3.9 mg L<sup>-1</sup> for samples of Assam, Darjeeling, Ceylon and a range of six common supermarket blends (French, Gardner, & Gunn, 1989).

In relation to Al toxicity, numbers of studies have reported the total concentration of Al in tea infusions (Kochian & Jones, 1997; Yokel & Florence, 2008). Wong et al. (1998) analyzed several tea infusions produced from black tea collected from major tea-growing countries and reported the presence of 0.7 to 3.5 mg Al L<sup>-1</sup> in tea infusion. In another study, Chen (1984) reported that tea infusion contained 2.0 to 6.0 mg Al L<sup>-1</sup>, whereas, Karimi et al. (2008) reported only 0.035 mg Al L<sup>-1</sup> in tea infusion from black tea marketed in Mashhad market, Iran. The total concentrations of Al in four tea infusions conducted by Flaten and Lund (1997) were 2.5 ± 0.2 (Georgia), 6.3 ± 0.3 (Argentina), 1.1 ± 0.2 (China) and 1.0 ± 0.1 (Kenya) mg L<sup>-1</sup> Al. These results indicated that Al was bound to the same relatively narrow size-range of large organic molecules in the tea infusions, irrespective of the origin of the tea.

Aluminium species in different tea infusions collected from Sri Lanka (black tea) were also investigated by Alberti, Biesuz, Profumo, and Pesavento (2003). It was found that Al in the tea infusions was present at a high total concentration of approximately 0.1 mM which was mainly linked to the less strong complexes. This could be one of the reasons for the high toxicity of aluminium in tea collected from Sri Lanka.

Tea contains 4 to 9% of inorganic matter, about one third of this is extracted during the brewing process (Erdemoğlu et al., 2000). The resulting concentration of Al in the tea infusion is in the range of 1 to 6 mg L<sup>-1</sup> (Baxter, Burrell, Crews, & Massey, 1989) and a cup of tea may thus contain approximately 0.2 to 1.0 mg of Al, where the estimated intake of Al is 3 to 10 mg per day (Sherlock, 1989). However, the concentration of Al in a cup of tea depends on the amount present in the original leaves as well as the time allowed and water used for the infusion process (Manickum & Verbeek, 1994; Mossion, Potin-Gautier, Delerue, Hecho, & Behra, 2008).

Fung et al. (2009) analyzed Al in tea infusion (1%, w/v tea infusion at 90 °C for 30 min) from 47 different tea bags and 28 samples of tea leaves marketed in Hong Kong. All tea samples released 0.703 to 5.931 mg Al L<sup>-1</sup> during infusion. These results were compared with other reports viz. 2 to 8 mg L<sup>-1</sup> (Flaten & Odegard, 1988), 2.5 mg L<sup>-1</sup> (Wong et al., 2003), 2.45 to 6.76 mg L<sup>-1</sup> (Fung, Zhang, Wong, & Wong, 2003), 1 to 6 mg L<sup>-1</sup> (Luo, Liu, Zhang, & Zhang, 2004) and an average of 2.75 mg L<sup>-1</sup> (Street, Drábek, Száková, & Mládkova, 2007). From the analytical data, Fung et al. (2009)

concluded that in comparison to the Joint FAO/WHO Provisional Tolerable Weekly Intake (JECFA, 2005) guidelines of 7 mg Al kg<sup>-1</sup> body weight, tea made with these tea leaves would not impose an adverse impact on human health.

Malik et al. (2008) analyzed 18 types of pure mixtures of tea leaves: four black teas (one decaffeinated and one with the addition of tea flowers), four green teas (one decaffeinated), four oolong teas (semi-fermented) and six white teas (one semi-fermented) for Al. It was observed that Al concentrations in tea infusions were 0.055 to 1.73 mg L<sup>-1</sup>. Malik et al. (2008) detected a broad spectrum for Al. In another study, a total of 29 tea samples from different origins, 13 green tea samples, 13 black tea samples, two semi-fermented and one white tea, imported from the Czech Republic, were collected and analyzed for total content of Al in tea infusions (Street et al., 2007). It was revealed that the average Al content in black tea was 1070 mg kg<sup>-1</sup> whereas, in green tea it was 1340 mg kg<sup>-1</sup>; this almost matched with the results reported by Matsuura et al. (2001). Street et al. (2007) reported that neither increased extraction time of tea infusion showed any effect on Al speciation nor did the addition of sugar (2.3 mg L<sup>-1</sup> and 3.2 mg L<sup>-1</sup> for black and green tea infusions, respectively). After the addition of lemon juice, the speciation of Al changed in one sample of black tea and five samples of green tea in their experiment. These findings suggested that lemon juice as an additive can significantly influence Al speciation in tea infusions. At present, according to the WHO guidelines, the maximum permissible level of Al in drinking water is 0.2 mg total Al per litre (WHO, 2003a). Therefore, the Al concentration in tea infusion is 10 to 100 times more than that of drinking water. However, this does not mean that tea infusion is 10 to 100 times more toxic, because toxicity is dependant mainly on Al speciation (Wickremasinghe, 1999). Aluminium is generally poorly absorbed in the gastrointestinal tract; roughly in the order of 0.1% of the dietary intake is absorbed, depending on the chemical species of Al (Priest et al., 1996). Thus, if the species of Al present in tea are more bioavailable than the species present in other dietary sources, tea could make a larger contribution towards human uptake of Al than is indicated by the total concentration present. However, the possible dietary risks of Al contents in tea may lead to the proposal that Al in tea soils should be at least partly immobilized. This may be achieved by increasing the organic matter content of the soils. The introduced organic matter should contain humic substances which are generally known for their high Al-binding capacity (Sposito, 1996).

In order to find out the amount of Al which may be released to Chinese tea liquor during tea infusion, a continuous infusion of 5 g tea (China tea) was infused in 500 mL (1% water extract, w/v) hot (90 °C) distilled water (pH 6.5) by Wong et al. (2003). It was reported that under continuous infusion, brick tea released higher concentrations of Al (2500 mg kg<sup>-1</sup>) after 30 min and on further infusions, it dropped noticeably after 1 h. In case of black tea infusion, only 750 mg kg<sup>-1</sup> Al was found. Therefore, consumers taking a large quantity of brick tea liquor may be exposed to a high level of Al. It has been observed that consumption of 0.3 L brick tea liquor per day would exceed the permissible limit of 15 mg L<sup>-1</sup> after 1 h infusion of brick tea as recommended by WHO (1997). Therefore, the potential of Al toxicity from drinking tea should not be overlooked, particularly among people who consume considerable tea that has a high Al content, as has been observed among some people in China who consume poor quality tea made from older leaves and branches (Wong et al., 1998).

Toxicity of trace element not only depends on their concentration, but also on their speciation. However, among the trace elements, the speciation study only focuses on very few selected trace elements. Aluminium bound in fluorides or organic complexes, phosphate or silicate Al polymers and Al(OH)<sub>3</sub> is said to be non-toxic (Boudot, Becquer, Merlot, & Rouiller, 1993). Toxicity of polymer [AlO<sub>4</sub>Al<sub>12</sub>(OH)<sub>24</sub>(H<sub>2</sub>O)<sub>12</sub>]<sup>7+</sup> is often considerably higher than that of Al<sup>3+</sup> (Parker & Bertsch, 1992). Street et al. (2007) reported that among 29 tea simple infusions (with no additives) showed similar Al speciation,

with no general differences between black and green tea. The  $Al^{3+}$  species, were not detected in any sample. This observation confirms the fact that Al present in tea infusions does not exist in free forms of hexahydrates or in the form of hydroxypolymers.

According to Mehra and Baker (2007), 1 L of tea can provide >100% of the daily dietary intake of Al however, the percentage available for absorption in the intestine is only between 4.96% and 9.13% for different tea samples. Therefore, from their findings they suggested that it is unlikely that moderate amounts of tea drinking can have any harmful effects on humans. However, it must be stated that tea drinking may contribute towards Al toxicity in individuals with impaired absorption or excretion of Al in their systems.

## 2.2. Arsenic

Arsenic is a mutagenic and carcinogenic element and highly toxic to plants and animals (Pais & Jones, 1997). Several reports about arsenic in food and beverages have been carried out to assess the daily dietary intake of arsenic for humans (Han, Shi, Ma, & Ruan, 2005; Kannamkumarath, Wuilloud, & Caruso, 2004; Laparra, Vélez, Barberá, Farré, & Montoro, 2005 and references there in).

### 2.2.1. Arsenic in tea leaves

Studies on distribution and uptake of As through tea plants have not been done exclusively so far. A recent experiment on As accumulation by Chinese tea grown in noncontaminated field and polluted soil simulated by As salts has been documented by Shi, Ruan, Ma, Han, and Wang (2008). In this field experiment, authors reported that As concentration in tea leaves were largely varied and were dependent on tea cultivars. For example, As accumulation by young shoots by five cultivars like Biyun, Jiuken1, Jiuken2, Longjing43 and Yinshuang were  $0.03 \pm 0.02$ ,  $0.04 \pm 0.02$ ,  $0.02 \pm 0.02$ ,  $0.07 \pm 0.05$ , and  $0.05 \pm 0.03$  mg kg<sup>-1</sup> respectively. In pot experiment, it was observed that As uptake by tea plants was increased remarkably by adding As salt in the form of Na<sub>2</sub>HAsO<sub>4</sub>. For example incorporation of 50 mg As per kg soil increased to 11.2 mg per kg young shoots (about 20 times over the control), while As concentration increased to 22.1 mg per kg young shoots when application rate of As was 200 mg per kg soil.

### 2.2.2. Arsenic in made tea

In addition to food, beverages like tea may act as an important potential dietary ingestion source of toxic elements in daily life. The regular consumption of tea may contribute to the daily dietary requirements of several elements. But at the same time, some toxic elements like arsenic may also be ingested into the body (Han et al., 2005). High arsenic level may occur in tea leaf, hence the case of higher consumption of tea may be necessary to figure out the arsenic species in tea (Han et al., 2005). Out of the 800 tea samples analyzed for As concentration, the ranges found were from below the detection limit to 4.43 mg kg<sup>-1</sup> dry weight of made tea samples, with a mean value of 0.26 mg kg<sup>-1</sup>. Yuan, Gao, He, and Jiang (2007) reported data of As, including As speciation in a total of 47 tea samples from 18 tea producing provinces in China. The total amount of arsenic in tea leaf samples was in the range from below the detection limit to 4.81 mg kg<sup>-1</sup>. Shi, Jin, and Zhu (2007) analyzed 328 tea samples for As during years 1997 and 1998 which came from the main tea producing regions of China. They found that the average content of As was 0.30 mg kg<sup>-1</sup> and its ranges were between the not detectable limit of the instrument and 2.0 mg kg<sup>-1</sup> in which 84.1% were less than 0.50 mg kg<sup>-1</sup> and 13.6% were between 0.50 and 1.00 mg kg<sup>-1</sup>. Out of the 2307 samples analyzed in the year 2004 for As, it was found that the average content of As was 0.65 mg kg<sup>-1</sup> and it ranged between the not detectable limit of the instrument and 2.00 mg kg<sup>-1</sup> in which 62.9% samples were less than 0.50 mg kg<sup>-1</sup> and 30.1% were between 0.50 and 1.00 mg kg<sup>-1</sup>.

### 2.2.3. Arsenic in tea infusion

Yuan et al. (2007) reported that leaching of arsenic from 47 tea infusions was strongly affected by extraction time and temperature. Because arsenic leaching ability by hot water was low and most of the arsenic was left in tea leaf residues after infusion, the concentration of arsenic in tea infusion was low even when some original tea leaf samples contained high level of arsenic. Yuan et al. (2007) also reported that the major species in tea infusion were in the form of inorganic arsenic (arsenite As<sup>III</sup> and arsenate As<sup>V</sup>). Compared with the amount of arsenic in infusion, more organic arsenic species were found in the original tea leaf samples. The contents of extractable inorganic arsenic in tea leaf samples were in the range from below the detection limit to 226 ng g<sup>-1</sup>. To evaluate the As intake from tea for safety assessment, not only the total amount of As in original tea leaf but also the leachability and species in infusion should also be considered because of its unique infusion and drinking ways. For this purpose, Yuan et al. (2007) also reported that the As species distributions in tea leaf were different from the corresponding tea infusions. Much more methylated arsenic was detected in tea leaf samples than in tea infusion. The extraction efficiency by microwave assisted extraction was also much higher than hot water. For example, there was no As<sup>III</sup>, DMA<sup>V</sup> and MMA<sup>V</sup> detected in a tea infusion among 47 tea samples but 0.106 mg kg<sup>-1</sup> As<sup>III</sup>, 0.092 mg kg<sup>-1</sup> DMA<sup>V</sup> and 0.146 mg kg<sup>-1</sup> As<sup>V</sup> were found in its corresponding tea leaf sample. Considering ingestion dose and assuming one person (60 kg body weight) consumes 10 g of Chinese tea per day, the maximum inorganic arsenic contribution from tea infusion is 2.26 µg, which is equal to 0.038 µg kg<sup>-1</sup> day<sup>-1</sup> excluding water contribution. This result was also consistent with the previous report which reported that heating of water increased arsenic concentration in the hot beverages which could contribute As in human body (Del Razo et al., 2002). Sofuoglu and Kavcar (2008) reported that the average As content in 100 black tea infusions in Turkey was 0.21 µg L<sup>-1</sup>.

Shen and Chen (2008) reported that mean As contents in oolong tea ( $n=18$ ) and black tea infusion ( $n=15$ ) were 0.005 µg L<sup>-1</sup> and 0.007 µg L<sup>-1</sup> respectively, whereas no As was found in 15 green tea samples from Taiwan when infusion was prepared from 2.0 g of tea sample with 100 mL of water. El-Hadri, Morales-Rubio, and de la Guardia (2007) reported that in tea marketed in Spain the total As content in all samples ranged from 0.13 ± 0.01 ng mL<sup>-1</sup> to 0.96 ± 0.01 ng mL<sup>-1</sup>. Arsenic content in all the samples were found to be below the Spanish regulation for arsenic content in soft drinks, where the maximum tolerable concentration level was stipulated at 250 µg L<sup>-1</sup> for tea drinks stored in both metallic and non-metallic containers. Karimi et al. (2008) reported that As contents in five made tea and tea infusion samples in Iran was found to be 0.09 mg kg<sup>-1</sup> and 0.001 µg/100 mL respectively. According to Tokalioglu and Kartal (2004), high As level may occur in tea leaf because of high As uptake from tea garden soils and in case of high consumption of tea, it is necessary to assess the contribution of tea to the daily dietary As intake.

The Provisional guideline value of As in drinking water prescribed by WHO (WHO, 2003b) is 0.01 mg L<sup>-1</sup> and this guideline value is mainly designated as provisional in view of the scientific uncertainties. The estimated maximum intakes of arsenic prescribed by the FAO/WHO for provisional tolerable weekly intake (PTWI) is 15 µg per kg body weight (i.e. 900 µg per person; FAO/WHO, 1989). For the safety of drinking tea with respect to As intake, Nookabkaew, Rangkadilok, and Satayavivad (2006) calculated average daily dietary intake (ADDIs) on 17 tea samples. The calculation was based on the experiment that one cup of infusion was prepared from 2 g or made tea and each person consumed three cups (total 300 mL tea infusion) of tea per day. On the basis of ADDIs, As intake was 0.000–0.0005 mg per day per person which was much more lower than ADDI value (0.1050–0.4060 mg per day per person). Therefore, consumption of tea may not lead to a threat in health because of As intake. On the other study, Yuan et al. (2007) reported that consuming 10 g of Chinese tea per day per person, the maximum

inorganic arsenic contribution from tea infusion is  $2.26 \mu\text{g}$ , which is equal to  $0.038 \mu\text{g kg}^{-1} \text{ day}^{-1}$  excluding water contribution. This value only accounts for 1.8% of provisional tolerable weekly intake (PTWI) ( $2.1 \mu\text{g}$  per kg body weight per day), recommended by the Food and Agriculture Organization/World Health Organization. On the contrary, results from sensitivity analysis of As consumption through black tea among 50 Turkish people, Sofuoglu and Kavcar (2008) concluded that carcinogenic risk was the most sensitive to the As concentration in tea.

### 2.3. Cadmium

Cadmium (Cd) has been designated as a human carcinogen (Waalkes, 2000) and is clearly a potent, multi-tissue animal carcinogen (Satarug, Baker, Reilly, Moore, & Williams, 2002). Phosphate fertilizer application is a significant contributor of trace elements, especially for Cd accumulation in cropped soils (Cupit, Larsson, de Meeus, Eduljee, & Hutton, 2002). Repeated application of phosphatic fertilizers may lead to a gradual buildup of Cd in agricultural soils over time (Kabata-Pendias & Pendias, 2001). According to Meeüs, Eduljee, and Hutton (2002), Cd from phosphate fertilizers constituted more than 50% of the total input in agricultural lands that were not heavily polluted or heavily industrialized. The phytotoxicity of Cd is also well known (Fodor, 2002), and is manifested as inhibition of plant growth (Titov, Talanova, & Boeva, 1995), nitrate assimilation (Hernández, Gárate, & Carpena-Ruiz, 1997) and photosynthesis (Barceló, Vázquez, & Poschenrieder, 1988; Larbi et al., 2002), as well as disturbances in plant ion (Wallace, Wallace, & Cha, 1992) and water balances (Barceló & Poschenrieder, 1990). The maximum permissible content of Cd in drinking water is  $0.003 \text{ mg L}^{-1}$  (WHO, 2003c).

#### 2.3.1. Cadmium in tea leaves

Cd concentration in tea leaves has not been frequently reported. Chen, Yu, Xu, Chen, and Shi (2009) reported that the concentration of Cd in different tea leaves ranged from  $0.03$  to  $0.08 \text{ mg kg}^{-1}$ . These were in good agreement with the levels obtained in previous study reported by Moreda-Piñeiro, Fisher, and Hill (2003). However, Yemane et al. (2008) analyzed five tea clones grown in Wushwush tea plantation farms, Ethiopia and Cd in the leaf tissues was present at the level which is too low to be detected by the analytical techniques.

#### 2.3.2. Cadmium in made tea

Various reports have discussed the potential health implications of some chemical factors such as Cd in tea (Tsushida & Takeo, 1977; Natesan & Ranganathan, 1990; Shi et al., 2008). Tea can be contaminated by Cd during growth period and manufacturing processes which might lead to an increase in the Cd body burden in humans. Cd constituents of tea leaves is normally different according to the type of tea (green or black) and geological source (Marcos et al., 1998). The main sources of Cd metal in tea plants are their growth media, nutrients, agro inputs and soil (Seenivasan, Manikandan, & Muraliedharan, 2008).

Tsushida and Takeo (1977) reported that the concentration of Cd in green tea in Japan was found to be  $0.11$  to  $1.93 \text{ mg kg}^{-1}$  with the mean of  $0.49 \text{ mg kg}^{-1}$ . AL-Oud (2003) has reported the level of Cd in tea samples marketed in Pakistan to be within the range from below the detectable limit to  $0.18 \text{ mg kg}^{-1}$ . Seenivasan et al. (2008a) analyzed one hundred black tea samples collected from the tea-growing regions of Valparai and Nilgiris in Tamil Nadu, Vandiperiyar, Wayanad and Munnar in Kerala and Karnataka states in south India for Cd. The results showed that the level of Cd in black tea was between  $0.05$  and  $0.38 \text{ mg kg}^{-1}$ . The lowest level of Cd was found in tea samples from Valparai and was highest from Vandiperiyar and Peermedu.

Analysing the tea samples collected from various countries of the world, Ferrara, Montesano, and Senatore (2001) found that Cd in tea was below the detectable limit. However, Narin, Colak, Turkoglu, Soyak, and Dogan (2004) reported that Cd level in fourteen Turkish black tea samples were  $<1$  to  $3.0 \text{ mg kg}^{-1}$  with the mean value of  $2.3 \pm 0.4 \text{ mg kg}^{-1}$ .

Mengel and Kirby (1987) noted that certain plants accumulated Cd without exhibiting any toxic symptoms. Further, they claimed that the availability of cadmium in soils was influenced positively by the addition of rock phosphate because rock phosphate is applied annually in many tea soils. Franklin, Duis, Brown, and Kemp (2005) reported  $4.9$  to  $5.5 \text{ mg g}^{-1}$  of Cd in phosphatic fertilizers and  $11.8$  to  $50.9 \text{ mg kg}^{-1}$  in zinc sources and these are major sources of Cd in tea.

Han et al. (2005) reported the Cd concentrations of a total of 798 tea samples collected from the main tea producing provinces in China. The Cd concentrations in the tea samples ranged from below the detection limit to  $1.07 \text{ mg kg}^{-1}$  dry weight of tea samples with a mean and median value of  $0.02 \text{ mg kg}^{-1}$  dry weight and under the detection limit, respectively. Among the different types of tea, green tea had the highest Cd concentration. In another study, Zhang and Fang (2007) reported that Cd in green tea leaves in China was at the lowest concentration among different trace metals, ranging from  $0.012$  to  $0.057 \text{ mg kg}^{-1}$ . From their study, it was concluded that the dissolved organic matter (DOM) in soil and acidification increased the bioavailability of Cd in soil towards tea plants. Shi et al. (2007) analyzed 328 tea samples for Cd, collected from the main tea producing regions of China during the years 1997 and 1998. It was found that the average content of Cd was  $0.06 \text{ mg kg}^{-1}$  and it ranged between ND (not detectable) and  $1.59 \text{ mg kg}^{-1}$  in which 94.2% were less than  $0.20 \text{ mg kg}^{-1}$  and 4.6% between  $0.20$  and  $0.50 \text{ mg kg}^{-1}$ . Further analysis of 2307 samples in year 2004 for Cd, revealed that the average content of Cd was  $0.10 \text{ mg kg}^{-1}$  and it ranged between ND and  $7.22 \text{ mg kg}^{-1}$  in which 58.3% was less than  $0.20 \text{ mg kg}^{-1}$  and 40.9% were between  $0.20$  and  $0.50 \text{ mg kg}^{-1}$ . Therefore, these data showed that the contents of Cd in tea were increasing constantly just like As. Cadmium levels in black tea samples on marketed black tea samples in Saudi Arabia were found to be in the range of  $0.32$  to  $2.17 \text{ mg kg}^{-1}$  (Ashraf & Mian, 2008) with mean cadmium value of  $1.1 \text{ mg kg}^{-1}$ .

#### 2.3.3. Cadmium in tea infusion

On the basis of the existing literature, it was observed that Cd in tea infusion was generally low. Odegard and Lund (1997) reported that Cd in Lipton Yellow Label tea commonly used in Norway contents was  $<0.08 \mu\text{g L}^{-1}$ . Shen and Chen (2008) reported that only 52.8% of the total Cd was released from eighteen samples of oolong tea ( $0.005 \mu\text{g}$  per  $100 \text{ mL}$ ), 40.3% from fifteen black tea samples ( $0.06 \mu\text{g}$  per  $100 \text{ mL}$ ), while no Cd was extracted from fifteen green tea samples in Taiwan. In order to assess Cd in Iranian consumed tea, Shokrzadeh, Saberyan, and Saravi (2008) analyzed 10 black tea samples of which five samples were from Iran and others from unknown foreign countries. The result showed that Cd level in Iranian and foreign tea infusions were from below the detectable limit to  $1.093 \text{ mg L}^{-1}$  and from below the detectable limit to  $0.673 \text{ mg L}^{-1}$ , respectively. However, no Cd was found in ten black tea samples collected from Iran, India and Sri Lanka (Salahinejad & Aflaki, 2010). The Joint FAO/WHO Provisional Tolerable Weekly Intake (PTWI) guideline for Cd is  $0.007 \text{ mg Cd}$  per kg body weight (i.e.  $0.420 \text{ mg Cd}$  per person per week) (WHO, 2003c). From the data it can be concluded that some tea infusion made with different tea leaves might have adverse impacts on human health however, a larger number of tea samples should be analyzed to make a concrete and robust model.

### 2.4. Chromium

Though chromium (Cr) is an essential trace element for both human beings and animals (Schwarz, 1972), introducing in human beings through food chain might cause adverse effect. Chromium is found in all phases of the environment, including air, water and soil. Naturally occurring in soil, Cr ranges from  $10$  to  $50 \text{ mg kg}^{-1}$  depending on the parent material (Adriano, 1986). Usually, Cr occurs in two forms, Cr (III) and Cr (VI), and plant uptake is in both forms. Cr (III) is sparingly soluble and less toxic, while Cr (VI) being more soluble in water and is highly toxic to biota (Adriano, 1986). Chromium interferes with several

metabolic processes, causing toxicity to the plants as exhibited by reduced root growth and phytomass, chlorosis, photosynthetic impairing, stunting and finally plant death (Gardea-Torresdey, Peralta-Videa, Montes, de la Rosa, & Corral-Diaz, 2004). Therefore, contamination of soils due to the use of Cr in various anthropomorphic activities has become a serious concern to plant and animal scientists over the past decade. Its complex electronic chemistry has been a major hurdle in unraveling its toxicity mechanism in plants. The impact of Cr contamination in the physiology of plants depends on the metal speciation, which is responsible for its mobilization, subsequent uptake and resultant toxicity in the plant system. Cr toxicity in plants is observed at multiple levels, from reduced yield through effects on leaf and root growth to inhibition on enzymatic activities and mutagenesis (Gardea-Torresdey et al., 2004). Provisional guideline for total Cr in drinking water recommended by WHO (2003d) is  $0.05 \text{ mg L}^{-1}$ .

#### 2.4.1. Chromium in tea leaves

Experimental data of chromium in tea leaves are very scanty. Natesan and Ranganathan (1990) concluded that to produce 1 ton of made tea, tea shoots assimilated about 1 g chromium from soils. Very recently Chen et al. (2009) reported that chromium in eight tea cultivar leaves was found between 0.39 and  $0.97 \text{ mg kg}^{-1}$ .

#### 2.4.2. Chromium in made tea

Wróbel et al. (2000) reported the average Cr contents in Mexican black and green tea samples as  $0.43 \pm 0.05$  and  $0.62 \pm 0.01 \text{ mg kg}^{-1}$  respectively. Seenivasan, Manikandan, Muraleedharan, and Selvasundaram (2008) in a survey of marketable black tea in South India found out that Cr content varied from 2.5 and  $11.4 \text{ mg kg}^{-1}$ . The lowest and highest contents of Cr were found in Karnataka and Munnar samples, respectively. However, in the majority (90%) of the samples tested, Cr was below  $10 \text{ mg kg}^{-1}$ . They also concluded that Cr content in black tea was positively correlated to the sharpening of Crush, Tear and Curl (CTC) rollers used for manufacturing. Ashraf and Mian (2008) reported the average Cr contents in some Saudi Arabia tea samples to be  $9.8 \pm 4.5 \text{ mg kg}^{-1}$ . Narin et al. (2004) found maximum Cr in Turkish tea samples at the levels of  $16.9 \pm 1.5 \text{ mg kg}^{-1}$ . Chromium concentration of total 801 tea samples tested in China ranged from below the detectable limit to  $16.10 \text{ mg kg}^{-1}$  (Han et al., 2005). Ferrara et al. (2001) reported that Cr in Chinese black tea samples were in the range of 17.9 to  $115.4 \text{ mg kg}^{-1}$  whereas, Cr content among the nine commercial tea collected from different countries were within the range between 19.8 and  $129.1 \text{ mg kg}^{-1}$ .

#### 2.4.3. Chromium in tea infusion

Natesan and Ranganathan (1990) had reported that 16.5% of Cr present in black tea was transferred into tea brew when the brewing time was 1 min and when the brewing time was increased to 5 min, the percentage transfer of Cr was 42.2. In general, Cr to the brew was reported positively correlated with the level of Cr in the black tea and the brewing time. Tascoglu and Kok (1998) reported that the transfer of Cr into tea brew from black tea was higher than from green tea. Seenivasan et al. (2008b) reported that black tea infusion contained very low amounts of Cr, between 0.04 and  $0.42 \text{ mg L}^{-1}$  when brewed for 6 min.

On the basis of the hazardous quotient (HQ) data and population health risk distribution of Cr among 50 participants (female 56% and male 44%; taking population age distribution into account), Sofuoglu and Kavcar (2008) reported that 95th percentile HQ values was 0.05. Therefore, Sofuoglu and Kavcar (2008) concluded that according to populations' noncarcinogenic risk distribution, the risk levels through consumption of black tea were not significant for Cr. However, while Cr (VI) is toxic and carcinogenic upon uptake, the data of Cr described in different literatures mainly focused on the total Cr rather than Cr in tea infusion and Cr speciation. Therefore, speciation of Cr in made tea and tea infusion needs to get special importance.

## 2.5. Copper

Copper (Cu) is a micronutrient for plants, but it is also highly phytotoxic at high concentrations (Brun, Maillat, Hinsinger, & Pépin, 2001). Overconsumption of Cu from food and beverages is detrimental to human health too (Kawada, Lee, Suzuki, & Rivai, 2002). Therefore, Cu content in water, food and beverages must be traced and controlled on a daily basis.

### 2.5.1. Copper in made tea

Although the judicious use of Cu fungicides may not cause any problem to human health and environmental contamination, its indiscriminate use may lead to the presence of undesirable levels of Cu content in black tea (Seenivasan et al., 2008a). Analysis of Cu content in tea samples from nine tea-growing regions of the world by Lepp and Dickinson (1985) showed that eight of the nine samples exceeded the normal range of 2–20  $\text{mg kg}^{-1}$  and samples produced from China had the highest content of  $78 \text{ mg kg}^{-1}$ . Experimental results published by Seenivasan et al. (2008a) on analysis of Cu content in 86 made tea samples collected from South India indicated the mean value of Cu to range between 15.9 and  $32.2 \text{ mg kg}^{-1}$ . Copper is one of the native metals found in tea, central to polyphenol oxidase enzyme. The lowest value of Cu was found in Nilgiris tea and the highest in Gudalur tea samples in south India. It was evident from this study that the Cu contents of all the made tea samples were less than  $35 \text{ mg kg}^{-1}$ , which is well below the permissible limit of  $150 \text{ mg kg}^{-1}$  under Prevention of Food Adulteration Act, 1954 (PFA), India.

Cu contents in Indian and US tea brands vary over a wide range, 1.60 to 35.0 and 4.4 to  $17.3 \text{ mg kg}^{-1}$ , with mean values of  $14.8 \pm 8.2$  and  $12.3 \pm 4.8 \text{ mg kg}^{-1}$ , respectively, which are comparable (Kumar, Nair, Reddy, & Garg, 2005). However, Cu content in Indian tea brands is present in a much wider range compared to that in the US brands. Wang, Ke, and Yang (1993) also reported that Cu content to be in the range of 9.6 to  $20.9 \text{ mg kg}^{-1}$  in three Chinese tea brands. Copper concentrations in 57 commercial tea samples purchased at the local market in Beijing, China, varied from 1.74 to  $21.15 \text{ mg kg}^{-1}$  of dry weight (average,  $12.63 \text{ mg kg}^{-1}$ ) for green tea;  $8.22$  to  $28.71 \text{ mg kg}^{-1}$  (average,  $16.35 \text{ mg kg}^{-1}$ ) for scented tea; 12.10 to  $48.19 \text{ mg kg}^{-1}$  (average,  $22.02 \text{ mg kg}^{-1}$ ) for oolong tea; 8.72 to  $28.78 \text{ mg kg}^{-1}$  (average,  $23.88 \text{ mg kg}^{-1}$ ) for black tea (Qin and Chen, 2007). The differences in methods used for processing and storage as well as the different types of leaves used could be the contributory factors to these variations.

Han et al. (2005) reported the Cu concentration of a total of 811 tea samples collected from the main tea producing provinces of China. The Cu concentrations in the tea samples ranged from 2.04 to  $447.50 \text{ mg kg}^{-1}$  dry weight of tea samples with a mean and median value of  $18.33 \text{ mg kg}^{-1}$  and  $15.24 \text{ mg kg}^{-1}$  dry weight, respectively. Among the different types of tea, black tea had the highest Cu concentration. The probable reasons of high concentration of Cu in black tea was the use of Cu-bearing Bordeaux mixture used to deal with disease problems and partly due to the copper boards used at the twisting stage (Jin, Du, Zhang, & Lin, 2008). Although Cu is an essential trace element for humans and is an indispensable component for many enzyme systems, it can also act as a toxic metal to which a number of pathogenic characteristics have been attributed, such as non-Indian childhood cirrhosis (Zietz et al., 2003) and Wilson's disease (Verissimo, Oliveira, & Gomes, 2005).

The Cu concentration in the 20 tea leaves collected from Yuyao County, China, ranged from 8.05 to  $33.50 \text{ mg kg}^{-1}$ , with an average of  $13.26 \text{ mg kg}^{-1}$  (Jin et al., 2008). None of the 20 tea leaf samples exceeded  $60 \text{ mg kg}^{-1}$ , the allowable limit given by the Chinese Ministry of Health. Therefore, even for people who consume a large amount of tea (e.g., 10 g of tea leaves per day), the daily intake of Cu from tea drinking is less than 0.4 mg, while the recommended daily consumption of Cu for adults, issued by the US Air Force, is 1.5 to

3.0 mg (US AF, 1990). Therefore, the Cu concentrations in the tea leaves from Yuyao County are acceptable from the perspective of the consumer health.

Ramakrishna, Palmakumbura, and Chatt (1986) reported that 10.0 to 25.0 mg kg<sup>-1</sup> Cu was found in tea samples from Sri Lanka. The significant differences in Cu were observed among tea varieties grown at different elevations in Sri Lanka. These levels were also compared with the results obtained using samples from other tea-growing countries like India (24.07 ± 2.25 mg kg<sup>-1</sup> Cu; Seenivasan et al., 2008a) and Iran (19.6 to 36.7 mg kg<sup>-1</sup>; Ansari, Norbakhsh, & Daneshmandirani, 2007). However, the value was much lower than other countries like Japan (100 mg kg<sup>-1</sup>; Fuchinokami & Fuchinokami, 1999), Australia, the United Kingdom and the United States (150 mg kg<sup>-1</sup>; Kumar et al., 2005) and Turkey (120 mg kg<sup>-1</sup>; Narin et al., 2004).

Moreda-Piñeiro et al. (2003) observed the concentration of Cu in the range of 12.34 to 19.14 µg g<sup>-1</sup> from 85 tea samples (36 samples from Asian countries, 18 samples from African countries, 24 commercial blends and 7 samples of unknown origin). The data showed that the concentrations of Cu in all the samples were within the permissible limit specified by the respective countries. The mean Cu content of the samples in Saudi Arabia was 18.1 mg kg<sup>-1</sup> (Ashraf & Mian, 2008). Among the 17 samples, the lowest level of Cu was found in Panda tea as 9.4 mg kg<sup>-1</sup>, whereas, highest in Ahmad tea and is 31.0 mg kg<sup>-1</sup>. Similar results for Cu in made tea were reported by other researchers (Ferrara et al., 2001; Narin et al., 2004; Xie et al., 1998).

Determination of Cu was also performed in several commercially available tea samples, such as Brooke Bond, Georgian, Dilmah, Ceylon and Chinese green tea leaves by Tautkus, Kazlauskas, and Kareiva (2004) using flame atomic absorption spectrometry (FAAS). The concentration of Cu in these tea samples ranged from 19.8 mg kg<sup>-1</sup> (Chinese green) to 33.9 mg kg<sup>-1</sup> (Brooke Bond). Tshushida and Takeo (1977) reported that the concentration of Cu in green tea in Japan was found to be 4.7 to 36.5 mg kg<sup>-1</sup> with a mean value of 11.4 mg kg<sup>-1</sup>.

### 2.5.2. Copper in tea infusion

Generally tea infusion contents 0.05 ± 0.003 mg L<sup>-1</sup> Cu (Gallaher, Gallaher, Marshall, & Marshall, 2006). Mehra and Baker (2007) analyzed copper extracted from eight different teas. Extraction was performed using 5 g tea with 200 mL distilled water with different infusion periods (2 min, 5 min and 10 min). The trends of Cu transfer at the first infusion (at 2 min) was Assam>Gunpowder Green>Tetley>Ceylon>Darjeeling>Ceylon decaffeinated>Lapsang Souchong>Kashmir Green, for the second infusion (5 min) it was Assam>Gunpowder Green>Tetley>Darjeeling>Ceylon>Lapsang Souchong>Kashmir Green>Ceylon decaffeinated and for the third infusion (10 min) it was Assam>Gunpowder Green>Tetley>Darjeeling>Ceylon>Lapsang Souchong>Kashmir Green>Ceylon decaffeinated. In addition, they concluded that the extraction rate of Cu in the first infusion was the highest followed by the second and the third infusions and was in decreasing order irrespective of the tea samples.

Average daily dietary intake of Cu by a person through food and beverages is 2.5 mg day<sup>-1</sup> with a range of 2–3 mg day<sup>-1</sup> (Powell, Burden, & Thompson, 1998). According to Powell et al. (1998), Cu concentrations in 2 min infusion from 12.4 g black tea in one litre water provided 0.05 ± 0.003 mg L<sup>-1</sup> Cu which is 2.0% of average daily dietary intake from 1.0 L tea. Mehra and Baker (2007) showed that 'available' Cu from drinking 1 L of tea per day provides 2.88% (loose tea) and 2.39% (tea bag tea) of the average daily dietary intake. Compared to Al, contribution of tea towards the daily dietary intake of Cu is low, which may be attributed to low concentrations of Cu in tea.

## 2.6. Fluoride

The pathways of fluoride from acidic tea soil to tea plants are well established (Wong et al., 2003). In brief, AlF<sub>x</sub> complexes contribute to

total soluble Al and F in tea garden soils where pH is generally less than 5.5. In this pH, AlF<sub>3</sub> would decompose into Al<sup>3+</sup> and F<sup>-</sup> when free F<sup>-</sup> ion will be absorbed by tea root passively and transported to young tea leaves (Ruan & Wong, 2001). Cao, Zhao, and Liu (1997) and Cao et al. (1997) reported that tea plant is a fluoride accumulator. Fluoride is a necessary element to human health, and a moderate amount of fluoride intake is confirmed that it is the effective way of reducing dental caries among children and adult (Levi et al., 1983). However, it has been noted that drinking a large amount of tea may cause of dental and skeletal fluorosis (Cao et al., 1995). Very recently Jin, Yan, and Jianwei (2000) concluded that the endemic fluorosis that occurs in Tibet was essentially due to heavy consumption of foodstuffs prepared with brick tea.

### 2.6.1. Fluoride in tea leaves

Shu, Zhang, Lan, and Wong (2003) analyzed five different parts of tea leaves like bud with two leaves, three leaves, four leaves, five leaves and six leaves from five locations of Sichuan Province in China. Their findings revealed that the F contents in leaves increased with the age of leaves, the bud with two leaves having the lowest (range: 54–181 mg kg<sup>-1</sup>) followed by three leaves (range: 228–1723 mg kg<sup>-1</sup>), four leaves (range: 450–2622 mg kg<sup>-1</sup>), five leaves (range: 574–2965 mg kg<sup>-1</sup>) and six leaves (range: 836–2893 mg kg<sup>-1</sup>). Fung and Wong (2002) reported that a concentration of F in tea leaves depends on soil pH. For example concentration of F in tea leaves ranged from 255 mg kg<sup>-1</sup> at pH 5 and 427 mg kg<sup>-1</sup> at pH 3.5.

### 2.6.2. Fluoride in made tea

The total F contents in 17 brands of tea leaves (green, oolong, black and brick teas) in Sichuan Province, People's Republic of China ranged from 170 to 878 mg kg<sup>-1</sup>. If brick tea samples were not included, the total F contents were ranged from 170 to 423 mg kg<sup>-1</sup>. The highest F contents in brick tea was due to the use of old leaves, fallen leaves and even branches, contained the highest total F contents which were 2 to 4 times higher than other brands made from young leaves (one shoot with two leaves) (Fung et al., 1999). The lowest F content was in oolong tea leaves (170 to 224 mg kg<sup>-1</sup>). Fung et al. (2003) reported that F contents in green tea and black tea (both were made of one bud and two leaves of the small Chinese leafed variety) were 297 and 442 mg kg<sup>-1</sup> respectively.

### 2.6.3. Fluoride in tea infusion

Fluoride contents in tea infusion prepared by repeated infusion have been reported by Fung et al. (1999). Authors reported that about 53% (green tea) to 80% (brick tea) of the total F contained in the tea leaves were released into the tea liquor. Additionally, result showed that there were similar patterns of F contents among different brands of the same types of tea leaves. The highest amount of total water-soluble F was obtained in tea liquor of brick tea (4.24 ± 7.05 mg L<sup>-1</sup>), which was 4 to 5 times higher than in the other types of tea and lowest one was in tea liquor of oolong tea (1.22 ± 1.51 mg L<sup>-1</sup>). Water-soluble F content of Turkish teas was found to range from 55–127 mg L<sup>-1</sup> (Tokalioglu, Sahin, & Kartal, 2001). These levels correspond to 0.55–1.27 mg L<sup>-1</sup> when converted into infusion concentrations (2 g tea infused in 200 mL of water). Fung et al. (2003) reported that higher concentration of F was released into tea liquor under repeated infusion method than continuous infusion method. Under these experiments, authors showed that five times repeated infusion produced 1.66 mg L<sup>-1</sup> and 2.20 mg L<sup>-1</sup> F for green tea and black tea respectively. However, continuous infusion of green tea and black tea for 360 min produced 1.58 mg L<sup>-1</sup> and 1.88 mg L<sup>-1</sup> F, respectively. Kalayci and Somer (2003) measured infusion concentrations of 2.60 and 3.92 mg L<sup>-1</sup> of F with 5 and 20 min extraction times respectively in black tea. Hudaykuliev, Tastekin, Poyrazoglu, Baspinar, and Velioglu (2005) reported a range of F content for Turkish teas as 88 to 289 mg kg<sup>-1</sup> which corresponds

between 0.8 and 2.6 mg L<sup>-1</sup> infusion concentrations with the assumption of water-soluble fraction as 90%. In order to assess the levels of fluoride ingestion through intake of four black teas, Gulati et al. (1993) conducted an experiment with or without milk by Indian and English style from four different brands of tea leaves commonly available in the Indian market. The results concluded that the variation in F leached was ranged from 2.1 to 4.22 mg L<sup>-1</sup> for English style without milk and 2.17 to 4.27 mg L<sup>-1</sup> with milk. Similarly, the value range from 2.26 to 4.27 mg L<sup>-1</sup> for Indian style without milk and 1.67 to 3.20 mg L<sup>-1</sup> for Indian style with milk for a leaching time between 2 and 6 min. Among 26 black tea samples originally produced in Turkey, Sri Lanka, India and Kenya, higher fluoride levels were found in black teas that originated from Turkey (3.72 mg L<sup>-1</sup>) when compared with teas originated from Sri Lanka (0.57 mg L<sup>-1</sup>). Moreover higher fluoride levels were determined in black tea bags compared with granular and stick-shaped black teas (Emekli-Alturfan, Yarat, & Akyuz, 2009). These results also suggested that consuming tea infusions prepared from black tea available in Turkish market, in large quantities may lead to exposure to a high amount of fluoride which may cause dental fluorosis. Similar finding was also reported by (Malinowska, Inkielewicz, Czarnowski, & Szefer, 2008). The authors reported that brewing time (5, 10 and 30 min) does increase the fluoride content, which in infusions of black tea (5 min brewing) was higher than that in green tea, with contents ranging between 0.32 and 4.54 mg L<sup>-1</sup> for black tea to 0.59 to 1.83 mg L<sup>-1</sup> for green tea. On the basis of their result, the Safe and Adequate Daily Intake (SAI) and Acceptable Daily Intake (ADI) was calculated and concluded that tea quality is important to protect human against too high uptake of F from black tea, which is the most popular beverage in Poland. In addition to this, Sofuoglu and Kavcar (2008) concluded that fluoride intake from drinking teas is not high enough to cause significant risk for dental fluorosis, not to mention the bone effect or skeletal fluorosis.

## 2.7. Lead

Lead (Pb) was identified as one of the most commonly occurring contaminants in the environment (Hafen & Brinkmann, 1996). The adverse health effects caused by low-level exposure to Pb have been extensively documented and, therefore, most countries have stipulated allowable levels of Pb in food and beverages as food and beverages are among the major sources of Pb in human body (Needleman, Schell, Bellinger, Leviton, & Allred, 1990; Rosen, 1992). Nowadays, Pb concentrations in commercial tea leaves have caused concern to both consumers and producers. The allowable limit of Pb for food and beverages in Europe and China is 5 mg kg<sup>-1</sup>, in Japan it is 20 mg kg<sup>-1</sup> while in Australia, Canada, and India it is 10 mg kg<sup>-1</sup>. The guideline value of Pb for drinking water is 0.01 mg L<sup>-1</sup> (WHO, 2003e).

### 2.7.1. Lead in tea leaves

Plants can take up Pb from the soil and under certain conditions high levels of Pb can be accumulated in the leaves and other edible parts of the plant. For instance, tea plants can uptake Pb from the soils, and inevitably, a portion will be transported to the tea leaves (Jin et al., 2005). Natesan and Ranganathan (1990) reported that older tea leaves tend to contain higher concentrations of Pb than younger leaves. Pb concentration of unwashed tea leaves collected from main tea leaf producing areas in Zhejiang Province in China ranged from 0.11 to 4.55 mg kg<sup>-1</sup> with an average of 2.21 mg kg<sup>-1</sup> (Jin et al., 2005). However, washing tea leaves with distilled water decreased average Pb concentration up to 43.8% of original Pb concentration. Jin et al. (2005) also reported that decrement rates of Pb by water washing along the road were 67.2%, 76.3%, 50.8%, 58.6%, 52.7% for the tea samples 10 m, 20 m, 40 m, 60 m and 90 m away from the road edge, respectively, indicating that there were larger deposits of Pb on the surfaces of leaves

growing nearer to the roads. These results further corroborated that Pb concentrations of all of the tea leaf samples were below 5 mg kg<sup>-1</sup>, indicating the Pb contamination in tea leaves of Zhejiang Province does not exceed the allowable limits of China. Han et al. (2007) also studied the effect of liming and seasonal variation on Pb concentration of tea plants in China. They observed about 1.13 mg kg<sup>-1</sup> Pb in tea shoots, when no lime was added to soils. The high amount of Pb in tea shoots was probably because of its enhanced bioavailability in acid soils. In their study, the highest lime treatment (7 tons ha<sup>-1</sup>) increased soil pH to around 4.5 (initial pH 4.43) and produced a small positive effect on tea yield. However, Pb concentration in the new shoots was also decreased by approximately 20 to 50% with respect to non liming tea plants, indicating that liming is an effective option to reduce tea Pb if soil is overly acidic. Yemane et al. (2008) reported that, five clones of *C. assamica* variety grown at Wushwush farms in Ethiopia did not show either the ability of Pb accumulation or too low to be detected. Concentration of Pb in the leaves of eight tea cultivars grown in China was between 2.28 and 5.61 mg kg<sup>-1</sup> (Chen et al., 2009).

### 2.7.2. Lead in made tea

Han et al. (2006) analyzed Pb concentrations in 1225 tea samples collected in China for a nationwide survey during the period 1999 to 2001. The Pb concentration varied from 0.2 to 97.9 mg kg<sup>-1</sup> dry weight (DW) of tea samples, with 32% of the samples exceeding the national maximum permissible concentration (MPC) of 2.0 mg kg<sup>-1</sup> DW stipulated for China. They found that green and scented teas had lower median and mean Pb concentrations than black and oolong teas. The higher Pb concentrations for black and oolong teas can be attributed to the different types of leaves picked for different teas; more and older leaves are harvested for producing black and oolong teas than for green and scented teas. In another study, the average Pb level in the Chinese black tea samples was found to be 1.42 mg kg<sup>-1</sup> (Xie et al., 1998).

Qin and Chen (2007) reported that the concentrations of Pb in the 57 tea samples marketed in Beijing (China) varied from 0.198 to 6.345 mg kg<sup>-1</sup> dry weight (median, 0.879 mg kg<sup>-1</sup>; mean, 1.320 mg kg<sup>-1</sup>). The highest level of Pb was found in Qimen Black tea (Anhui Province), and the lowest level was found in Longjing green tea (Zhejiang Province). A report also showed that the Pb concentrations in 85 Chinese teas varied from 0.15 to 11.61 mg kg<sup>-1</sup> dry weight (Tang, Zeng, Lu, & Liang, 2003). The level of Pb in made tea that originated from different countries and imported to Pakistan, was reported to be within a range of 0.03 to 14.84 mg kg<sup>-1</sup> (AL-Oud, 2003). AL-Oud (2003) also reported that the highest concentration of Pb was found in Chinese green tea and suggested that the level was too high to be considered safe for tea drinkers. 2.31 mg kg<sup>-1</sup> of Pb in black tea and 0.01 µg L<sup>-1</sup> Pb in tea infusion in Iran was reported by Karimi et al. (2008). However, Yemane et al. (2008) found no Pb in 19 made tea samples in Ethiopia.

Among the 100 tea samples collected from South India, Seenivasan et al. (2008a) reported that the Pb content in tea samples varied between 0.04 and 1.36 mg kg<sup>-1</sup>. The lowest Pb content was found in the made tea samples collected from Munnar and the highest from Wayanad. The tea samples from Nilgiris-Wayanad and Karnataka regions of south India also showed marginally higher levels of lead. Potassic fertilizers, phosphatic fertilizers, NPK blends, manganese, zinc, boron and magnesium sources contain lead (Franklin et al., 2005). Zinc is the only micronutrient whose deficiency is widely noticed in tea and zinc sulphate is given as a foliar spray to supply zinc. Zinc sulphate, if contaminated with lead, and is applied as foliar spray, may contaminate tea plant with Pb entry into its tissues. **Spraying of copper fungicides with heavy metal impurities may increase the accumulation of Pb and Cd in tea.** Copper oxychloride is considered as one of the main sources of Pb contamination (Semu & Singh, 1996). In this study, Pb content in all the made tea samples was less than 10 mg kg<sup>-1</sup>, the limit prescribed under the Prevention of

Food Adulteration (PFA) Act of India. Ramakrishna et al. (1986) studied the Pb concentration in made tea produced in Sri Lanka and found it to vary between 0.188 and 0.561 mg kg<sup>-1</sup>.

Ashraf and Mian (2008) in a study on black tea in Saudi Arabia found that the level of Pb in the 17 tea versions ranged between 0.3 and 2.2 mg kg<sup>-1</sup>. In their study the lowest concentrations of Pb were found in Abu Jabal, Deemah and Dilmah, while the highest was encountered in Manasul. Narin et al. (2004) found maximum Pb in Turkish tea samples at the level of 27.3 ± 0.1 mg kg<sup>-1</sup>. It was indicated that the higher levels of Pb in tea samples could be attributed to the dust particle entry during tea processing and solder being used in packaging. Tsushida and Takeo (1977) reported that the concentration of Pb in green tea in Japan was found to be 0.11 to 1.93 mg kg<sup>-1</sup> with the mean of 0.49 mg kg<sup>-1</sup>. It was concluded that the green tea produced in some districts of Japan near the metropolis areas contained the highest concentration of lead. Matsuura et al. (2001) found Pb to be in the range of 0.69 to 0.73 mg kg<sup>-1</sup> for black tea samples in Japan. Chen et al. (2009) reported that among the tested toxic heavy metals, the most abundant metal in the tea leaves was Pb, which is of the most concern by tea drinkers, ranging between 2.28 and 5.61 mg kg<sup>-1</sup> among the eight cultivars. Among 41 tea plant varieties, Chen, Xu, Yu, Chen, and Shi (2010) further reported that the ratio of Pb concentration in mature leaves to that in young leaves ranged from 1.7 to 6.5 where the minimum concentration of Pb in young leaf was 1 mg kg<sup>-1</sup> and the maximum was 19.8 mg kg<sup>-1</sup>.

### 2.7.3. Lead in tea infusion

Study on Pb concentration in tea infusion has not been frequently reported. Xie et al. (1998) analyzed 39 different types of tea infusion comprising three typical kinds of tea (20 green teas, 8 black teas and 5 oolong teas) by total-reflection X-ray fluorescence method for multi-element. Authors reported that there were no distinct differences between the mean contents of Pb in green and oolong teas. In different literatures it has been seen that limited dissolution of Pb from tea leaves occurs during soakage with boiling water (Jin et al., 2005). However, soluble Pb concentration in some tea infusion could still exceed the 0.05 mg L<sup>-1</sup> limit set for drinking water by WHO (WHO, 2003e). Therefore, the Pb contamination in tea leaves remains a concern, and practices should be developed to avoid problems in the future.

## 2.8. Manganese

Manganese (Mn) has widespread significance because of their effects on acceptability as an essential element for humans and other animals. However, adverse effects of Mn can result from both deficiency and overexposure. Manganese is known to cause neurological effects following inhalation exposure, particularly in occupational settings. There have been epidemiological studies that report adverse neurological effects following extended exposure to very high levels in drinking water (Powell et al., 1998).

### 2.8.1. Manganese in tea leaves

AL-Oud (2003) suggested the ability of the tea plant to accumulate higher levels of Mn in tea leaves within the range of 390 to 900 mg kg<sup>-1</sup>. Range of Mn in five tea clones of Ethiopia was ranging between 501 and 1281 mg kg<sup>-1</sup> (Yemane et al., 2008) and level of Mn in eight different tea cultivars grown in Zhejiang University Tea Research Institute, located in Hangzhou, Zhejiang Province, China was observed within the range of 950.1 to 1224.2 mg kg<sup>-1</sup> (Chen et al., 2009). From these studies it can be concluded that Mn contents in tea leaves among cultivars were mainly attributed to cultivar variations as these plants were grown in same soils.

### 2.8.2. Manganese in made tea

Because of its biochemical importance, Mn has been the most analyzed element in tea leaves from various countries. Chute,

Weginwar, and Garg (1990) reported Mn concentrations of 802 ± 4 and 707 ± 2 mg kg<sup>-1</sup> in two Indian brands of tea leaves by a substoichiometric isotope dilution method. Kumar et al. (2005) analyzed fifteen different brands of Indian black tea leaves and seven US brands black tea. They found that in Indian tea, Mn concentration was in the range of 371 and 758 mg kg<sup>-1</sup> with a mean concentration of 575 ± 96 mg kg<sup>-1</sup> whereas in US tea it was in the range of 79–768 mg kg<sup>-1</sup> with a mean concentration of 329 ± 231 mg kg<sup>-1</sup>. A comparison of Mn concentrations in tea leaves from different countries was also performed by Kumar et al. (2005). This comparison revealed that most tea leaves from various countries have Mn content in the range of 300–900 mg kg<sup>-1</sup>, except those from Turkey and Japan where much higher Mn contents (1100–2678 mg kg<sup>-1</sup>) have been reported (Lamble & Hill, 1995; Ozdemir & Gucer, 1998). Furthermore, Mn was found between 148 and 1595.4 mg kg<sup>-1</sup> (average 824.8 mg kg<sup>-1</sup>) among forty six commercial made tea samples in Spain of which 12 were Chinese, 3 were Japanese, 3 were Indian, 2 were Kenyan, and 4 were from Sri Lanka (Fernandez-Caceres, Martin, Pablos, & Gonzalez, 2001). From the individual concentration of Mn among tea samples, authors conducted the linear discriminant analysis (LDA) in order to get a separation relating to the geographical origin. LDA clearly revealed the separation between the African (Kenya) and the Asian teas (China, Japan, Sri Lanka, and India), which appears as two well-differentiated clusters.

### 2.8.3. Manganese in tea infusion

According to the literature data, Mn is the only element with a significant dietary amount in tea, especially in black tea (Heydorn, 1988; Powell et al., 1998; Street, Száková, Drábek, & Mládkova, 2006). Concentration of Mn from two brands of Iranian and foreign tea was between 132.7 and 481 mg kg<sup>-1</sup> was reported by Kasrai, Shoushtarian, and Bozorgzadeh (1977). Manganese in tea infusion was also significantly varied with the origin of tea samples. For example Kumar et al. (2005) reported that Mn was widely different in tea infusion from India and US tea brands. To determine Mn contents in three teas (Chinese green tea, decaffeinated green tea and authentic green tea) available in USA market, Gallaher et al. (2006) analyzed tea infusions from these teas. The results reported that the infusions were a good source of Mn within the range between 1.86 and 2.48 mg L<sup>-1</sup> tea infusion. On comparing Mn into solution from black (Assam, Darjeeling from India; Lapsang Souchong from China) and green teas (Kashmir Green from India; Gunpowder Green from China), Mehra and Baker (2007) concluded that the percentage of Mn transfer for green teas was lower than that of black teas for all infusion times. Pohl and Prusisz (2007) analyzed three black tea and green tea samples each for Mn concentration in infusion available in Poland and analyzed results showed that 1.56 to 6.02 mg L<sup>-1</sup> and 1.53 to 2.02 mg L<sup>-1</sup> Mn was available in black and green tea infusion respectively. Fractionation pattern of Mn in these samples showed that the broadly meant cationic species of Mn was the most abundant, i.e., contributed to 53–68% of the totally dissolved metal content in the black tea infusions and to 80–82% of the whole metal amount in the brews of the green teas. The fraction of Mn bound to polyphenols contributed to 31–46% (39% on average) in case of the black teas and from 13% to 20% (16% on average) in case of the green teas. This difference between the tea species can be presumably attributed to the fact that in the manufacturing processes subjected to the black teas, the flavanoids present in the leaves polymerize into more complex and condensed polyphenols of a much higher molecular weight. The concentration of Mn in the residual metal fraction, likely attributed to the anionic and/or neutral metal species associated with small ligands, was found to be the lowest, i.e., from 0.5% to 3% of the total metal content. Therefore, the fractionation pattern assessed for Mn indicates that the complexation of this metal in the tea infusions is not so simple (Pohl & Prusisz, 2007). Ozdemir and Gucer (1998) reported that 30% Mn by water extracted tea infusion was present in the form of Mn

(II), and also 2.5% of Mn was distributed in the total organic bound and was passed into the various solvent.

The guideline value of Mn in drinking water prescribed by WHO is  $0.4 \text{ mg L}^{-1}$  (WHO, 2003f) and estimated safe and adequate daily dietary intake is 2–5 mg (Powell et al., 1998). Nookabkaew et al. (2006) calculated that average daily dietary intakes of Mn through tea infusion were 0.413–2.225 mg per day. Results from Mehra and Baker (2007) showed that in general, tea provides about 35.5% of available Mn (except 115.5% available Mn from the Tetley tea bag sample), as a percentage of the average daily dietary intake. Hence, tea drinking may be regarded as a major source of essential dietary Mn. However, Tetley tea bag sample is of some concern as high Mn in the diet can lead to long-term toxicity. As this finding is based on only one tea bag sample, further studies are needed to confirm the findings and its related consequences.

## 2.9. Nickel

It is almost impossible to visualize soil without trace levels of heavy metals like nickel (Ni) (Adriano, 1986) which is one of the most important heavy metals in terms of its potential toxicity to plants and animals (Bazzaz, Carlson, & Rolfe, 1974; Gouugh, Shacklette, & Case, 1979; Kabata-Pendias & Pendias, 2001). Activities such as mining and agriculture have polluted extensive areas throughout the world by Ni (McLaughlin, 2002; Everhart et al., 2006; Peltier, Allada, Navrotsky, & Sparks, 2006). It is well known that an element like Ni is essential for plant growth at low concentrations (Brady & Weil, 1999; Reeves & Baker, 2000). Nevertheless, beyond certain threshold concentrations, this element becomes toxic for most plant species (Brady & Weil, 1999; Peralta-Videa et al., 2002).

### 2.9.1. Nickel in made tea

Among 100 analyzed black tea samples from South India, Seenivasan et al. (2008a) reported that tea contained 1.1 to  $5.3 \text{ mg kg}^{-1}$  Ni. Among the analyzed samples the lowest nickel (Ni) content was found in Nilgiris teas ( $1.1 \text{ mg kg}^{-1}$  Ni) and highest in tea from Vandiperiyar and Peermedu ( $5.3 \text{ mg kg}^{-1}$  Ni) in south India. Marcos et al. (1998) reported that Ni in made tea sample ranged between 2.99 and  $22.6 \text{ mg kg}^{-1}$ . Franklin et al. (2005) reported that potassic fertilizers contained 2.7 to  $16 \text{ mg kg}^{-1}$  nickel as impurity while commercial phosphatic fertilizers contained 19 to  $24 \text{ mg kg}^{-1}$  Ni. It was further reported that manganese sources contained 340 to  $613 \text{ mg Ni kg}^{-1}$ , zinc sources contained 241 to  $2399 \text{ mg Ni kg}^{-1}$ , boron sources  $39 \text{ mg kg}^{-1}$  and magnesium sources contained  $77 \text{ mg kg}^{-1}$ . It is evident that Ni mainly comes through the foliar and soil application of low quality fertilizers and micro nutrients. Since Ni is a toxic element, not having any tolerance limit in tea, the agro inputs used in tea fields should be analyzed for heavy metal impurities.

Ferrara et al. (2001) analyzed various commercial tea samples coming from China, Russia and Syria. The analysis revealed that teas from Syria and Russia contained 131.6 to  $1964 \text{ mg kg}^{-1}$  and  $97.2 \text{ mg kg}^{-1}$  to  $2025 \text{ mg kg}^{-1}$  Ni respectively. Nookabkaew et al. (2006) analyzed popular herbal tea products from Thailand by using ICP-MS. They reported that the Ni content in herbal tea product was 2.281 to  $9.194 \text{ mg kg}^{-1}$  with average of  $5.633 \text{ mg kg}^{-1}$  on dry weight basis. However the mean concentrations of Ni in tea infusions and the percent of Ni released into infusion was  $8.044 \mu\text{g}/100 \text{ mL}$  and 67.71% respectively.

Moreda-Piñeiro et al. (2003) analyzed Ni concentration in 85 tea samples (36 samples from Asian countries, 18 samples from African countries, 24 commercial blends and seven samples of unknown origin). The data showed that the average values of Ni concentrations in tea samples from China was  $4.92 \text{ mg kg}^{-1}$  ( $n = 13$ ) and from India and Sri Lanka it was  $4.08 \text{ mg kg}^{-1}$  ( $n = 13$ ). The average concentration of Ni in Asian tea was  $4.24 \text{ mg kg}^{-1}$ . However, Ni concentration

in tea samples collected from Africa was found to be  $4.76 \text{ mg kg}^{-1}$ . Feng, Wang, and Li (2003) analyzed Ni from three tea samples (a green tea and an oolong tea from China and a black tea from Singapore) by a capillary electrophoresis method. The Ni concentration in the three tea samples ranged from 3.07 to  $7.51 \text{ mg kg}^{-1}$ . In another study, Ni was found to be 10.8 to  $38.8 \text{ mg kg}^{-1}$  with the mean concentration of  $23.3 \text{ mg kg}^{-1}$  among fourteen tea samples produced and marketed in Turkey (Narin et al., 2004).

### 2.9.2. Nickel in tea infusion

Concentrations of Ni in tea infusion are very limited. Five Aamallais black tea infusion of South India released 59.8% of total Ni in made tea when 1 g tea sample was brewed for 1 min however, it was 74.9% when tea was brewed for 5 min (Natesan & Ranganathan, 1990). Similarly, Salahinejad and Aflaki (2010) reported that Ni from black tea was infused moderately (22–55% of total Ni). Nookabkaew et al. (2006) reported that among different herbal tea products consumed in Thailand, the highest average levels of Ni were detected in *C. sinensis* infusion, prepared by 2 g of herbal tea powders in 100 mL water. Among the infusions of black, green and oolong tea taking four each and infusions of six white teas, Ni was found between  $0.026 \text{ mg L}^{-1}$  and  $0.269 \text{ mg L}^{-1}$  and Ni was found significantly higher in decaffeinated black Japan Sencha tea (Malik et al., 2008). With respect to the acceptable daily intake of Ni (<1 mg per day) as toxic elements in daily dietary and safety standards, the infusion of black tea samples analyzed in different study were found to be safe for human consumption (Nookabkaew et al., 2006; Street et al., 2006; Seenivasan et al., 2008a; Salahinejad & Aflaki, 2010).

## 3. Conclusions

There is no denial the fact that tea is the most popular beverage in the world, next to water and therefore, it can be recognized as an important part of a sound diet. However, from the present literature survey, strong evidence has been shown that oral intake of different trace elements may take place from tea as tea plants have a genetic potential to uptake nonessential trace elements. The presence of trace elements in all analyzed tea samples surveyed in this review was within the safe limits towards human beings, but it appeared that it still provides a significant additional source of trace elements. However, it is still difficult to find out the adverse human health effect of tea related to trace elements and at the same time it is also difficult to translate diverse scientific findings into public health messages and setting a health based standard for trace elements. Besides this, toxicity of trace elements depends not only on the total amount of the metal but also on the existing species. Therefore, total element determination is not adequate for risk assessment. However, except few elements (particularly Al), much less attention has been paid to elemental speciation in tea and tea infusion and therefore, it is difficult to form a consistent picture of the speciation of discussed trace elements in tea infusions from the studies published until today. Despite significant research efforts, more research is required to comprehend the science of accumulation of trace elements by tea plants, specification of uptake trace elements and the effect of trace elements on human health by regular consumption of tea. However, concentrations of trace elements in tea products could be reduced through variatal selection, use of young leaves and the agronomic practices to lower uptake of trace elements and are very essential.

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