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Multi-element analysis of tea beverages by inductively coupled plasma atomic emission spectrometry

P.L. Fernández, F. Pablos, M.J. Martín*, A.G. González

Department of Analytical Chemistry, Faculty of Chemistry, University of Seville 41012-Seville, Spain

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Abstract

The metal content of several tea beverages, infusions, instant and tea soft drinks was determined. Eleven metals Al, Ba, Ca, Cu, Fe, K, Mg, Mn, Na, Sr and Zn were determined by inductively coupled plasma atomic emission spectrometry (ICP–AES). A study of the differences in the metal contents of tea beverages has been carried out. Pattern recognition techniques such as principal components analysis and linear discriminant analysis were used to discriminate between the tea beverages. The metal intake related to tea consumption has also been studied. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Tea; Metals; Pattern recognition; Chemometrics

1. Introduction

Tea is one of the most popular beverages in the world. Health benefits of tea have been documented since the early Chinese and Japanese texts and these are being seriously considered by a number of researchers in the scientific community (Hara, Luo, Wickremasinghe, & Yamanishi, 1995; Stagg & Millin, 1975). There are, therefore, a number of reports of studies investigating the benefit of tea in reducing the incidence of cancer (Shi et al., 1994; Wang et al., 1994), vascular disorders (Hertog, Feskens, Hollman, Katan, & Kromhout, 1993), hypertension (Henry & Stephens-Larson, 1984) and dental caries (Rasheed & Haider, 1998). The average tea drink, or infusion, is prepared by seeping the dried leaf in near-boiling water, and contains little protein, vitamins, fibre or carbohydrate, but may be a source of some essential dietary metals and metal binding polyphenols (Powell, Burden, & Thompson, 1998). The regular consumption of tea may contribute to the daily dietary requirements of several elements (Xie, Bohlen, Klockenkämper, Jian, & Günther, 1998). For instance, tea could be an important source of manganese and the large amount of potassium in comparison with sodium that could be beneficial for hypertensive patients. Due to the importance of the minerals present in tea, several studies have been carried out to determine their levels in leaves and tea infusions by using inductively coupled plasma atomic emission spectrometry (ICP–AES; Marcos, Fisher, Rea, & Hill, 1998; Natesan & Ranganathan, 1990), neutron activation analysis (Ma, et al., 1993) and total reflection X-ray fluorescence (Xie et al., 1998). In the present paper the most relevant metals (Al, Ba, Ca, Cu, Fe, K, Mg, Mn, Na, Sr and Zn) occurring in several tea beverages (infusions, instant teas and soft tea drinks) were determined by ICP–AES). The metal content was used to discriminate them by applying Pattern Recognition tools. These differences were emphasised in order to evaluate the importance of tea beverages as dietary source of minerals.

2. Experimental

2.1. Analysis of the metal content in tea beverages

The metal content was analysed in three different kinds of tea beverages, i.e. infusions, instant teas and

* Corresponding author. Tel.: +34-954-557173; fax: +34-954-557168.

E-mail address: mmartin@cica.es (M.J. Martín).

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soft drinks. Tea infusions were prepared using green and black tea leaves from different geographic origins. Those tea leaves as well as instant teas and the soft drinks were obtained in the market. An identification code was assigned to each sample consisting of a number followed by a letter (G for infusions made of green tea leaves, B for infusions made of black tea leaves, I for instant teas and S for soft drinks). Table 1 presents a short description of the samples.

Tea infusions were made with distilled water following the usual way of preparing these kinds of beverages. Nevertheless, in order to unify and make comparisons all the infusions were prepared with the same amount of tea and water. Accordingly, 1.50 g of tea leaves (exactly weighed) were placed into a 250 ml beaker and then 100 ml of boiling water were added. After an adequate extraction time, the infusion was filtered through a folded filterpaper. Once cooled to room temperature, the infusion was transferred into a 100 ml volumetric flask, adding milli-Q water till the mark. In the case of instant teas the recommendations of the manufacturer were followed. Samples 1I and 2I are 100% tea extract and they were prepared weighing 0.5 g with a final volume of 100 ml. For samples 3I, 4I and 5I, 10 g were taken because they have a lower content of tea extract. The metal content of the tea soft drinks was directly measured.

Analyses were carried out by ICP–AES (Fisons-ARL 3410, FISONS Instruments, Valencia, California, USA). The instrumental conditions, analytical lines and linear range for each element are given in Table 2. The wavelength of the analytical lines was selected in most of the cases considering the most prominent line and, alternatively, secondary lines were used to prevent possible interferences. Due to the high stability of the ICP plasma, an internal standard was not necessary and instrumental drift was not detected during the analysis. The precision of the measurements was less than 2% in all cases. All analysis were performed in duplicate and after checking by standard addition, matrix effect was not detected. Standards were prepared by dilution of a multi-element standard (1000 mg l^{-1}) obtained from Merck (Darmstadt, Germany). All aqueous solutions and dilutions were prepared with ultrapure water (Milli-Q, Millipore, Bedford, MA, USA).

2.2. Data analysis

The 11 metals determined were considered as chemical descriptors of each sample. Chemometric calculations were made using the obtained data. Several pattern recognition (PR) techniques were applied to the data set including display methods such as principal component analysis PCA; Chatfield & Collins, 1981) and cluster analysis (CA; Massart & Kaufman, 1983) and supervised learning procedures for classification such as linear discriminant analysis (LDA; Coomans, Massart, & Kaufman, 1979). The statistical package STATISTICA 99 from Statsoft (Statsoft^{TM}) was used for PR computations.

Table 1
Analysed tea beverages

Sample code	Class	Comments
1G	Green	
2G	Green	
3G	Green	
4G	Green (Mint)	
5G	Green	
6G	Green	
7G	Green	
8G	Green (Jasmine)	
9G	Green (Gunpowder)	
10G	Green	
1B	Black (Darjeeling)	
11G	Green (Earl Grey)	
12G	Green (Chum Mee)	
13G	Green (Sencha)	
14G	Green	
15G	Green (Gunpowder)	
16G	Green (Genmaicha)	
17G	Green	
18G	Green	
19G	Green (Gunpowder)	
20G	Green (Jasmine)	
2B	Black (Earl Grey)	
3B	Black	
4B	Black (English Breakfast)	
5B	Black (Teekane)	
6B	Black (Ceylon)	
7B	Black (Assam)	
8B	Black (Ceylon)	
9B	Black	
10B	Black	
11B	Black	
12B	Black (Earl Grey)	
13B	Black	
14B	Black (Lemon)	
15B	Black	
16B	Black	
17B	Black	
18B	Black	
19B	Black	
20B	Black (Ceylon)	
21B	Black (Ceylon)	
22B	Black (Assam)	
23B	Black	
1I	Instant	100% tea extract
2I	Instant	100% tea extract
3I	Instant apple tea	1.4% tea extract
4I	Instant lemon tea	1.8% tea extract
5I	Instant peach tea	–
1S	Lemon Nestea	Soft drink
2S	Diet Lemon Nestea	Soft drink
3S	Peach Lipton Ice tea	Soft drink
4S	Lemon Lipton Ice tea	Soft drink
5S	Lemon Trina tea	Soft drink
6S	Diet Lemon Trina tea	Soft drink
7S	Radical Lemon Ice tea	Soft drink
8S	Diet Lemon Ice tea	Soft drink

3. Results and discussion

3.1. Metal content in tea beverages

The Chinese standard procedure for judging the flavour of tea (Xie et al., 1998) recommends an extraction time of 5 min. Nevertheless for some of the manufacturers 3 min of infusion is adequate to obtain a good quality tea beverage. In order to check possible differences in the metal content of tea infusions prepared in these two ways, two sets of analysis of teas with 3 and 5 min of infusion were run. In addition, tea beverages prepared with instant teas and tea soft drinks were also analysed. Table 3 summarises the obtained results, including ranges, mean values and the corresponding standard deviations. Sample 4G, that is a mint flavoured tea, presented very high values for all the analysed metals except for manganese and, consequently, it was considered an outlier and was not used for further calculations. At first glance it can be indicated that the results for infusions present a wide variation. It could be due to the natural variation of the metal content in the tea leaves and also to the random errors present in the experimental process (preparation of the infusions and measurements). For the majority of the elements, the obtained concentrations are similar to those reported by Powell et al. (1998) and Xie et al. (1998). Potassium was the metal with higher contents in all infusions with values ranging from 94 to 364 mg l^{-1}. The contents of magnesium and calcium ranged, approximately, between 5 and 22 mg l^{-1}. Manganese and aluminium appear with lower concentrations that were included in the intervals 1–6 mg l^{-1} for Mn and 1–10 mg l^{-1} for Al. The content of the other analysed metals was lower than 1 mg l^{-1}.

In the case of instant teas, Zn, Mn, Fe, Mg and Cu were present in higher concentrations for the beverages prepared with samples 1I and 2I. For the rest of the metals, there were no apparent differences except for sodium with a major content in samples 3I, 4I and 5I. By comparing instant teas with infusions it could be stated that samples 1I and 2I present similar concentrations to those in the infusions except for Mn and Mg that are present at higher levels and calcium that was present at a lower concentration in all instant teas.

Table 2
Operating parameters for inductively coupled plasma atomic emission spectrometry (ICP–AES)

RF frequency	27.12 MHz
Operating power	650 w
Coolant argon flow rate	7.5 l min^{-1}
Plasma argon flow rate	0.8 l min^{-1}
Burner type	Minitorch
Nebuliser type	Meinhard
Sample flow rate	2.3 ml min^{-1}

Element	λ (nm)	Linear range (µg ml^{-1})
Al	396.152	2.4–98.4
Ba	455.403	0.2–1.0
Ca	393.366	2.4–98.3
Cu	324.754	0.2–1.0
Fe	259.940	0.2–9.8
K	766.490	50–960
Mg	279.553	2.4–98.6
Mn	257.610	2.4–97.5
Na	589.592	2.4–9.8
Sr	407.771	0.2–1.0
Zn	213.856	0.2–1.0

Table 3
Metal content (mg l^{-1}) of the analysed tea beverages

Type of beverage	Zn	Mn	Fe	Mg	Cu	Al	Sr	Ca	Ba	Na	K
G3 mean[a]	0.17(5)	2.1(5)	0.08(3)	9(2)	0.07(2)	3(1)	0.06(9)	7.0(8)	0.014(6)	0.8(4)	190(35)
G3 range	0.27–0.08	3.5–1.3	0.16–0.02	14–6	0.12–0.04	6–2	0.08–0.04	9.1–5.7	0.029–0.008	2.4–0.4	259–115
G5 mean[a]	0.19(6)	2.6(5)	0.08(3)	10(2)	0.06(3)	3(1)	0.05(1)	7.71(9)	0.02(2)	0.9(6)	170(35)
G5 range	0.31–0.08	3.8–1.5	0.17–0.03	14–5	0.12–0.01	7–2	0.08–0.04	9.88–5.87	0.07–0.01	3.3–0.5	227–94
B3 mean[a]	0.29(6)	3(1)	0.07(5)	12(2)	0.09(3)	4(2)	0.08(2)	9(1)	0.03(2)	1(1)	250(20)
B3 range	0.50–0.19	6–1	0.23–0.03	17–8	0.16–0.03	9–1	0.16–0.05	16–6	0.12–0.01	5–0	280–205
B5 mean[a]	0.20(5)	3(1)	0.06(3)	15(3)	0.01(3)	6(2)	0.08(2)	10(2)	0.03(3)	1(1)	290(32)
B5 range	0.29–0.12	6–1	0.16–0.03	22–9	0.15–0.04	10–1	0.15–0.04	18–6	0.13–0.01	6–1	364–179
I[b] mean[a]	0.230(2)	6.8(7)	0.09(4)	20.2(2)	0.05(3)	3.48(2)	0.017(4)	1.7(5)	0.014(8)	0.3(2)	255(6)
I[b] range	0.233–0.226	7.3–6.3	0.13–0.06	20.4–20.0	0.07–0.03	3.49–3.47	0.020–0.014	2.1–1.4	0.019–0.008	0.4–0.2	260–251
I[c] mean[a]	0.051(8)	3.1(8)	0.056(7)	3(1)	0.012(2)	3.7(7)	0.011(5)	2.4(5)	0.013(4)	1.4(2)	213(15)
I[c] range	0.060–0.044	4.0–2.5	0.063–0.050	5–3	0.013–0.010	4.3–2.9	0.011–0.010	2.6–1.8	0.016–0.009	1.6–1.2	230–204
S mean[a]	0.04(2)	0.7(5)	0.1(1)	14(14)	0.005(2)	1.1(8)	0.2(3)	17(20)	0.021(6)	146(95)	78(51)
S range	0.08–0.02	1.1–0.0	0.3–0	50–5	0.008–0.003	2.5–0.2	0.7–0	57–1	0.031–0.011	330–26	176–10

G: green teas; B: black teas; 3 or 5: minutes of infusion; S: tea soft drinks.
[a] Average of triplicate determinations. Values between parentheses refer to the error corresponding to the last significant figure.
[b] Instant tea (100% tea extract, 1I, 2I).
[c] Instant tea (<2% tea extract, 3I, 4I, 5I).

The metal contents in the analysed tea soft drinks presented a wide variability. Between the studied metals sodium was at a higher concentration with values included in the range 330–26 mg l^{-1}. These values are higher than those encountered in the infusions and instant teas. This could be due to the additives added as sodium salts present in this type of drink. The potassium contents vary between 176 and 10 mg l^{-1}, which in this case are lower than those found in the other studied tea beverages. Calcium and magnesium present contents ranging between 57 and 1 mg l^{-1} and 50 and 5 mg l^{-1}, respectively. Manganese appears with lower concentrations comparing with the other tea beverages.

It could be very useful to make comparisons between the different types of infusions, i.e. G3 vs. G5, B3 vs. B5, G3 vs. B3 and G5 vs. B5. In order to find significant differences between the infusions prepared with green or black teas and 3 or 5 min of extraction, a Student t-test was performed. According to the null hypothesis and for a confidence level $P=0.05$ several significant differences were found. The obtained results are gathered in Table 4. It appears that for green teas Zn, Mn and Ca are significantly higher in 5 min infusions while for black ones the concentrations of Mg, Al, Ca and K are significantly higher in 5 min infusions while Zn and Cu concentrations are lower. After making the comparison between black and green teas at 3 min Mn, Mg, Cu, Al, Ca, Ba and K are significantly higher in black teas whereas Zn concentration is higher in green teas. Similarly, for black teas vs. green teas at 5 min, Mg, Al, Sr, Ca and K are significantly higher in black ones and Fe and Cu in green teas.

3.2. *Differences in the metal content of tea beverages*

In order to get a better understanding on the possible differences between the metal content of the considered tea beverages several chemometric methods were applied. The metal contents of infusions made with green and black teas prepared with 3 or 5 min of extractions, instant teas and soft drinks were considered as data matrix. This study was carried out trying to find differences between all types of tea beverages. First of all, PCA was performed in order to visualise the data trends. After calculating the two first principal components (PC 1 and PC 2) the scores plot of the samples was depicted and it is shown in Fig. 1. Only the separation between the soft drinks and the rest of samples is appreciated. To obtain classification rules for assigning categories to all types of tea beverages, a supervised PR method like LDA was applied. In this case, four classes were considered, i.e. infusions, soft drinks and two classes of instant teas. After performing LDA, three discriminant functions were calculated. Fig. 2 shows the scatter plot of the tea beverages using as axes the two first ones. It can be observed that soft drinks are situated at the right side of the plot well separated from the rest of beverages. Infusions are grouped at the left side of the figure being the two classes of instant teas separated, but close to them. This can be explained because

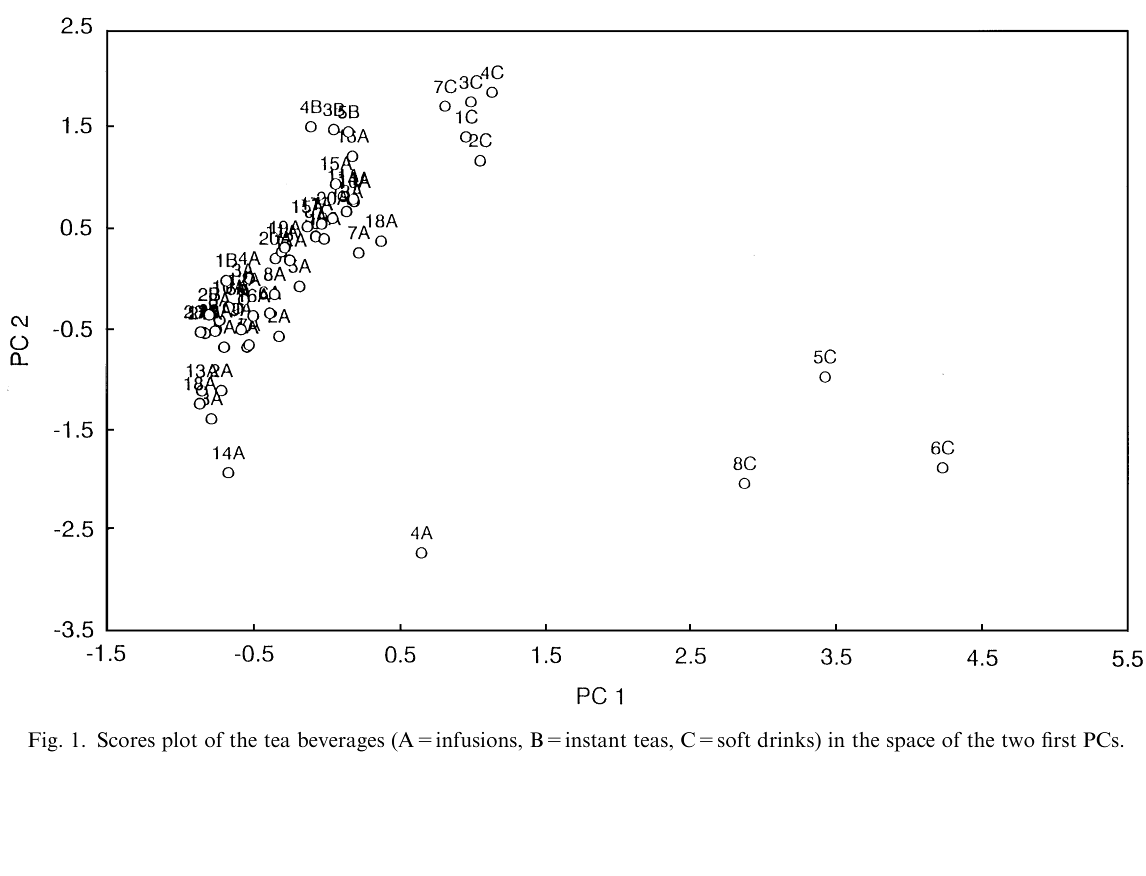

Fig. 1. Scores plot of the tea beverages (A = infusions, B = instant teas, C = soft drinks) in the space of the two first PCs.

the preparation of instant teas is in such a way that the resulting beverage is similar to an infusion made with tea leaves. Looking at the a posteriori probabilities, all samples were correctly classified except samples 5I and 7S, which represent a recognition ability of 94.6%. In order to evaluate the classification performance the leave-one-out method (Su, Danzer, & Thiel, 1997) was used as a validation procedure. A prediction ability of 91.1% was obtained.

3.3. Metal intake from tea consumption

As indicated earlier, tea beverages act as suitable sources of metal intake for humans (Xie et al., 1998). In order to discuss the contributions of the different tea beverages on the average daily dietary intake (ADDIs) for the studied metals (Mahan & Scott-Stump, 1996; Powell et al., 1998), a first calculation gave the average metal content by cup (μg/50 ml beverage). The results are gathered in Table 5, by indicating between brackets their contribution to ADDIs as%. It is remarkable the high contribution to ADDIs in the case of manganese. For potassium, tea beverages provide an important amount of this metal though its percentage of contribution to ADDIs is low because of the high potassium requirements of the human diet. To make useful comparisons, these calculations were carried out considering a volume of 50 ml per cup, though it is very usual to drink higher volumes of tea beverages, i.e. 120–250 ml of tea infusions and 330 ml of tea soft drinks. Consequently, the average metal content and the contribution to ADDIs by cup would be higher in a factor up to five for tea infusions or more than six in the case of the soft drinks. On the other hand, by considering the average consumption per person per day of tea beverages, the amount of metal intake per person and day

Table 4
Student *t*-test results to compare the metal content between tea infusions

Tea infusions	Zn	Mn	Fe	Mg	Cu	Al	Sr	Ca	Ba	Na	K
G3 *vs* G5	G5 +	G5 +						G5 +			
B3 *vs* B5	B3 +			B5 +	B3 +	B5 +		B5 +			B5 +
G3 *vs* B3	G3 +	B3 +		B3 +	B3 +	B3 +		B3 +	B3 +		B3 +
G5 *vs* B5			G5 +	B5 +	G5 +	B5 +	B5 +	B5 +			B5 +

Sign + means significantly higher concentration according to the Student *t*-test ($t_{calculated} > t_{tabulated}$). $P = 0.05$.

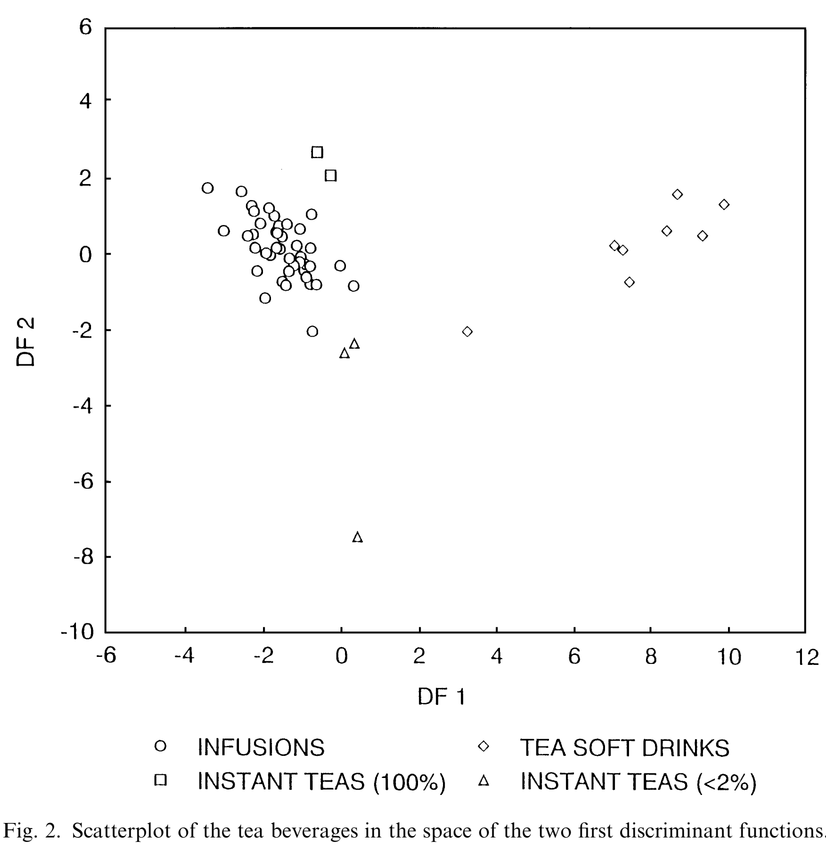

Fig. 2. Scatterplot of the tea beverages in the space of the two first discriminant functions.

Table 5
ADDI[a] for each metal and average metal content by cup of tea beverage

Metals	ADDIs[a] (mg day^{-1})	Infusions[b] (μg cup^{-1})	Soft drinks[b] (μg cup^{-1})	Instant teas[b] (μg cup^{-1})
Zn	15	10.6 (0.07)	2.0 (0.01)	7.0 (0.05)
Mn	4 (2–5)	133.7 (3.34)	35.0 (0.88)	247.5 (6.19)
Fe	15 (10–18)	3.6(0.02)	5.0 (0.03)	3.7 (0.02)
Mg	350 (300–400)	575.0 (0.16)	700.0 (0.20)	580.0 (0.17)
Cu	2.5 (2–3)	2.9 (0.12)	0.3 (0.01)	1.6 (0.06)
Al	5 (2–10)	200.0 (4.00)	55.0 (1.10)	179.5 (3.59)
Sr	1.6 (0.98–2.2)	3.3(0.21)	10.0 (0.62)	0.7 (0.04)
Ca	1000 (800–1200)	421.4 (0.04)	850.0 (0.09)	102.5 (0.01)
Ba	1.1 (0.65–1.7)	1.2 (0.11)	1.1 (0.10)	0.7 (0.06)
Na	2200 (1100–3300)	46.2 (0.002)	7300.0 (0.33)	42.5 (0.002)
K	3800 (1900–5600)	11250.0 (0.30)	3900.0 (0.10)	11700.0 (0.31)

[a] ADDIs, average dietary daily intake; mean value and range in parenthesis. (Mahan & Scott-Stump, 1996; Powell et al., 1998).
[b] In parenthesis, % of average daily dietary intake from a cup (50 ml) of tea beverage.

Table 6
Average amount of every metal per person and day (μg person^{-1} day^{-1})

Metals	Infusions[a]	Soft drinks[b]	Instant teas[a]
Zn	31.33	0.66	20.65
Mn	394.56	11.51	730.13
Fe	10.62	1.64	10.92
Mg	1696.25	230.14	1711.00
Cu	8.56	0.10	4.72
Al	590.00	18.08	529.53
Sr	9.74	3.29	2.07
Ca	1243.13	279.45	302.38
Ba	3.54	0.36	2.07
Na	136.29	2400.00	125.38
K	33187.50	1282.19	34515.00

[a] Considering an average consumption of 2.95 cups per person per day; beverage content from table 5 multiplied by 2.95 (http://www.teaparty.co.uk/tp/content/facts.htm).
[b] Considering an average consumption of 6 l per person per year; beverage content from Table 5 multiplied by $\frac{6000\ \text{ml}}{50\ \text{ml} \bullet 365\text{days}}$ (http://www.boiss-sons.ch/n/new1999a.htm).

was estimated and depicted in Table 6. As can be seen and due to their higher consumption infusions, prepared with tea leaves or instant teas constitute a better source of metals. The contribution of tea soft drinks is lower except for sodium.

4. Conclusion

Considering the chemometric calculations, PCA and specially LDA it can be concluded that the studied metal profile (Zn, Mn, Fe, Mg, Cu, Al, Sr, Ca, Ba, Na and K) is a very good descriptor for the differentiation of the common types of tea beverages (infusions, instant teas and soft drinks). Tea infusions are a good source of dietary minerals, especially black teas which are considered the better source of manganese and potassium. On the other hand, from statistical comparisons, it can be concluded that infusions prepared with black teas present higher levels of Mn, Mg, Al, Ca, K. Furthermore there are differences between infusions prepared with 3 or 5 min of extraction. Zn, Mn and Ca present higher concentrations for green teas and Mg, Al, Ca and K for black teas prepared with the longer time of extraction.

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