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

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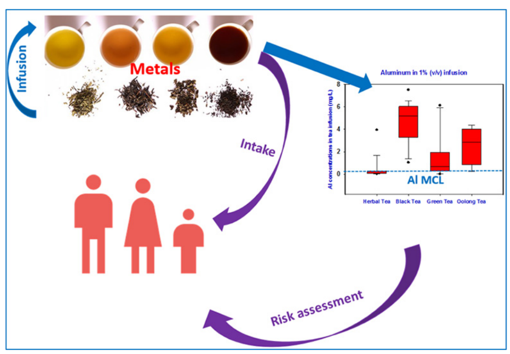
HIGHLIGHTS

- 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):

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

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

Table 1
Tea samples collected for the study.

Tea Type	Country of origin	Shape	Tea type and ingredients
Herbal			
Tonghai	China		Herbal: buckwheat tea
Clipper	USA	Tea bag	Herbal mix: chamomile
Herbal tea	USA	Tea bag	Herbal mix: chamomile
Stash Premium	USA	Tea bag	Herbal mix: chamomile
Runa	Ecuador		Herbal mix: guayusa tea
Yogi	USA	Tea bag	Herbal mix: carob pod
Matte Leao	Brazil		Herbal mix: yerba mate shrub
Bioherbal	Peru		Herbal mix
Traditional medicine	USA	Tea bag	Herbal mix: lemon Echinacea
Tajo	USA	Tea bag	Herbal mix: chamomile flowers
Lipton	Canada	Tea bag	Herbal mix: chamomile flowers
Wuyutai	China	Needle-like	Herbal mix: with jasmine
Carmencita	Spain		Herbal mix
Maracuja farms	Brazil		Herbal mix
Boldus	Brazil	Barks	Herbal mix: boldus
Carcueja	Brazil		Herbal mix
Black			
Brooke bond	India	Granular	Black tea
Twinings	UK	Tea bag	Black tea
Twinings	UK		Black tea
African Pride	Tanzania	Granular	Black tea
Garden Eagle	Uganda	Granular	Black tea
Stash Premium	USA	Tea bag	Black tea
Twinings	UK		Black tea
Cha Preto Leao	Brazil	Tea bag	Black tea
Madui Zhang	China	Compressed ball	Black tea
Lapsang Sauchang	China	Granular	Black tea
Brooke Bond	India	Tea bag	Black tea
kte	Nepal	Tea bag	Black tea
Lipton	Pakistan	Granular	
Tajo	USA	Tea bag	Black tea
Lipton	Pakistan	Granular	Black tea
Lipton		Granular	Black tea
Green tea			
Bigelow	USA	Tea bag	Green tea
Lipton	USA	Tea bag	Green tea
Yogi	USA	Tea bag	Green tea
Revolution tea	USA	Tea bag	Green tea
Mast Chang	China	Granular	Green tea
Chayuchanxin	China	Ball	Green tea
West lake Long Jing	China	Leaf-like	Green tea
Huangshan Maofeng	China	Needle-like	Green tea
Wuyutai	China	Needle-like	Green tea
West lake Long Jing	China	Leaf-like	Green tea
Wuyutai	China	Needle-like	Green tea
Oolong			
Tie Yin	China	Tea bag	Oolong tea
Dongdingwulong	China	Granular	Oolong tea
Lingfeng	China	Tea bag	Oolong tea
Gaoshan	China	Granular	Oolong tea

± 11; As = 0.110 ± 0.003; Cd = 1.59 ± 0.005; Cr = 1.86 ± 0.110 μg g^{-1}) were in agreement with the certified values (Al = 598 ± 12; As = 0.112 ± 0.004; Cd = 1.52 ± 0.04; Cr = 1.99 ± 0.06 μg g^{-1}). QA/QC protocols included triplicate analyses and the use of standard and spiked solutions every 10 samples. The mean recovery of standard solutions was 96 ± 1.5%, while the spike recovery was 94 ± 3.6%. The performance of the ICP-MS was checked by running an intermediate calibration standard every 10 samples. All calibration standard checks were within the acceptable range (80–120%).

All data are expressed as means of three replicates with standard error. Analyses of variance (ANOVA) by Tukey's multiple grouping were used to determine significance differences among different teas.

3. Results and discussions

The benefits of drinking tea are multiple, contributing to daily intake of essential minerals and benefit overall human health. However, teas may also contain toxic metals including Al (Table 2), and As, Cd, Cr and Pb (Table 3). The mean Al content in herbal teas was 548 mg kg^{-1}, which was lower than 885, 1089 and 1655 mg kg^{-1} in green, oolong and black teas. All traditional teas had detectable metal concentrations and the mean concentrations for Cd, As, Pb and Cr was 0.04, 0.19, 0.70 and 4.58 mg kg^{-1}, with Cr being the highest among 4 metals (Table 3 and Fig. 1). Compared to traditional teas, herbal teas exhibited higher mean concentrations of Cd, As, and Pb (0.19, 0.26 and 2.37 mg kg^{-1}) than traditional teas. Among teas from different countries, black tea from India had high Cr at 31.2 mg kg^{-1}, and green tea from China had low Cr at 0.41 mg kg^{-1} (Table 3). Regulations regarding metals in medicinal plants and plant-based products have been established for several countries, including Brazil, Canada, China, German, India, Japan and Thailand (Table 4). However, in the US there is no regulation on metal content in teas. WHO (2007) recommends MCLs in plant materials for Cd, As and Pb are at 0.3, 1.0 and 10 mg kg^{-1}. However, no value for Cr though hexavalent Cr is toxic (WHO, 2007).

Table 2
Aluminum in tea samples and infusion, and percent of Al and pH in tea infusion.

Tea type	Al in tea (mg kg^{-1})	Al in 1% (w/v) infusion (mg L^{-1})	% of Al infused	pH of infusion
Herbal				
Tonghai	1745	3.95	22.0	5.50
Clipper	1287	0.17	1.30	5.30
Herbal tea	777	0.13	1.60	5.30
Stash Premium	739	0.06	0.90	5.30
Runa	571	0.70	12.0	5.50
Yogi	553	0.09	1.60	5.50
Matte Leao	512	0.68	13.0	5.50
Bioherbal	489	0.08	1.70	5.40
Traditional medicine	407	0.06	1.50	5.10
Tajo	332	0.10	2.90	5.30
Lipton	291	0.13	4.40	5.10
Wuyutai tea	291	0.34	12.0	4.60
Carmencita	261	0.05	1.90	5.10
Maracuja Farms	240	0.05	2.00	4.90
Boldus	229	0.04	1.90	5.40
Carcueja	47.0	0.09	20.0	4.80
Mean	548 ± 111	0.42 ± 0.25	6.29 ± 1.84	5.23 ± 0.07
Black				
Brooke Bond	2517	7.51	29.0	4.30
Twinings	2384	6.05	25.0	4.40
Twinings	2285	6.05	26.0	4.30
African Pride	2232	6.00	26.0	4.70
Garden Eagle	2080	5.70	27.0	4.60
Stash Premium	1971	5.51	27.0	4.80
Twinings	1735	3.55	20.0	4.40
Cha Preto Leao	1518	5.28	34.0	4.30
Madui Zhang	1510	1.51	14.0	3.90
Lapsang Sauchang	1472	5.11	34.0	4.10
Brooke bond	1409	4.60	32.0	4.20
Kte	1406	2.66	18.0	4.30
Lipton	1269	3.56	28.0	4.10
Tajo	1229	6.12	49.0	4.60
Lipton	1168	3.20	27.0	4.00
Lipton	300	1.05	35.0	4.10
Mean	1655 ± 147	4.59 ± 0.47	28.2 ± 2.06	4.35 ± 0.07
Green				
Bigelow	2286	5.20	23.0	4.70
Lipton	1964	6.13	31.0	5.30
Yogi	1533	1.22	7.90	4.50
Revolution	1500	1.96	13.0	4.80
Mast Chang	969	0.97	27.0	4.10
Chayuchanxin	426	0.67	16.0	3.80
West lake Long Jing	300	0.28	9.30	4.20
Huangshan maofeng	259	0.31	12.0	4.10
Wuyutai tea	229	0.31	14.0	4.10
West lake Long Jing	221	0.14	6.50	3.80
Wuyutai	50.3	0.02	4.00	4.40
Mean	885 ± 253	1.56 ± 0.67	14.8 ± 2.75	4.35 ± 0.14
Oolong				
Tie Yin	1797	4.36	24.0	4.30
Dongdingwulong	1272	3.05	16.0	3.80
Lingfeng	969	2.64	31.0	3.90
Gaoshan	318	0.26	8.10	4.40
Mean	1089 ± 356	2.58 ± 0.99	19.8 ± 5.72	4.10 ± 0.17
Mean Traditional Tea	1309 ± 135	3.3 ± 0.43	22.4 ± 1.88	4.30 ± 0.06
Mean All Tea	1050 ± 192	2.29 ± 0.62	16.9 ± 3.11	4.61 ± 0.14

3.1. Al contents in tea leaves and tea infusions

Aluminum is one of the most abundant metals in soils (Ochmański and Barabasz, 2000). After drinking water, tea is the most consumed beverage in the world, thereby making it a major dietary source of Al. Tea plants are known to accumulate Al, which can reach 8700–23,000 mg kg^{-1} in the leaves (Chen et al., 2006; Ghoochani et al., 2015).

For all teas, its pH in infusion was acidic, being 3.8–5.5 (Table 2). The mean Al concentrations were 584 in herbal, 885 in green, 1089 in oolong, and 1655 mg kg^{-1} in black teas (Fig. 2A). Herbal tea from Brazil had low Al at 47 mg kg^{-1} and black tea from India has high Al at 2517 mg kg^{-1}. Fung et al. (2009) found that Al concentration in old leaves of mature plants in black teas was as high as 19,316 mg kg^{-1}, which is 11 times higher than the present study. The mean Al content in traditional teas was higher than herbal teas (1309 vs. 584 mg kg^{-1}), consistent with Polechońska et al. (2015) who reported 961 mg kg^{-1} Al in traditional teas.

Similarly, Al contents in infusions for herbal, green, oolong and black teas were 0.04–3.95, 0.02–6.13, 0.26–4.36, and 1.05–7.51, mg L^{-1}, accounting for 0.9–49% Al being solubilized after 5 min in boiling water. The data were consistent with 6.0 mg L^{-1} reported by Fernandez-Calvino et al. (2008). Among teas, black tea infusions contained highest

Table 3
As, Cd Cr and Pb concentrations in tea leaves and infusion and percentage of release from traditional and herbal teas.

Tea Brand	Tea Type	As			Cd			Cr			Pb		
		Tea mg kg^{-1}	Infusion μg L^{-1}	% Infusion	Tea mg kg^{-1}	Infusion μg L^{-1}	% Infusion	Tea mg kg^{-1}	Infusion μg L^{-1}	% Infusion	Tea mg kg^{-1}	Infusion μg L^{-1}	% Infusion
Bioherbal	Herbal	1.10	1.02	9.00	0.31	0.16	5.00	4.60	1.00	2.28	0.90	1.10	12.2
Clipper		0.40	0.73	18.0	0.05	nd	–	5.40	1.60	3.00	0.70	2.20	31.0
Maracuja Farms		0.30	0.19	6.00	0.06	nd	–	1.80	0.90	5.06	0.70	2.00	28.0
Carmencita		0.30	0.19	6.00	0.12	0.19	15.0	3.00	1.40	4.60	26.4	19.9	7.50
Tajo		0.30	0.44	15.0	0.21	nd	–	3.30	1.90	5.82	0.60	1.20	20.0
Tonghai		0.27	1.25	46.0	0.05	nd	–	1.53	2.20	14.4	2.37	2.90	5.99
Runa		0.20	0.72	36.0	0.82	0.41	5.00	1.30	1.10	8.69	0.20	1.60	80.0
Matte Leao		0.20	nd	9.00	0.42	0.61	14.0	2.20	4.40	20.2	0.60	3.30	55.5
Stash Premium		0.19	0.48	25.0	0.31	0.40	13.0	3.29	1.10	3.43	0.79	3.60	45.5
Traditional medicine		0.18	0.84	47.0	0.22	0.18	8.00	2.07	1.60	7.97	0.33	1.80	54.5
Yogi		0.14	0.36	26.0	0.09	0.13	14.0	1.27	0.90	7.72	0.74	1.10	14.8
Herbal tea		0.10	0.60	60.0	0.06	nd	–	1.64	1.20	7.13	0.36	1.40	38.8
Carcueja		0.10	nd	–	0.05	nd	–	1.40	0.90	7.00	0.20	2.50	100
Boldus		0.08	nd	–	0.02	nd	–	0.97	0.90	9.28	0.35	1.20	34.2
Lipton		0.05	0.33	66.0	0.09	0.14	15.0	1.06	1.40	13.1	0.24	0.90	37.5
Mean		0.26 ± 0.07	0.60 ± 0.09	28.3 ± 6.00	0.19 ± 0.06	0.28 ± 0.07	11.1 ± 1.66	2.32 ± 0.36	1.52 ± 0.24	7.98 ± 1.28	2.37 ± 1.78	3.11 ± 1.26	37.7 ± 7.10
Lipton	Black	0.30	0.27	9.00	0.06	nd	–	6.90	6.50	9.48	0.50	0.90	18.0
Brooke bond		0.30	0.27	9.00	0.02	nd	–	31.2	11.0	3.56	0.70	1.40	20.0
Twinings		0.30	nd	–	0.06	nd	–	9.00	3.70	4.12	0.80	1.10	13.7
Twinings		0.30	0.48	16.0	0.02	nd	–	2.70	2.10	7.85	1.10	3.90	35.4
Brooke Bond		0.30	nd	–	0.05	nd	–	10.6	5.30	5.02	0.50	1.40	28.0
Lipton		0.20	0.41	21.0	0.05	nd	–	13.7	7.40	5.46	0.30	0.90	30.0
Cha Preto Leao		0.20	nd	–	0.01	nd		1.72	0.90	5.47	0.30	1.60	53.3
Lipton		0.20	0.34	17.0	0.05	nd	–	12.1	3.70	3.10	0.30	0.21	7.00
Kte		0.20	nd	–	0.03	0.27	9.00	2.90	1.90	6.62	0.80	2.30	28.7
Madui Zhang		0.19	nd	–	0.04	nd	–	1.42	nd	–	nd	nd	–
Twinings		0.15	0.34	23.0	0.06	nd	–	1.63	2.80	17.6	1.90	8.40	44.2
Tajo		0.06	nd	–	0.19	0.19	10.0	2.17	1.10	5.21	0.67	1.60	23.8
Lapsang Sauchang		0.05	nd	–	0.01	nd	–	1.43	2.40	16.9	0.35	1.90	54.2
Stash Premium		0.05	nd	–	0.04	nd	–	2.77	4.20	15.2	0.25	1.80	72.0
Garden Eagle		0.36	0.24	6.60	nd	nd	–	19.7	10.0	5.43	1.11	3.00	27.0
African Pride		0.23	0.31	13.4	nd	nd	–	4.45	3.00	6.76	0.26	0.95	36.5
Caykur		0.32	0.20	6.66	nd	nd	–	1.03	0.80	7.76	0.40	1.15	28.8
Mean		0.22 ± 0.02	0.32 ± 0.03	13.5 ± 2.16	0.05 ± 0.01	0.23 ± 0.57	9.50 ± 0.71	7.37 ± 2.04	4.20 ± 0.80	7.84 ± 1.19	0.64 ± 0.11	2.03 ± 0.49	32.5 ± 1.29
Bigelow	Green	0.70	0.77	11.0	0.07	nd	–	3.50	2.70	7.89	1.70	2.20	12.9
Yogi		0.50	1.33	27.0	0.08	0.17	21.0	4.60	2.10	4.57	1.00	1.01	10.1
Revolution tea		0.30	0.58	19.0	0.08	nd	–	2.50	1.30	5.52	1.00	2.30	23.0
Chayuchanxin		0.09	nd	–	0.02	nd	–	0.85	0.90	10.4	0.75	1.60	21.3
Mast Chang		0.08	nd	–	0.05	nd	–	1.37	nd	–	0.55	2.40	43.6
Wuyutai tea		0.06	nd	–	0.01	nd	–	0.42	1.00	24.1	0.37	2.80	75.6
West lake Long Jing		0.06	nd	–	0.01	nd	–	0.75	0.89	11.8	0.77	1.70	22.0
West lake Long Jing		0.05	0.18	36.0	0.04	nd	–	0.73	0.80	11.3	1.08	4.00	37.0
Lipton		0.05	nd	–	0.02	nd	–	1.49	3.20	21.8	0.48	1.42	29.5
Huangshan meofeng		0.04	0.18	45.0	0.02	nd	–	0.41	1.10	28.7	0.40	2.20	55.0
Wuyutai tea		0.01	nd	–	0.01	nd	–	0.45	0.90	21.1	0.36	nd	–
Mean		0.18 ± 0.07	0.60 ± 0.24	27.6 ± 6.72	0.04 ± 0.01	0.17 ± 0.00	21.0 ± 0.00	1.55 ± 0.42	1.52 ± 0.27	14.7 ± 2.56	0.76 ± 0.12	2.16 ± 0.25	33.0 ± 6.43
Dongdingwulong	Oolong	0.13	0.53	41.0	0.07	nd	–	0.69	1.10	15.9	1.11	0.21	1.89
Lingfeng		0.08	0.18	23.0	0.05	nd	–	1.37	2.80	20.8	0.55	2.40	43.6
Tie Yin		0.07	nd	–	0.02	nd	–	1.35	0.90	7.19	0.40	2.10	52.5
Gaoshan		0.07	nd	–	0.05	nd	–	0.46	0.70	15.8	0.96	5.30	55.2
Mean		0.08 ± 0.01	0.35 ± 0.24	32.0 ± 12.7	0.04 ± 0.01	–	–	0.97 ± 0.27	1.41 ± 4.71	14.9 ± 4.87	0.75 ± 0.19	2.50 ± 0.25	38.2 ± 6.43
Mean Traditional Tea		0.19 ± 0.03	0.41 ± 0.05	22.9 ± 2.26	0.04 ± 0.01	0.21 ± 0.01	11.1 ± 1.53	4.58 ± 1.20	2.91 ± 0.48	±1.26	0.70 ± 0.56	2.14 ± 0.45	33.5 ± 3.33
Mean All Teas		0.21 ± 0.03	0.49 ± 0.01	23.8 ± 0.42	0.09 ± 0.15	0.26 ± 0.15	13.3 ± 4.71	3.86 ± 0.83	2.41 ± 0.35	10.0 ± 0.97	1.24 ± 0.56	2.46 ± 0.45	34.8 ± 3.14

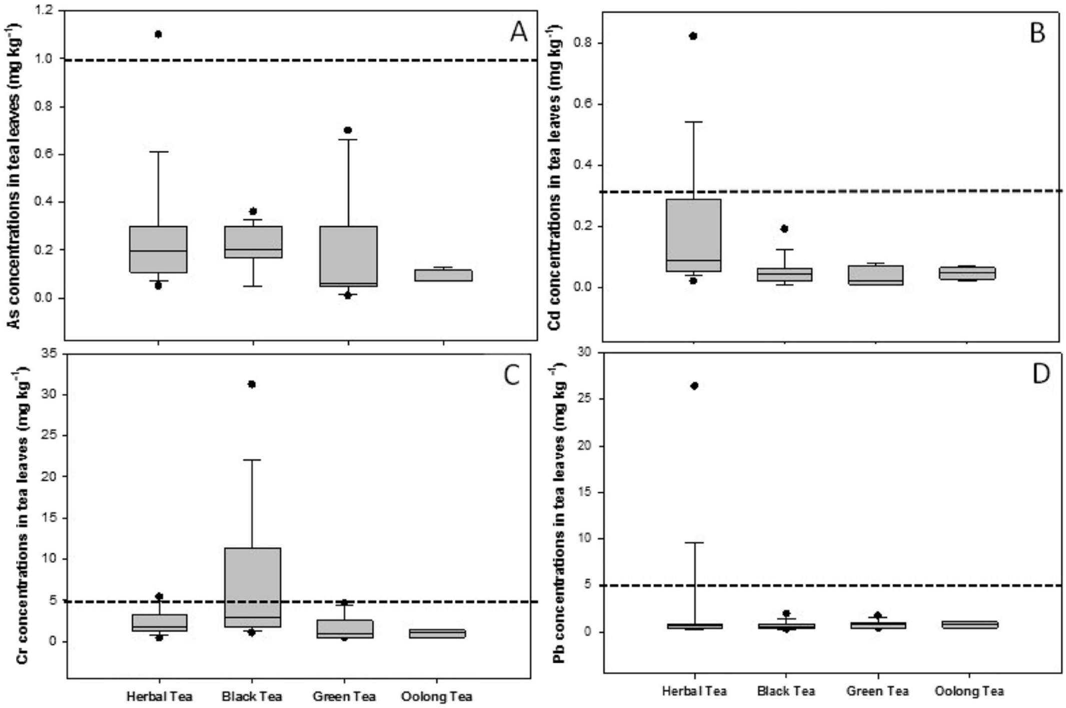

Fig. 1. Total As (A), Cd (B), Cr (C) and Pb (D) concentrations in the tea leaves from traditional and herbal teas. Dashed lines in AB are the limit in teas for As and Cd (1 and 0.3 mg kg^{-1}) in medicinal plants (WHO, 2007), and in CD are the limit in teas for Cr and Pb (5 mg kg^{-1}) based on Chinese standard (Table 1).

soluble Al at 14–49% (Table 2), similar to those reported for black tea from India and Java (43–50%) (Schulzki et al., 2017). All black tea and 83, 75 and 25% of green, oolong and herbal teas exceeded the USEPA's secondary MCL for Al in drinking water at 0.2 mg L^{-1}, with the Al concentration in tea infusion being 1.3–37 times higher than 0.2 mg L^{-1} (Table 2; Fig. 2B). These results suggest that drinking teas may contribute to Al intake, which are potentially harmful to those who drink large quantities of teas (Malik et al., 2013).

3.2. Weekly Al intake in children and adults

Exposure to high Al has been associated with Alzheimer's disease. Tea plants accumulate Al, making tea a major source of dietary Al intake. Therefore, it is necessary to evaluate the weekly Al intake with tea drinkers to assess the associated health risks. Considering a weekly tea consumption of 0.525 L by children <15 and 5.25 L by adults >35 years (Sofuoglu and Kavcar, 2008), we calculated the tolerable weekly Al intake from teas based on Eq. 1. They were 0.001–0.39 and 0.003–0.56 mg kg^{-1} for children and adults (Fig. 2CD). Higher weekly Al intakes were associated with black tea followed by green tea and oolong tea (Fig. 2CD).

Table 4
Regulatory limits for As, Cd, Cr and Pb in tea or herbal materials from different countries.

Countries		As	Cd	Cr	Pb	References
		mg kg^{-1}				
Brazil	Tea	0.6	0.4	–	–	ANVISA, 2013
Canada	Raw herbal materials	5.0	0.3	2.0	10	Street et al., 2006
China	Herbal materials	2.0	1.0	5.0	5.0	Karimi et al., 2008
German	Products of plant origin	–	0.2	–	5	Gasser et al., 2009
India	Tea	–	–	–	10	PFA, 1955
Japan	Tea	–	–	–	20	Karimi et al., 2008
Thailand	Herbal materials	4.0	0.3	–	10	Karimi et al., 2008
WHO	Dried plants material	1.0	0.3	–	10	WHO, 2007

The joint FAO/WHO Expert Committee on Food Additives (JECFA) established the provisional tolerable weekly intake (PTWI) for Al at 1.0 mg kg^{-1} (JECFA, 2011), which applies to all Al in foods. In our study, the weekly Al intake in black tea was 0.06–0.39 and 0.08–0.56 mg kg^{-1} in children and adults (Fig. 2C), which was lower than the PTWI at 1.0 mg kg^{-1}. Some studies showed that Al above the permissible level may develop Alzheimer's disease (USEPA, 2000).

3.3. Cr and Pb contents in tea leaves and infusions

Among the 4 metals including As, Cd, Cr, and Pb, Cr had the highest concentration in teas. The average Cr contents in oolong herbal, green, black, and tea samples were 0.97, 1.55, 2.32, and 7.37 mg kg^{-1} (Fig. 1C and Table 3). The Cr contents in black tea ranged from 1.03 to 31.2 mg kg^{-1}, which was similar to those from Turkey (10.0–14.8 mg kg^{-1}; Narin et al., 2004), but lower than those from China (17.9–115 mg kg^{-1}; Ferrara et al., 2001). The maximum Cr limit in teas is 5 mg kg^{-1} based on Chinese standard and 2 mg kg^{-1} based on Canadian regulations (Table 4). In this study, 47% black, 71% herbal, and 27% green teas exceeded 2 mg kg^{-1} (Fig. 1C and Table 3). The Cr in black tea may come from crushing and curling process during manufacturing (Polechońska et al., 2015). Also, black tea is produced from old leaves presumably containing higher Cr concentration (Szymczycha-Madeja et al., 2012). Chromium contents in the infusions of oolong, green, herbal, and black teas were 0.73–2.80, 0.83–3.20, 0.90–4.40, and 0.80–11.0 μg L^{-1} (Fig. 3C and Table 3), with 2.3–29% being soluble. This was lower than Li et al. (2013) who found 38% Cr was soluble from green tea, but similar to Karak and Bhagat (2010) who found 16% in Pu-erh tea.

The mean Pb concentrations in herbal teas was 2.37 mg kg^{-1}, which was higher than those of black, green, and oolong teas at 0.65–0.76 mg kg^{-1} (Fig. 1D and Table 3). Low Pb at 0.20 mg kg^{-1} was in herbal tea from Chile while high Pb at 26.4 mg kg^{-1} was in herbal tea from Spain. The Pb contents in herbal tea were higher than those from China at 2.90–8.40 mg kg^{-1} (Zhu et al., 2013). The Pb contents in

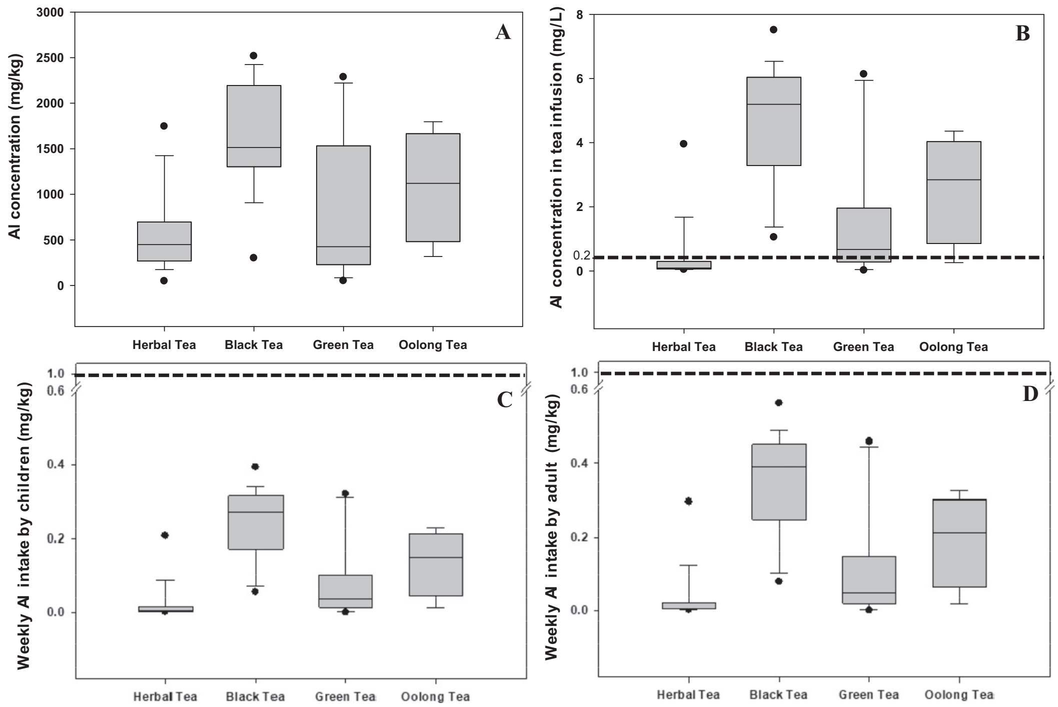

Fig. 2. Aluminum in tea samples (A), in tea infusion (B), weekly Al intake by children (C) and adults (D) from traditional and herbal teas. Dashed line in B is USEPA's secondary maximum contaminant level for Al in drinking water at 0.2 mg L^{-1} and CD is the provisional tolerable weekly intake for Al at 1.0 mg kg^{-1} (JECFA, 2011).

black tea were similar to those from Saudi Arabia at 0.30–2.20 mg kg^{-1} (Ashraf and Mian, 2008), but lower than those from Iran at 8.3–15.5 mg kg^{-1} (Zazouli et al., 2010).

The allowable Pb limit in tea leaves is 5 mg kg^{-1} in China and Europe, 10 mg kg^{-1} in Australia, Canada and India, and 20 mg kg^{-1} in Japan (Table 4). As such, Pb contents in all teas were below the limits excluding an herbal tea from Spain (Fig. 1D, Table 3). Excluding two teas, 0.21 to 19.9 μg L^{-1} Pb was soluble in infusion, corresponding to 1.9–80% total Pb (Fig. 3D, Table 3). These results are consistent with literature at 52% (Schulzki et al., 2017). The high Pb in infusion was from one herbal tea at 19.9 μg L^{-1}, which also contained high Pb in tea leaves at 26.4 mg kg^{-1}. During infusion, herbal and oolong teas released the highest Pb contents, averaging 38%.

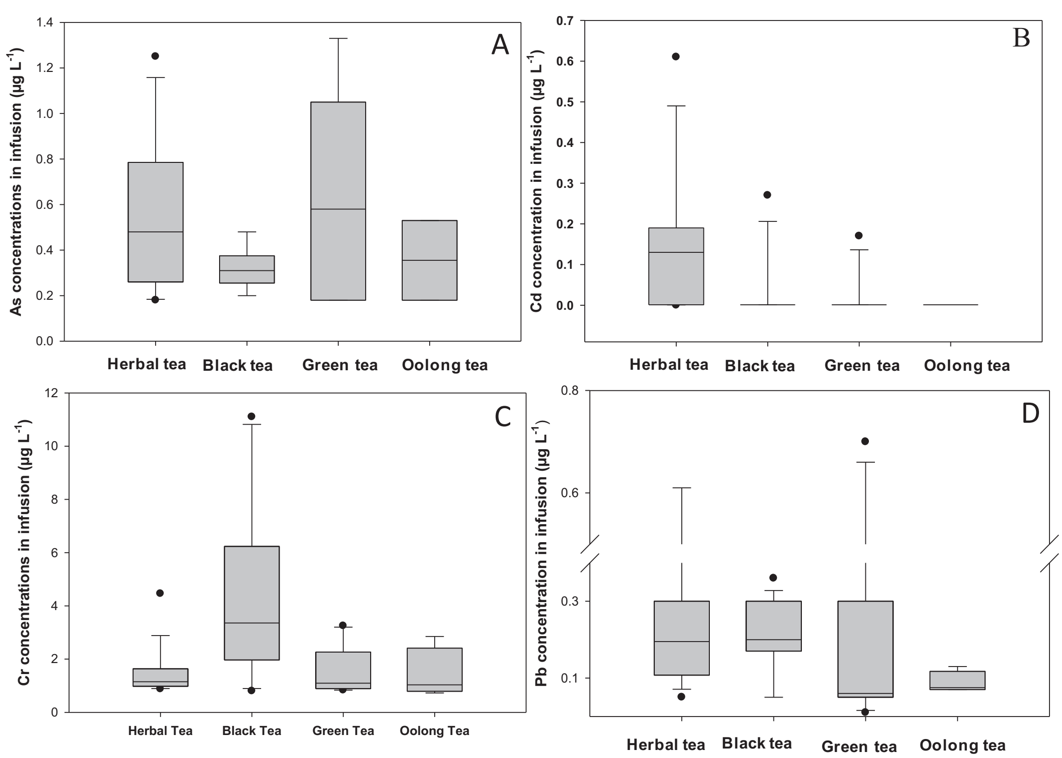

Fig. 3. Soluble As (A), Cd (B), Cr (C) and Pb (D) concentrations in tea infusion from traditional and herbal teas.

3.4. As and Cd contents in tea leaves and tea infusions

Chronic exposure to As in drinking water results in various adverse health effects including skin and internal cancers (USEPA, 2000). Mean As concentrations were generally low, ranging from 0.08 to 0.26 mg kg^{-1} (Fig. 1A and Table 3). In herbal teas, the sample from Peru had high As content at 1.10 mg kg^{-1}, exceeding the limit of 1 mg kg^{-1} for medicinal plants (WHO, 2007). The As concentrations in tea infusions was also low (Fig. 2A and Table 3), with little transfer of As from tea leaves to the infusion. Based on Szymczycha-Madeja et al. (2012), the highest metal solubilization occurs during the first 5-minute of contact between boiling water and tea leaves. Arsenic was detected above the detection limit of 0.18 μg L^{-1} in 54% tea infusions, ranging 0.19 to 1.33 μg L^{-1} (Fig. 2A and Table 3), but all below the As MCL in drinking water at 10 μg L^{-1}. Schwalfenberg et al. (2013) also found low As concentrations in tea infusions, ranging 0.06 to 1.12 μg L^{-1}. Szymczycha-Madeja et al. (2012) reported that the As contents in the infusion were also low. The authors did As speciation in the infusion, finding that up to 29–88% of As was in inorganic form.

The Cd contents in teas were 0.01–0.82 mg kg^{-1}, averaging 0.09 mg kg^{-1} (Fig. 1B and Table 3), higher than the mean of 0.02 mg kg^{-1} based on 798 teas by Han et al. (2005). The value was lower than 0.62–0.69 mg kg^{-1} from Zheng et al. (2014) and Seenivasan et al. (2008). Compared to traditional teas, herbal teas had higher Cd (0.19 vs. 0.01–0.08 mg kg^{-1}). In our study, 11% of teas exceeded the MCL of 0.3 mg kg^{-1} Cd in herbs (WHO, 2007). The average Cd content in 2307 tea samples from China was 0.1 mg kg^{-1}, with <1% of the samples having >0.5 mg kg^{-1} (Shi et al., 2008). The Cd in teas may result from phosphate and zinc fertilizers, a significant contributor to Cd contamination in soil (Cupit et al., 2002; Zhong et al., 2016). Cadmium contents in tea infusions were 0.13–0.61 μg L^{-1}, with 5–21% being soluble and being below the MCL in drinking water at 5 μg L^{-1} (Fig. 3B and Table 3). Sofuoglu and Kavcar (2008) also found low Cd content at 0.19 μg L^{-1} in tea infusions from Turkey.

4. Conclusion

In this study, we assessed the health risk associated with tea consumption based on 47 traditional and herbal teas originating from 17 countries. We found that As, Cd and Pb did not exceed the limit for medicinal plants recommended by WHO (2007) excluding one herbal tea for As (1.10 mg kg^{-1}) and Pb (26.4 mg kg^{-1}). High Cr levels in 47% herbal and 73% of traditional teas exceeded the Cr concentration allowed at 2 mg kg^{-1} (Table 4). Total Al was lower in herbal teas (47–1745 mg kg^{-1}) and its infusion (0.09–3.95 mg L^{-1}) compared to traditional teas (50.3–2517 mg kg^{-1}) and in infusions (0.02–7.51 mg L^{-1}). During tea infusion, 0.9–22% and 4–49% of the Al was released into water from herbal and traditional teas. All black tea and 83, 75 and 25% of green, oolong and herbal teas exceeded the secondary MCL for drinking water at 0.2 mg L^{-1}. Based on PTWI of 0.01–0.39 and 0.003–0.56 mg kg^{-1} for children and adults from drinking teas, tea consumption may contribution Al intake, especially those heavy drinkers and those like black tea. Based on our study, it is important to consider metal intake from tea consumption, especially for Al and Cr. More research is needed to expand the study to include more data.

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