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Metal concentrations in traditional and herbal teas and their potential risks to human health

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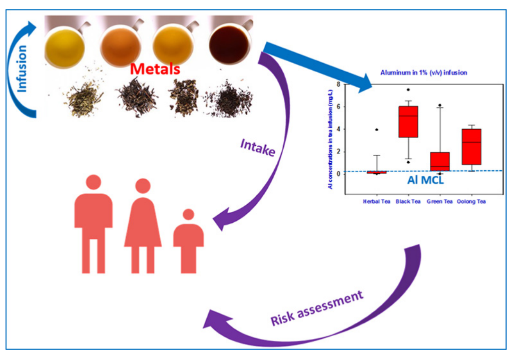
HIGHLIGHTS

- Total and fusion concentrations of Al, As, Cd, Cr, and Pb were determined in 47 teas.
- Cr in 47% herbal and 73% of traditional teas exceeded Canadian Cr limit at 2 mg kg^{-1}.
- Al was higher in traditional teas (50.3–2517 mg kg^{-1}) and infusion (0.02–7.51 mg L^{-1}).
- Al was lower in herbal teas (47–1745 mg kg^{-1}) and infusion (0.09–3.95 mg L^{-1}).
- All black tea & 83, 75 & 25% green, oolong & herbal teas >secondary MCL of 0.2 mg L^{-1}
- Tea consumption may contribute Al and Cr intake, especially for black tea.

GRAPHICAL ABSTRACT



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ABSTRACT

Food and beverage consumption is an important route for human exposure to metals. Traditional tea (*Camellia sinensis*) is a widely-consumed beverage, which may contain toxic metals. This study determined total and infusion concentrations of 5 metals including Al, As, Cd, Cr, and Pb in 47 traditional and herbal teas from 13 countries and assessed their potential risks to human health. The data showed that herbal teas exhibited higher As (0.26 mg kg^{-1}), Cd (0.19 mg kg^{-1}) and Pb (2.32 mg kg^{-1}) than traditional teas. Black tea from India had high Cr at 31 mg kg^{-1} while white tea from China had low Cr at 0.39 mg kg^{-1}. Arsenic, Cd and Pb did not exceed the WHO limit for medicinal plants excluding one herbal tea with 1.1 mg kg^{-1} As and 26.4 mg kg^{-1} Pb. However, Cr in 47% herbal teas and 73% traditional teas exceeded the Canada limit of 2 mg kg^{-1}. Metal concentrations in tea infusions were below the MCL for drinking water except for Al. Total Al and its infusion was lower in herbal teas (47–1745 mg kg^{-1} and 0.09–3.95 mg L^{-1}) than traditional teas (50.3–2517 mg kg^{-1} and 0.02–7.51 mg L^{-1}), with 0.9–22% and 4–49% of the Al being soluble in infusion. The Al concentrations in infusion in all black tea and 83, 75 and 25% of the green, oolong and herbal teas exceeded the secondary MCL for drinking water at 0.2 mg L^{-1}. However, the weekly intake of Al from drinking tea (0.001–0.39 and 0.003–0.56 mg kg^{-1} for children

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and adults) was lower than the provisional tolerable weekly intake for Al at 1.0 mg kg^{-1}. Our data showed that it is important to consider metal intake from tea consumptions, especially for Cr and Al in heavy tea drinkers.

1. Introduction

Since traditional teas (*Camellia sinensis*) contain polysaccharides, caffeine, polyphenols and amino acids, they are a popular drink (Nkansah et al., 2016). Based on fermentation degree and tea color, there are six types of tea including green, black, oolong, dark, white, and yellow (Zhang et al., 2015). Green and yellow teas are non-fermented, and oolong and white teas are semifermented, while dark and black teas are fully fermented. Unlike traditional teas, herbal teas may or may not possess *C. sinensis* leaves.

Tea is the second most consumed beverage worldwide, representing a significant part of the human diet. Originating from China, tea consumption has spread globally over the past 2000 years, becoming one of the most popular beverages in the world (Karak and Bhagat, 2010). The Food and Agriculture Organization (FAO) estimated that ~4.8 million tons of teas were consumed worldwide in 2013, with 1.0–1.6 million tons of teas being consumed in China and India (FAO, 2015). In 2015, Americans consumed over 80 billion servings of tea, with 85% being black tea and 14% being green tea. Studies showed that *C. sinensis* is an Al-hyperaccumulator, accumulating both Al and other metals (Karak and Bhagat, 2010). Aluminum exhibits evidence of toxicity to living organisms, which is of health concern as excessive exposure has been associated with an increased risk of Alzheimer's disease (Gupta et al., 2005).

The primary dietary source of Al in the US is from foods and beverages including teas. The allowable Al intake in food is 5 mg day^{-1}, with the USEPA's secondary maximum contaminant level (MCL) in drinking water being at 0.2 mg L^{-1} (ATSDR, 2008). According to Yokel and Florence (2008), drinking water provides ~0.1 mg of Al, accounting for ~2% of daily Al intake. In countries where Al from other sources is low and tea consumption is high, tea consumption may contribute up to 50% of daily Al intake (UKMAFF, 1993; Yokel and Florence, 2008). While elements such as Cu, Mn, Fe and Zn are essential nutrients for plants and animals, metals like As, Cd, Cr and Pb are toxic to humans even at low concentrations, which may cause mutagenic, teratogenic and carcinogenic toxicity (Abdul et al., 2015). For example, the high stomach cancer rate in the Van region of Turkey was closely related to the high levels of Cd, Pb, Cu, and Co in their soils, fruits and vegetables (Türkdoğan et al., 2002).

Depending on tea origin, metal accumulation can occur naturally or result from manufacturing and agronomic processes. This includes application of pesticides and fertilizers, and plant uptake from acidic soils where tea plants are cultivated (Brzezicha-Cirocka et al., 2016; Karak and Bhagat, 2010). Elevated Al concentrations in teas have been reported in different countries including China (Fung et al., 2009), Brazil (Milani et al., 2016), Iran (Ghoochani et al., 2015, Parviz et al., 2015), and Czech Republic (Malik et al., 2013). For example, the Al concentration in tea infusions was at 0.70–5.93 mg L^{-1} in teas from Hong Kong, which is ~30 times higher than USEPA's secondary MCL for Al in drinking water at 0.2 mg L^{-1} (Fung et al., 2009). In addition to Al, Seenivasan et al. (2008) reported Cr concentrations of 1.1–21 mg kg^{-1} in 100 teas from India. Ning et al. (2011) found Pb content at 0.26–3.2 mg kg^{-1}, As at 0.035–0.24 mg kg^{-1}, Cu at 12–22 mg kg^{-1}, and Cd at 0.0059–0.085 mg kg^{-1} in 30 different teas from China. However, there is no report on the levels of Al, As, Cd, Cr, and Pb in teas consumed in the US. The main route of human exposure to these metals are through dietary intake and continuous exposure may lead to nervous, bone and kidney diseases, and cardiovascular disfunctions (WHO, 1992). Therefore, it is important to investigate toxic metals in traditional and herbal teas to reduce their risks to humans.

We determined the total concentrations of Al, As, Cd, Cr, and Pb in 47 traditional and herbal teas from US markets. In addition, metal concentrations in tea infusion were also determined to assess their potential risks to humans. The results from this study can shed light on the potential risk of tea consumption on metal intake by heavy tea drinkers.

2. Material and methods

2.1. Collection of tea samples

A total of 47 samples representing four types of tea (herbal, black, green and oolong) were collected in the US, which originate from 13 countries (Brazil, Canada, China, Ecuador, India, Nepal, Pakistan, Peru, Spain, Tanzania, Uganda, UK and USA). They included 16 herbal, 16 black, 11 green and 4 oolong teas (Table 1). The contents of Al, As, Cd, Cr and Pb were determined both in total concentrations and in 5-minute infusions in boiling water.

2.2. Total metal contents in teas and 5-minute tea infusion

Tea samples were dried at 65 °C to constant weight and ground into fine powder to obtain a representative sample. About 0.5 g of the sample was digested with HNO_3/H_2O_2 using USEPA Method 3050B on a hot block (Environmental Express, Ventura, CA). All elements from the digested samples were analyzed by inductively coupled plasma mass spectrometry (ICP-MS; Perkin-Elmer Corp., Norwalk, CT).

Tea infusions were prepared by adding 50 mL of boiling double DI-water to 0.5 g of tea leaves in a 50 mL conical flask. The tea infusion was mixed using a glass rod to ensure adequate wetting, then covered and allowed to boil for 5 min based on tea industry's recommended brew time. The solution was filtered through a Whatman N. 40 filter, cooled, and diluted with double DI-water to 50 mL. Concentrations of Al, As, Cd, Cr and Pb were then determined via ICP-MS. A standard solution was prepared by dilution of a 1000 mg L^{-1} stock solution (Merck) prior to use. Double DI-water was used throughout this experiment.

2.3. Health risk assessment

In this study, potential risk of metal exposure from tea ingestion was assessed. The weekly intake of metals from tea ingestion was estimated following USEPA (1992):

$$\text{TWI} = C \times \text{WI}/\text{BW} \quad (1)$$

where TWI is the tolerable weekly intake (mg kg^{-1} BW per week), C is the metal concentration in tea infusion (mg L^{-1}), WI is the average weekly intake rate of tea (L week^{-1}), and BW is body weight (kg). For children and adults, default BW is 10 kg and 70 kg.

Potential health risks from ingesting metals in teas were estimated for both children and adults. As per Sofuoglu & Kavcar (2008), an estimated of weekly tea intake of 0.525 L by children <15 and 5.25 L by adults >35 years were consumed.

2.4. Quality control and data analysis

All reagents including HNO_3 and H_2O_2 were of analytical reagent grade (Merck). All glassware and equipment were soaked with 10% HNO_3 and HCl for 8 h and then rinsed with double DI-water prior to use. Certified reference material (Tomato leaves SRM1573a, NIST MD, USA) was analyzed as samples with each batch. The results (Al = 560