


Assessment of the aluminium contents of black tea and black tea infusions

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Abstract

The aluminium (Al) can cause health problems to human being at overdose. The black tea is among the most common beverages in the world but may contain higher amount of Al. This study was conducted to determine Al content in black tea produced in Rize, Trabzon and Giresun/ Turkey and amount of Al transferred into tea infusions. Tea infusions were prepared for 15, 30, 45 and 60 min and the Al content was determined using the inductively coupled plasma optical emission spectroscopy (ICP-OES). The Al content in black teas was ranged from 8177.75 ± 1167.76 to $15,657.72 \pm 1060.05$ mg/kg. Al content of samples taken from Trabzon were lower than the Al contents of samples collected from Rize and Giresun. Infusion time effected significantly the amount of Al transferred into the tea infusions ($p < 0.05$). The Al content in black teas was significantly influenced by the production location and depended on the harvest season ($p > 0.05$).

Keywords Tea · Aluminium · Infusion · Heavy metal

1 Introduction

Tea is one of the most common drinks in all over the world and is produced from the leaves of many plants [1] and particularly from *Camellia sinensis*. Among all the beverages, about 98% of people prefer drinking the tea for the first choice. 75% (approx. 2.5 millions) of the desiccated tea produced annually are processed into black tea. According to Shekoochyan et al [2] about 18-20 billion cups of tea are consumed daily in the world. Turkey holds a significant place among the world's largest producer countries owing more than 4% of world production. According to the Food and Agriculture Organization of the United Nations [3] statistics, Turkey is the sixth major tea producer in world after China, India, Kenya, Vietnam and Sri Lanka. The production of tea in Turkey began in the early years of the Republic along the Eastern Black Sea Region and most of the tea plantations are centred in Rize, Turkey [4].

Teas have shown tremendous beneficial effects on human health, since they intervene in the prevention of Parkinson's disease, various skin problems, myocardial and coronary artery diseases [5]. Nonetheless, teas can contain some elements which are harmful to the human health [6]. The tea plants contain a higher concentration of aluminium (Al) than many other plants and would be a potentially important source of dietary Al [6]. Al variants in environmental and physical condition together with its physicochemical properties are often the determinants for its bio-availability and toxicity [7]. Al is the third most abundant element in the earth's crust and is therefore, a natural component of drinking water and foodstuffs. Excessive intake of Al can be harmful for human health, particularly for kidney. Kidney insufficiency results in an increase in Al concentrations in the kidneys of the dialysis patients [8]. Even though some epidemiological reports

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were contradictory, there was a mounting scientific evidence suggesting a relationship between the neurotoxicity of Al and the pathogenesis of Alzheimer's disease [6, 9].

Although Al content of tea has been determined in various teas from many countries [10–12], there is no detailed researches investigating Al content in the teas produced in different locations of Turkey. This study has two aims: (1) measure the concentrations of Al contained in the manufactured black tea leaves in Turkey; (2) to determine the rates and concentration of Al released after the preparation of infusions with boiling distilled water.

2 Materials and methods

2.1 Materials

Black tea leaves (*Camellia sinensis*) were collected three times during three harvest seasons (May, July and August) from three factories located in Giresun, Trabzon and Rize in Turkey.

2.2 Methods

2.2.1 Determination of dry matter

About 5 g of tea plant powder was put in tared nicel containers and dried at 105 °C up to constant weight. The dry matter was calculated by considering the loss of weight [13].

2.2.2 Preparation of tea infusions

For the preparation of tea infusions, 5 g black tea samples and 200 mL of hot deionized water (approximately 96 °C) were put in the cleaned glass teapot. After different brewing times (15, 30, 45 and 60 min), the infusions were filtered with Nylon to remove big particles and leaves. The so-called blank water followed the same procedure without tea leaves.

2.2.3 Preparation samples for ICP-OES analysis

The total Al was investigated for both tea infusions and tea plants. Briefly, 1 g of the black tea samples were digested with 16 mL HNO₃ (65% AR nitric acid) and 4 mL HClO₄ (70% perchloric acid) at 200 °C in a 100-mL volumetric flask [14].

ICP-OES standard of Al was purchase from Merck (Darmstadt, Germany). 1 mg.ml⁻¹ of stock solution was used and the standard solutions were prepared by diluting the stock solution in 5% (v/v) HNO₃ immediately before use. All chemicals and reagents employed were of analytical grade.

2.2.4 Instrumentation

ICP-OES (Perkin Elmer model Optima 2100 DV, Norwalk, CT, USA) was used for determination of Al in black tea leaves and their infusions. The ICP-OES operating parameters and conditions were given in Table 1.

Table 1 Operation Parameters and conditions for ICP-OES

Parameters	
RF power	1500 watts
Number of replication	3 times
Sample uptake rate	2 mL min ⁻¹
Flush time	15 sn
Delay time	10 sn
Wash time	15 sn

2.2.5 Modeling of infusion kinetics

The modeling kinetics was achieved by fitting the experimental data to Peleg (Eq. 1) and Weibull (Eq. 2) empirical rehydration models [15]. The data of the kinetics were statistically analyzed based on the values of coefficient of determination (R^2), chi-square (χ^2) and root mean square error (RMSE) using MATLAB software [16, 17]. The models with higher R^2 , as well as the lowest χ^2 and RMSE were selected as the best models to interpret the rehydration behaviors of dried fruit (Table 2).

$$R_r = \frac{1}{X_e - X_0} \times \frac{t}{(a + bt)} \quad (1)$$

$$R_r = 1 - \exp\left(-\left(\frac{t}{b}\right)^a\right) \quad (2)$$

where R_r is the ratio of aluminium infused at time t ; X_0 and X_e are the amounts of aluminium at time t_0 and t ; and a and b are the model constants.

2.2.6 Statistical analysis

The significant differences in Al contents of the tea leaves obtained from different harvest sessions and locations were determined with the two-way analysis of variance (ANOVA) method. The significant differences in Al contents

Table 2 Dry matter and aluminium contents of black tea samples

Locations	Plant	Plucking periods	Dry matter,%	Al, mg/kg ^a
Rize	1	1	94.29 ± 0.13	11,066.77 ± 672.67
		2	94.17 ± 0.05	11,548.51 ± 908.30
		3	94.41 ± 0.27	14,058.03 ± 817.09
	2	1	94.22 ± 0.16	13,847.27 ± 276.61
		2	93.69 ± 0.05	11,230.24 ± 1296.12
		3	94.10 ± 0.16	10,145.58 ± 1028.49
	3	1	93.85 ± 0.32	13,510.14 ± 293.62
		2	94.16 ± 0.27	8894.84 ± 637.81
		3	94.20 ± 0.06	10,509.89 ± 1237.66
Trabzon	1	1	92.83 ± 1.61	8780.01 ± 1735.33
		2	94.20 ± 0.11	10,266.88 ± 701.87
		3	93.56 ± 1.24	11,226.51 ± 970.78
	2	1	91.91 ± 0.50	8177.75 ± 1167.76
		2	94.01 ± 0.20	11,066.43 ± 406.94
		3	94.57 ± 0.11	10,792.31 ± 1310.49
	3	1	92.18 ± 0.05	11,102.63 ± 2072.67
		2	94.12 ± 0.06	10,709.45 ± 160.31
		3	94.57 ± 0.27	9896.10 ± 673.40
Giresun	1	1	91.96 ± 0.08	8809.94 ± 69.51
		2	94.10 ± 0.15	13,889.29 ± 1462.42
		3	95.23 ± 0.07	15,657.72 ± 1060.05
	2	1	92.59 ± 1.12	10,019.57 ± 1664.05
		2	94.10 ± 0.20	10,395.93 ± 469.73
		3	94.12 ± 1.63	10,080.76 ± 319.05
	3	1	92.71 ± 1.12	14,327.21 ± 628.14
		2	94.08 ± 0.18	9828.63 ± 336.61
		3	94.91 ± 0.48	9883.87 ± 410.88

^aIn dry basis

among the tea infusions as affected by infusion times were determined by using one-way ANOVA. Statistical differences among the groups were determined by Duncan multiple-range test.

3 Results and discussion

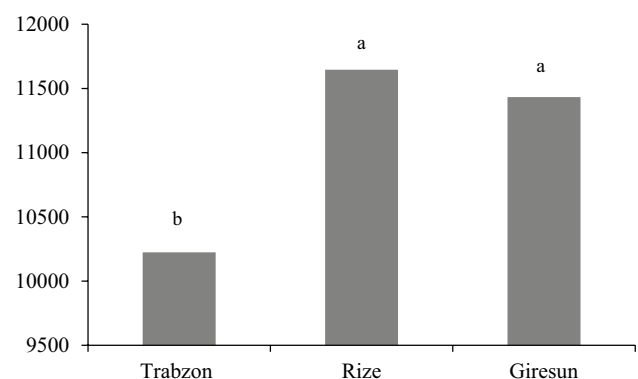
3.1 Aluminium content in black tea leaves

Al contents of black tea manufactured in different provinces during different shooting periods were given in Table 2. As can be seen, the mean of Al contents of black tea samples varied between 8177.75 and 15,657.72 mg/kg. The average Al contents of the tea were found between 8894 and 14,058.03 mg/kg in Rize, between 8177.75 and 11,226.51 mg/kg in Trabzon, and between 8809.94 and 15,657.72 mg/kg in Giresun. The differences among the average Al content of the districts are statistically significant ($p < 0.05$). There is no differences between Al contents of the leaves collected from Rize and Giresun, while samples from Trabzon showed significant differences when compared to other provinces (Fig. 1). Moreover, no significant difference ($p > 0.05$) was observed in tea leaves collected from different shooting periods. Erdemođlu et al. [18] reported the average Al content of black tea as 1054 $\mu\text{g/g}$ by using ion exchange resin and atomic absorption spectroscopy and Matsuura et al. [19] determined Al concentration of 807 $\mu\text{g/g}$ in black tea leaves using ICP-MS and ICP-AES.

Carr et al. [20] investigated total Al in young and old leaves of *C. sinensis* var. *sinensis* grown in acidic soil, and found as 380 and 6866 $\mu\text{g g}^{-1}$, respectively. Tang et al. [7] detected the Al contents of dry leaves and tea infusion using the spectrophotometer. They found the Al contents of the dry tea leaves were 325 $\mu\text{g/g}$, 483 $\mu\text{g/g}$ and 637 $\mu\text{g/g}$ for green tea, black tea and oolong tea, respectively. The concentrations of Al in the tea infusion were 4.9 $\mu\text{g/ml}$ for green tea, 6.7 $\mu\text{g/g}$ for black tea, and 9.5 $\mu\text{g/g}$ for oolong tea. Ansari et al. [10] determined the amounts of heavy metals such as Cd, Pb, Ni and Al and macro-elements Fe, Zn, Mn, and Cu using atomic absorption spectrometry in black tea cultivated in Iran and imported. They found that the mean level of Al was 699.2 mg/kg for Iranian and 388.3 mg/kg for imported black tea. Xie et al. [6] investigated the distribution of Al and F contents and the relationship between Al and F in tea plants and soils of 12 tea. They found the Al content ranged from 1196 to 7976 mg/kg for old leaf, 370 to 2681 mg/kg for young leaf and 285 to 525 mg/kg for a stem. Mossion et al. [21] determined total Al and calcium concentrations, total organic carbon and total polyphenol content, and compared with respect to the origin of tea leaves, their particle size (broken or whole leaves), and also the mineral composition of waters used to prepare infusions. They found that the higher the mineral content, the lower the extraction yields of aluminium, total organic carbon and total polyphenols were. Our findings were higher than the values Al reported in the previous studies.

The highest values of Al detected in black tea leaves produced in Turkey may be mainly associated to cultivation conditions and plant characteristics. Kröppel et al. [22] and Olivier et al. [23] have mentioned that the environmental factors such as soil features, climate, altitude, rainfall are the factors affecting the contents of metals in tea leaves are the environmental factors such as soil features, climate, altitude and rainfall. Olivier et al. [23] have precisely pointed out the fertilisation and genetic characteristics as factors influencing the Al content of black tea leaves. Moreover, it has been noted that older leaves accumulate more Al compared to fresh leaves [22, 24]. This assessment is in accordance to our results, since the tea leaves produced in Turkey are quite older. So, they accumulated more Al. The soil properties are other main factors affecting Al content of tea leaves. Wong et al. [25] have found that the tea leaves grown in acidic

Fig. 1 Aluminum content of black tea leaves from different provinces ($p < 0.05$)



and older soils contained higher amount of Al. The soils on which the tea plantations are installed in Turkey have acidic features. Özyazıcı et al. [26] analysed the soil features of the districts where tea plantations are dominant in Turkey. They determined ranges pH of 3.14–5.18 in Rize and 3.17–6.39 in Trabzon. Özkutlu et al. [27] reported pH varying between 3.49 and 5.01 in Rize, where 65% of tea plantations are installed in Turkey. They also affirmed that the harvest takes place when the pH of the soil is between 4.5 and 6.

3.2 Aluminium content in black tea infusions

The results of Al contents in black tea infusions were given in Table 3. The black tea leaves collected from Rize were used to prepare the infusions since they gave highest Al content (Fig. 1). The infusions' preparation lasted 45 min and the samples were taken in every 15 min for Al content analysis. As seen in Table 3, the amount of Al in tea infusion was ranged from 1068.56 to 1148.01 mg/kg. It was remarked that the duration of infusion had significant effects on the Al content ($p < 0.05$). The Al content increased with the increase of infusion time. Varo et al. [28] reported the Al concentration of 3 mg/kg in the tea samples infused for 5 min, while Coriat and Gillard [29] found more than 1000 mg/kg. Fairweather et al. [30] detected 3.9 mg/kg of Al after infusing 1 g green and black teas during 5 min in 100 ml boiled water. Baxter et al. [31] found 3.6 ppm of Al concentration in teas from India, Ceylon and China while Rao [32] determined 30–480 µg/L of Al content in commercial black tea powder bought from Southern India. It was remarked that the Al contents found in the present study were higher than those discovered in the explored previous studies. This may be linked to the higher amount of Al in the black tea leaves investigated. Moreover, for infusion samples; the average accumulated concentration of Al based on WHO guidelines was 0.05 µg/kg commonly). This indicates that Al content in dry tea and infusions was higher than the WHO guidelines. It can be concluded that the daily intake dose of Al through the consumption of dry tea and infusions will harm the health of consumers during their lifetime.

In present study, about 10% of Al was transferred in the tea infusion after 15 min. Similarly, Moghaddam et al. [33] have found out that 28.8%, 10.31% and 3.72% of Al were transferred at 2, 5 and 10 min, respectively. Erdemoğlu et al. [34] have determined 32.73% of Al transfer during the preparation of black tea infusions. Nookabkaew et al. [35] reported 21.79% of Al transfer while Han et al. [24] noticed 31.1% of Al transfer. Furthermore, the highest transfer of the amount of Al had occurred in the infusion prepared with ultra-pure water when compared to infusions prepared with the water containing lower, middle and higher mineral [21]. It can be assumed that the Al transfer detected in this study was significantly low when compared to data of the previous studies. Many factors infusion such as duration of infusion, composite of tea, density of water, type of teapot and infusion method affect the amount of Al transferred during the preparation of infusions. This study only focused on the effect of the duration of tea infusion and the results showed significant effects of infusion time on Al content. These findings are in agreement with previous studies [36, 37]. Even though there is an evidence in bioavailability of a small amount of Al, there is no question that Al is a potent neurotoxicant for both animals and human. It was reported that intracerebral inoculation of Al phosphate in rabbits resulted in neurofibrillary degeneration of "striking resemblance" to the neurofibrillary tangles of Alzheimer's disease [38, 39]. Rogers and Simon [40] have found out that the past consumption of foods containing large amounts of Al additives differed between people with Alzheimer's disease and controls, indicating that dietary intake of Al might promote the risk of developing this disease.

3.3 Results of kinetics and modeling

The release of aluminum during the different infusion times was steeper during the earlier period of infusion (0–40 min), slowed down after 40 min to reach almost constant rate from 55 min (Fig. 2). Similar behaviors have been previously observed during infusions of different kinds of teas, suggesting the time-dependency of the extraction components from teas [41–43]. The experimental data were applied to Peleg and Weibull empirical models and results were given in

Table 3 Total aluminium concentrations (mg/kg) in the black tea infusion

Time (min)	Mean ± std deviation
15	1068.56 ± 22.86b
30	1117.02 ± 16.10a
45	1140.48 ± 9.28a
60	1148.01 ± 25.49a

*The averages with same letters have no statistical differences ($p > 0.05$)

Fig. 2 Kinetics of the experimental data fitted to Peleg and Weibull models

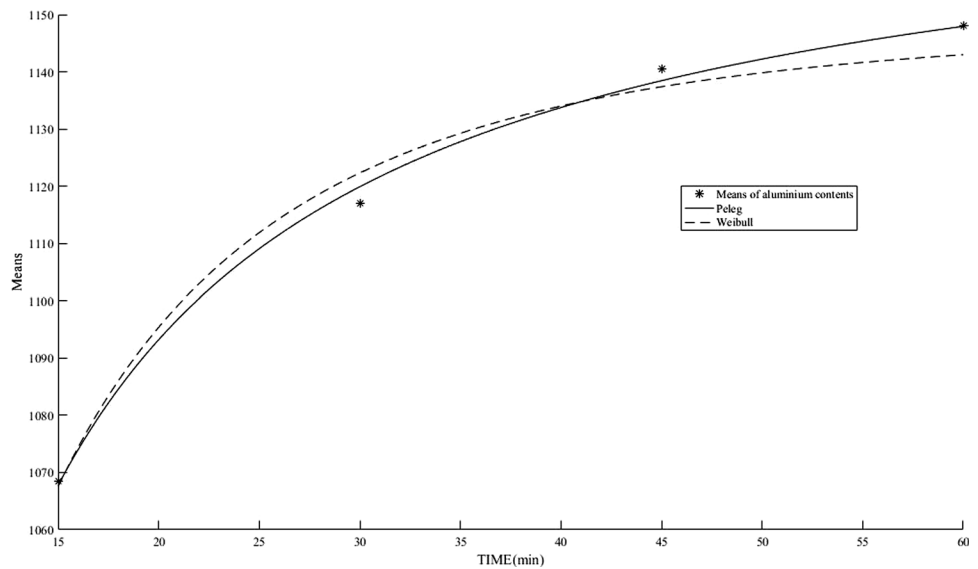


Table 4 Results of Peleg and Weibull models applied to the experimental data of the infused aluminium in function of time

NO:	Models	Equations	References	Results			
				R ²	RMSE	a	b
1	Peleg		(Benseddik et al., 2019; Vega-Gálvez et al., 2009)	0.9949	2.5633	0.0013	0.0008
2	Weibull		(Benseddik et al., 2019)	0.9751	5.6520	0.5158	2.2551

Table 4. Although the studies models represented better the kinetics of the release of aluminium during the infusions, it was remarked that the fitness of these models depended on the time. Both Peleg and Weibull described better the release behavior of aluminium, presenting high R² (0.9751–0.9949) and low RMSE (2.5633–5.6520) (Table 4). Our results are consistent with previous studies which have reported that these models are adequate to predict the release behavior of components from teas [41, 42]. The model of Peleg was revealed to be the best model giving the highest R² = 0.9949 and the lowest RMSE = 2.5633 (Table 4).

4 Conclusion

The Al contents of tea leaves and transferred during the preparation of infusions of teas from Giresun, Trabzon and Rize were analysed. The average Al contents of teas in Rize, Trabzon and Giresun were ordered between 8894 mg/kg and 14,058.03 mg/kg; 8177.75 mg/kg and 11,226.51 mg/kg; 8809.94 mg/kg and 15,657.72 mg/kg, respectively. Results of the analyses showed that the samples of Trabzon contain a statistically lower amount of aluminium comparing to the other cities ($p < 0.05$). Considering the harvest seasons, it was found out that the Al content is not statistically important ($p > 0.05$). When comparing to the previous studies, the results of the Al content detected in the present study were found higher. In this case, Turkish people who have high tea consumption habits may have health problems. This study showed that the Al content of tea increased with the increment of the duration of infusion ($p < 0.05$). In order to reduce the occurrence of Al content in the tea infusion, it is necessary to keep the infusion time short.

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Authors' contributions Conceptualization (ZO, OZ and IK); Methodology (ZO, OZ and IK); Validation (IK); Formal Analysis (ZO and OZ); Data Curation (IK); Writing—Original Draft Preparation (ZO and OZ); Writing—Review & Editing (IK); Supervision (IK). All authors read and approved the final manuscript.

Data availability The dataset generated for this study are available on request to the corresponding author.

Declarations

Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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