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Elemental analysis of teas, herbs and their infusions by means of total reflection X-ray fluorescence

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ABSTRACT

This work shows that total reflection X-ray fluorescence (TXRF) is a suitable tool for multi-elemental analysis of teas, herbs and their infusion. A low power benchtop TXRF spectrometer was used. Safety of infusion consumption was evaluated. Many commercially available teas,herbs and roots samples were analyzed. Total concentrations of thirteen elements K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Rb, Sr, Ba and Pb as well as their extraction efficiencies into infusion were determined. The content of Pb is highlighted in all teas and herbs, in the concentration range between 0.5 and 4.8 mg/g, but only in infusions of herbs. Chemometry was applied for classification purposes. Elemental daily intakes with respect to infusion drinking was calculated and compared with dietary reference intake values. Results show that TXRF is a fast and simple technique for safety check of tea and herb infusions on a routine basis.

1. Introduction

Tea and herb infusions are, one of the most popular non-alcoholic beverages world-wide, due to aroma, taste, smell, variety of types. Multiple positive health-promoting effects are reported in literature (Heiss and Heiss, 2007; McKenzie et al., 2010; Pohl et al., 2016; Salahinejad and Aflaki, 2010; Welna et al., 2013). Tea is an evergreen plant belonging to the family of *Camellia sinensis*. The best-known varieties of tea are: black, green, oolong, pu-erh, white and yellow. Among them, black, green, and oolong are the most popular and they differ only for the fermentation degree of leaves (Desideri et al., 2011a; Szymczycha-Madeja et al., 2012). Instead, herbal infusions are prepared from different parts or a mixture of medicinal plants, *i.e.* flower, leaves, fruits, herbs, barks and roots (Pohl et al., 2016).

Tea and herbs have a complex chemical composition. Beside organic compounds, such as flavonoids, amino-acids, proteins, enzymes, vitamins, aroma-forming substances, volatile oils and carbohydrates, they have a high content of major, minor and trace elements (Desideri et al., 2011b; Pohl et al., 2016; Soylak et al., 2007; Szymczycha-Madeja et al., 2012; Welna et al., 2013). Elemental composition is characteristic of the type of tea or herb and it is principally attributed to the production and the geographical origin of plants, i.e. soil composition, climate, local environmental conditions and agricultural practices (Aksuner et al., 2012; Chen et al., 2009; Desideri et al., 2011a; McKenzie et al., 2010; Pohl et al., 2016; Soylak et al., 2007; Welna et al., 2013).

Daily consumption of tea and herb infusions significantly contributes to the dietary intake of elements essential for human health like Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Se, V and Zn, (Aksuner et al., 2012; Desideri et al., 2011a; Pohl et al., 2016; Salahinejad and Aflaki, 2010; Xie et al., 1998). Beneficial effects are associated with anti-oxidant activity (Łuczaj and Skrzydlewska, 2005; Pohl et al., 2016; Zhu et al., 2002), protective effects against cancer (Pohl et al., 2016; Siddiqui et al., 2005; Way et al., 2004), vascular diseases, hypertension, dental caries (Sofuoglu and Kavcar, 2008) and reduction of blood cholesterol levels (Fujita and Yamagami, 2008). However, adverse effects on human health have been sometimes observed. Indeed, C*amellia sinensis* is known to tolerate and accumulate large quantities of F and Al associated with dental and skeletal fluorosis (Sofuoglu and Kavcar, 2008; Whyte et al., 2008) and Alzheimer's disease (McLachlan, 1995; Shu et al., 2003), respectively, even if tea will always be a minor source. Other potentially toxic elements, such as Pb, As, Cd and Hg, originated by the growth environment pollution, such as atmospheric dusts, rainfalls, fertilizers used, or during the tea manufacturing processes, can also be accumulated (Aksuner et al., 2012; Desideri et al., 2011a; Welna et al., 2013). It should be also highlighted that the elements bioavailability plays an important role. Therefore, the knowledge of essential and toxic elements concentration, is of major importance for consumers and producers (Han et al., 2007; Salahinejad and Aflaki, 2010), leading to the definition of tea quality.

Different tea and herb products are on the market. For obvious

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Table 1
Description of the studied teas and herbs samples.

Sample type	Sample name	Commercial name	Brand or Importer	Country of Origin
Black tea	B1	Uva Bombagalla Pekoe	PETER'S Tea House Trading	Sri Lanka
	B2	El Arosa Tea	Elyouser for Food Industries	Kenya
	B3	Broken Orange Pekoe	Two Lambs and Shield	Sri Lanka
	B4	PG Tips	Unilever UK	Kenya
	B5	Vanilla Tea	Twinings of London	China
	B6	The N25 Noir	Palais Des Thés	Sri Lanka
	B7	Earl Grey	Twinings of London	Sri Lanka
	B8	Four Red Fruits	Twinings of London	China
	B9	The au Tiare	Manutea	Sri Lanka
	B10	Prince of Wales	Twinings of London	China
	B11	Earl Grey	Twinings of London	Sri Lanka
	B12	English Breakfast	Dulcimea	Sri Lanka
	B13	Yunnan Black tea	–	China
	B14	Thé Infrè Decaffeinated	Bonomelli	Unknown
	B15	Star Tea Berries	Star	Unknown
	B16	Misaki Peach Tea	Dulcimea	Unknown
	B17	Misaki Lemon Tea	Dulcimea	Unknown
	B18	Golden Assam	Twinings of London	India
	B19	Earl Grey Classic	Twinings of London	China
Green tea	G1	Green Tea	Primia	China
	G2	Thé vert supérieur 555	Palais Imperial	China
	G3	Temple of Heaven China Green Tea	Shanghai Tiantan International Trading Co., LTD	China
	G4	Earl Grey Green Tea	Everton	India
	G5	Pure Green Tea	Lipton	Kenya
	G6	Green Tea with Mint	Aboca	Unknown
	G7	The N25 Green	Palais Des Thés	China
	G8	Pure Green Tea	Twinings of London	China
	G9	Jasmine Green Tea	Twinings of London	China
	G10	Huang Shan Mao Feng	–	China
Oolong tea	O1	Tie Guan Yin Oolong	Twinings of London	Korea
Ginseng	E1	Red Ginseng Powder	Korea Ginseng Corporation	Korea
Herbs	I1	Lemon Twist	Twinings of London	Unknown
	I2	Misaki Woodland Fruits	Dulcimea	Unknown
	I3	Strawberry & Mango	Twinings of London	Unknown
	I4	Peach & Passion Fruits	Twinings of London	Unknown
	I5	Raspberry & Echinacea	Twinings of London	Unknown
	I6	Blackcurrant, Ginseng & Vanilla	Twinings of London	Unknown
	I7	Temptation Fruits Strawberry & Raspberry	Lipton	Unknown
	I8	Misaki Elderberry & Brier Rose	Dulcimea	Unknown
	I9	Fresh Mint Organic-Bio	Whittington	Unknown

reason (availability and time) we were only able to analyze black, green and oolong tea, and some herb samples. Indeed, it would be very helpful to be able to predict composition of similar products. With this respect chemometric evaluations are the most powerful. In the last years, the use of chemometric for food quality evaluation is increasing. In particular, in the context of holistic approach to food differentiation, where it is fundamental to integrate different analytical techniques providing independent data. Elemental analysis is a good source of data for chemometric. Indeed, it is possible to investigate relationships with elemental composition and samples classification according to their geographical or botanical origin (Szefer, 2007).

Usually, elemental analysis of teas and herb infusions is carried out by inductively coupled plasma atomic emission spectrometry (ICP-AES) (Chen et al., 2009; McKenzie et al., 2010; Salahinejad and Aflaki, 2010), ICP mass spectrometry (ICP-MS) (Pohl et al., 2016; Sofuoglu and Kavcar, 2008; Tokalıoğlu, 2012), and flame and graphite furnace atomic absorption spectrometry (Han et al., 2007; Pohl et al., 2016; Srividhya et al., 2011). Total reflection X-ray fluorescence (TXRF) is another technique that during the last years has gained interest for elemental composition analysis of vegetal foodstuff (Borgese et al., 2015; Dalipi et al., 2017; De La Calle et al., 2013). TXRF is a variation of energy dispersive X-ray fluorescence spectrometry that has a higher sensitivity and improved limits of detection at ppb level (μg/kg and μg/L). This is due to the geometrical configuration (Klockenkämper and von Bohlen, 2015). In total reflection conditions of X-rays, matrix effects, such as absorption and secondary excitation can be neglected and quantification can be done by the addition of a standard analyte, called internal standard (Bilo et al., 2015). TXRF has several advantages. Low amounts of sample are required for analysis (few μL or ng). Qualitative and quantitative analysis is faster and simpler. The introduction of new low power benchtop TXRF systems which are cost-effective and do not require gas or cooling media for operation has increased the popularity of TXRF. So far, only three contributions related with analysis of tea and herb infusions by means of TXRF were reported in literature. In these works, large-scale TXRF instrumentations with high-power X-ray tubes (Khuder et al., 2009; Xie et al., 1998) and a low-power benchtop TXRF system equipped with tungsten (W) X-ray tube (Marguí and Voutchkov, 2017) were employed.

In this study, we evaluate the capabilities of a low-power benchtop TXRF system equipped with Mo X-ray tube for multielement analysis of 40 different types of commercially available tea and herb samples. Infusions were also analyzed and elemental solubility was determined. Elemental daily intakes were calculated to evaluate consumer's exposure. Principal component analysis (PCA) of elemental composition data was performed for differentiation of tea and herb samples according to their botanical origin.

2. Experimental

2.1. Materials and reagents

Stock solution of Ga 1000 mg/L (in 5% HNO_3, TraceCERT®, standard for ICP, Sigma-Aldrich, Buchs, Switzerland) was used for the preparation of internal standard solution. Nitric acid (≥65%, puriss.