



THE CONTENTS OF INORGANIC COMPONENTS IN SELECTED POLISH AND ROMANIAN HERBAL TEAS INFUSIONS

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Abstract

The aim of this study was to analyze the contents of the selected inorganic ions (F^- , Cl^- , NO_3^- , PO_4^{3-} , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Ca^{2+}) and trace metals (Cr, Mn, Fe, Ni, Cu, Zn, Cd, Al, Pb, Co) in infusions prepared from popular herbal teas available in the Polish and Romanian markets. Ion chromatography with conductivity detection and microwave plasma-atomic emission spectroscopy were applied for this purpose. The experimental data were subjected to chemometric analysis to identify common traits and differences among them. Principal component analysis was employed to create a model using selected variables. The findings revealed that certain herbal teas exhibited elevated concentrations of potentially hazardous analytes, such as nitrate, cadmium, lead, and chromium. Based on these results, an assessment of the associated risk from these analytes was conducted.

Introduction

Herbal teas and medicinal plants are largely used mainly due to their health benefits and fragrance characteristics that result from the contents of the organic and inorganic compounds they contain. They are important

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sources of substances for human health as they contain various biologically active compounds (i.e. polyphenols, carbohydrates, proteins, amino acids, volatile organic compounds) (BUTIUK et al. 2016, DALAR and KONCZAK 2013, KACZMARCZYK and LOCHYŃSKI 2014, THEUMA and ATTARD 2020), as well as metals, inorganic anions and cations (MARTÍN-DOMINGO et al. 2017). Herbal teas are a large group, including herbs, fruits and their mixtures (VUONG 2014); some of them are available in supermarkets, while others can be bought at pharmacies.

The condition of the environment in which the herbs plant are grown (geographical location, climate, soil type, cultivation method) has an important impact on the composition and the quality of the final products (KOJTA et al. 2012, NGURE and KINUTHIA 2020). For that reason, the cultivation areas should be the subject to specific strict rules and monitoring (MUNTEAN et al. 2016). If we consider also that tea drinking is a ritual and a lifestyle in many countries, herbal teas are among the most popular beverages in the world and hold the second place in the beverage popularity ranking just after water.

However, there is a question whether using herbal teas or any other products derived from such raw materials have only positive effects, since besides beneficially biologically active constituents, medicinal plants contain also variable amounts of hazardous substances, such as heavy metals, pesticides, polyhydroxy aromatic compounds, mycotoxins, nitrates, etc. (MUNTEAN et al. 2013, 2016, STREET 2012). For most cases, anthropic pollution is the major cause, leading to unwanted side-effects of using contaminated plant remedies, many cases of liver, kidney or other organ damage being recorded (RODRIGUEZ-FRAGOSO et al. 2008).

Inorganic compounds present in food (including herbal teas) are responsible for a number of body functions. They have to be delivered with food as human beings are not able to synthesize those (ABD EL-ATY et al. 2014). Among the inorganic substances present in herbal teas, some metals and metalloids (Cr, Mn, Fe, Ni, Cu, Zn, Cd, Al, Pb, Co), and inorganic ions (F^- , Cl^- , NO_3^- , PO_4^{3-} , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} and Ca^{2+}) play important roles.

In turn, certain metals, such heavy metals which can be present in medicinal herbal teas, can have also a negative effect on human organisms. The World Health Organization (WHO) and the Food and Agriculture Organization of the United Nations (FAO) highlighted several metals because of their toxicity: Cd, Pb, Hg, As, Sb, Cu, Sn, Mn, Ca, Ni, Cr, Zn, Se, and Fe (BOLAN et al. 2017). Some of them play important biological roles in the living organisms and are necessary in certain concentration ranges for their normal functioning; microelements such as Cu, Zn, Co, Mo, Cr, V,

Mn and Fe belongs to this group. Nonetheless, they are harmful to the organism at high concentrations, when they become toxic (ERDEMIR 2018).

When taking into account the chemical composition of the consumed tea, its preparation and consumption methods are important (WELNA et al. 2013). Usually, herbal teas are prepared by infusion with water; hot water is poured over the prepared product portion or a tea bag and left under cover for approx. 10–20 minutes. Fresh or dried plant parts can be used, such as roots, leaves, fruits, or grains. However, more complex procedures are also used (PERRY 1996).

Various analytical methods and techniques are employed to study the chemical compositions of medicinal herbal teas, and tea infusions. They mainly include: chromatographic methods (MICHALSKI 2006a), capillary electrophoresis (HORIE and KOHATA 2000), neutron activation analysis (CHAJDUK 2009) or spectroscopic methods (FRASER et al. 2014, SZYMCZYCHA-MADEJA et al. 2012). For organic and inorganic ions, the ion chromatography is the dominant analytical method; it has been used for over 35 years as a reference method for determining anions and cations in water and wastewater (KONCZYK et al. 2019; MICHALSKI 2006b). Recently, its use largely expanded, including even nutrient analysis from seeds (YALDIZ and CAMLICA 2019) or phosphate from oils (ZHANG et al. 2013). On the other hand, microwave plasma-atomic emission spectroscopy has a very low utilization cost as it does not require expensive gases, while its detection limits are lower than those in the atomic absorption spectrometry (POIRIER et al. 2017).

The aim of the present study is to evaluate the elemental composition of 56 different herbal teas, and one green tea available on Polish and Romanian market. The contents of the main inorganic anions (F^- , Cl^- , NO_3^- , PO_4^{3-} , SO_4^{2-}) and cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}) as well as metals (Cr, Mn, Fe, Ni, Cu, Zn, Cd, Al, Pb, Co) were analyzed in teas infusions. Additionally, the chemometric evaluation of the studied teas was carried out to determine the common traits and differences. Based on the obtained results, an assessment of the associated risk from these analytes was conducted.

Materials and Methods

Reagents

To prepare the eluents for ion chromatography (IC) analysis, the following substances were used: Na_2CO_3 , $NaHCO_3$, PDCA (pyridine-2,6-di-

carboxylic acid) and 70% nitric acid (Sigma-Aldrich, Poland). The anion and cation standards (concentrations of $1,000 \pm 2$ mg/L) used to prepare the standard curves were from Fluka (Steinheim, Switzerland). The examined metal standards (concentrations of $1,000 \pm 2$ mg/L) used to prepare the standard curves for the microwave plasma-atomic emission spectroscopy (MP-AES) analysis were delivered by SPEX (Montreal, Canada). The deionized water (conductivity <0.08 $\mu\text{S}/\text{cm}$) used for the tea infusions, eluents and standard solutions was obtained from a Direct-Q 3 UV Millipore system (Merck, Germany).

Teas

54 herbal teas bought from supermarkets from Poland (48) and Romania (6) were used for the tests: peppermint (*Mentha piperita* L.), chamomile (*Matricaria chamomilla* L.), melissa (*Melissa officinalis* L.), linden (*Tilia platyphollos*), sage (*Salvia officinalis* L.), horsetail (*Equisetum arvense* L.), violet tricolor (*Viola tricoloris herba*), St. John's wort (*Hypericum perforatum* L.), calendula (*Hypericum perforatum* L.), nettle (*Urtica dioica* L.), wild rose (*Rosa canina* L.), 11 herbal mixtures with various compositions and only for comparison one popular green tea with prickly pear (*Opuntia ficus-indica*). The tea names were coded taking into account the country of origin (Poland – P; Romania – R) and the base plants: mint – MI; chamomile – CH; melissa – ME; linden – L; sage – S; horsetail – H; violet tricolor – VT; St. John's wort – SJ; calendula – CA; nettle – N; wild rose – WR; herbal mixtures – HM; green tea with prickly pear – GT.

For obtaining comparable data, water infusions were prepared as follows: 1.0000 ± 0.0001 g herbal tea samples was weighed in 250 mL Berzelius beakers, in which 100 mL of boiling deionized water were poured then left under cover for 10 minutes. The obtained infusions were filtered through a medium-density filter paper; after cooling, the infusions were transferred in 100 mL volumetric flask, bringing the volume to the mark with deionized water and homogenizing the filtrate. Afterwards, they were filtered through a $0.45\text{-}\mu\text{m}$ Nylon 66 syringe filter (Sigma Aldrich, Germany) in plastic containers, being analyzed with IC (inorganic ions) and MP-AES (metals). From each tea, two independent infusions were prepared, and each infusion was analyzed triplicate.

Analytical methods

The inorganic ions' content in the herbal tea infusions was determined with a 930 Compact IC Flex ion chromatograph (Metrohm, Switzerland) equipped with a conductivity detector; the separation conditions for IC

analysis are presented in Table 1. Calibration of the IC methodologies was carried out in accordance with ISO 8466-1 (POLISH NORM 2004). In order to determine the validation parameters, 6 standard solutions of anions and cations with concentration ranges given in Table 2 were prepared. The calibration solutions were analyzed in triplicate. The obtained areas of the analytes were used to calculate the standard deviation, the coefficient of variance, the limits of detection and limits of quantification, as well as the correlation coefficient. In addition, the recovery for the analyzed ions was calculated; the results are presented in Table 2. The range of concentrations in the standard solutions was matched to the expected concentrations in the real samples. The standard deviation ranges from 0.102 for PO_4^{3-} to 0.172 mg/L for SO_4^{2-} . The limits of detection (LOD) and quantification (LOQ) are also at good levels (from 0.1 for F^- and K^+ up to 0.25 mg/L for SO_4^{2-} , and from 0.27 for Na^+ up to 1.23 mg/L for PO_4^{3-} , respectively), with recoveries varied from 96.8 for PO_4^{3-} up to 104.2% for Mg^{2+} .

Table 1

Parameter	Anions analysis	Cations analysis
Analytical column	Metrohm Metrosep Supp 2 (150 x 4.0 mm)	Metrohm Metrosep C6 (150 x 4.0 mm)
Eluent	3.6 mM Na_2CO_3 + 1 mM NaHCO_3	1.7 mM PDCA + 1.7 mM HNO_3
Eluent flow rate	0.7 mL/min	0.9 mL/min
Detection	suppressed conductivity	non-suppressed conductivity

Table 2

Parameter	F^-	Cl^-	NO_3^-	PO_4^{3-}	SO_4^{2-}	Na^+	K^+	Mg^{2+}	Ca^{2+}
Concentration range [mg/L]	0.1–10.0	0.5–20.0	0.5–30.0	0.2–20.0	0.5–30.0	0.2–10.0	1–100.0	0.2–10.0	0.5–20.0
Standard deviation [mg/L]	0.105	0.167	0.142	0.102	0.172	0.103	0.121	0.104	0.137
Coefficient of variance [%]	1.02	0.85	0.62	2.07	0.95	0.68	3.41	1.47	1.30
Limit of detection (LOD) [mg/L]	0.10	0.17	0.23	0.41	0.25	0.09	0.10	0.17	0.21
Limit of quantification (LOQ) [mg/L]	0.30	0.51	0.70	1.23	0.75	0.27	0.28	0.51	0.63
Correlation coefficient (r)	0.9997	0.9989	0.9997	0.9996	0.9996	0.9997	0.9994	0.9996	0.9998
Recovery [%]	103.1	99.6	101.7	96.8	101.2	97.9	102.2	104.2	99.6

The trace metal content was determined by MP-AES, using a 4200 Agilent spectrometer (Agilent Technologies, USA). The limits of quantification for the determined metals by the applied method (based on 10-fold repeats for the standard samples) and other validation parameters are given in Table 3.

Table 3

MP-AES validation parameters for determined metals

Element	λ [nm]	Adjustment of the calibration curve	Concentration range [mg/L]	Limit of detection [$\mu\text{g/L}$]	Limit of quantification [$\mu\text{g/L}$]	Correlation coefficient (r)
Cr	425.433	linear	1.0–50.0	0.34	1.04	0.9997
Mn	403.076	linear	1.0–100.0	0.08	0.25	0.9998
Fe	371.993	linear	1.0–100.0	0.56	1.69	0.9999
Ni	352.454	linear	1.0–60.0	0.36	1.08	0.9995
Cu	324.754	linear	1.0–40.0	0.18	0.54	0.9995
Zn	213.857	nonlinear	1.0–20.0	0.43	1.29	1.0000
Cd	228.802	linear	1.0–20.0	0.32	0.97	0.9998
Al	396.152	linear	1.0–60.0	0.39	1.18	0.9996
Pb	405.781	linear	1.0–60.0	0.35	1.04	0.9997
Co	340.512	linear	1.0–60.0	0.35	1.04	1.0000

The data matrices were processed in Excel (Microsoft, USA), then principal component analysis (PCA) and cluster analysis were performed using MatLab (The Mathworks, USA) after mean center preprocessing.

Results and Discussion

Content of the inorganic anions and cations

The determination results concerning the contents of the inorganic anions and cations in milligrams per product portion [mg/p.p.] in the analyzed herbal teas are presented on Figure 1 and given in Table 4 (the product portion (p.p.) relates to the mass of analyzed tea listed in Appendix 1 – Table 1.1).

The contents of inorganic ions in the examined herbal tea infusions were different and depended on the tea type, its producer and country of origin (Table 4). The fluoride contents in the studied infusions ranged from 0.11 to 5.20 mg/p.p. (max. for mint tea P-MI2); the chloride contents ranged between 0.32 and 20.78 mg/p.p. (max. for herbal mixture tea P-HM5); the

nitrate contents ranged between 0.11 and 22.26 mg/p.p. (max. for nettle tea P-N1), the phosphate contents ranged between 0.16 and 17.75 mg/p.p. (max. for wild rose tea P-WR2) and the sulphate contents ranged from 0.31 to 27.92 mg/p.p. (max. for herbal mixture tea P-HM5).

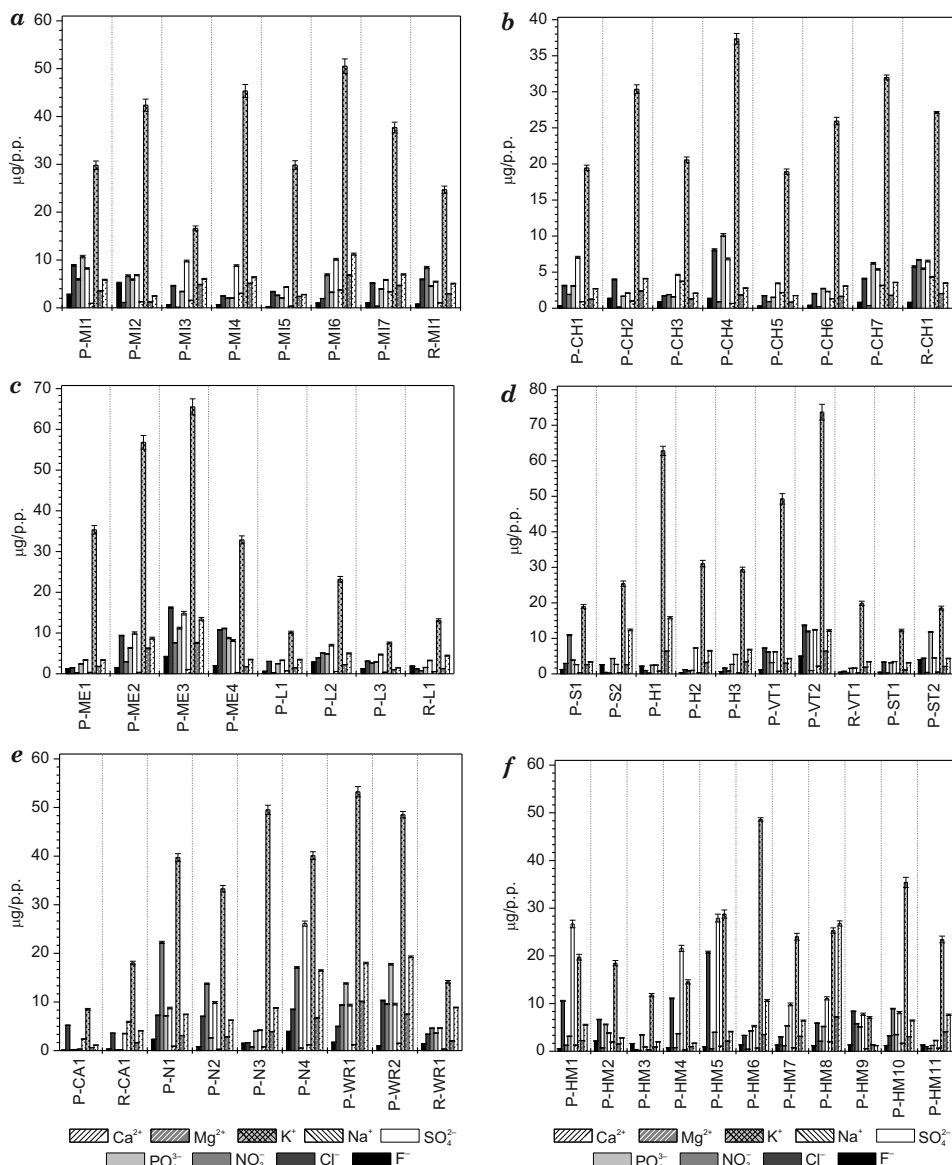


Fig. 1. Mean values of determined ions in particular tea types: *a* – mint; *b* – chamomile; *c* – melissa and linden; *d* – sage, horsetail, violet tricolor and St. John's wort; *e* – calendula, nettle and wild rose; *f* – herbal mixture

Table 4

Inorganic ions content in the studied herbal teas [mg/p.p.]

Ion	Minimum	Maximum	Median	Mean
F ⁻	0.11	5.2	1.23	1.47
Cl ⁻	0.32	20.78	3.76	5.11
NO ₃ ⁻	0.11	22.26	1.23	3.87
PO ₄ ³⁻	0.16	17.75	3.45	4.41
SO ₄ ²⁻	0.31	27.92	6.03	7.46
Na ⁺	0.18	5.92	0.86	1.27
K ⁺	7.05	73.69	27.95	30.30
Mg ²⁺	0.62	10.11	2.39	3.13
Ca ²⁺	1.13	26.8	4.72	6.51

Fluorine in the form of the F⁻ anions in large concentrations has a toxic effect on the human organism (BALCERZAK and JANISZEWSKA 2013); its excessive amount can disturb the absorption and metabolism of iodine, which is necessary for the normal functioning of thyroid and parathyroid glands. Tolerable upper intake level (UL) of fluoride for adults was established on value 7 mg/day. It means that drinking P-MI2 mint (5.20 mg/p.p.) provides the body with as much as 74% of UL (EFSA 2005a). Chloride is a major anion of the extracellular fluid; together with other electrolytes, it influences the water distribution in the organism and acid-base homeostasis. The Scientific Committee on Food did not establish a Population Reference Intake for chloride (EFSA 2021), but concluded that the requirements should match those for sodium (on a molar basis), i.e. 25–150 mmol/day. The US Institute of Medicine established an Adequate Intake (AI) for chloride at a level equivalent (on a molar basis to that of sodium), equaled to 2.3, 2.0 and 1.8 g/day, respectively for younger adults, older adults and the elderly (EFSA 2005b). The tested teas make a negligible contribution to achieving these AI values.

Nitrate and nitrite ions may be the cause of unfavorable changes as they are responsible for the oxidation reaction of iron in the hemoglobin to its Fe³⁺ form. As a result, methemoglobin (unable to bind oxygen) is formed. Phosphate is part of adenosine triphosphate (ATP) – a basic source of energy for the majority of living organisms. The phosphorous deficiency in the diet severely hampers human development, while its excessive amount has adverse effects because it contributes to osteoporosis and damages kidneys. At higher concentrations (up to 1,000 mg/L), sulphate can have a laxative effect and cause stomach problems. When having in mind the highest permissible levels of anions in food products, the obtained contents were not high enough to have a negative impact on human health.

However, a daily consumption of a few strong herbal tea infusions containing around 20 mg nitrate and more, phosphate or sulfate in one portion recommended by the producer may have an adverse effect. In the acidic environment of the stomach, nitrates undergo reduction to nitrites, with demonstrate toxic influence on the human organism. It ought to be remembered that humans ingest large amounts of nitrates with other foods (mainly vegetables, fruit, and meat products) every day (RACZUK et al. 2015) and any additional dose of these ions may exceed the acceptable daily intake (ADI) established at the level of 3.7 mg/kg of the body weight by the Joint FAO/WHO Expert Committee on Food Additives in 2002 (JECFA 2002). Therefore, the consumption of three P-N1 nettle tea portions in a day by a child weighing 20 kg will result in reaching the ADI level.

When comparing the mean contents of specific anions in the Polish tea infusions with the values obtained for the Romanian teas (Table 4), important differences were observed in the following cases: for fluoride in the mint and violet tricolor teas (contents two and six times higher in the Polish teas, respectively); for chloride in the linden and wild rose teas (contents over three times higher in the Polish teas) and violet tricolor tea (content nearly four times higher in the Romanian teas); for nitrates in the chamomile and mint teas (contents over seven and two times higher in the Romanian teas, respectively), and linden and wild rose teas (contents three-and-a-half and two times higher in the Polish teas, respectively); for phosphate in the wild rose and linden teas (contents four and two times higher in the Polish teas, respectively) and violet tricolor tea (content nearly three times higher in the Romanian teas) and for sulfate in the violet tricolor and wild rose teas (contents more than twice as high in the Polish teas).

The contents of the sodium, potassium, magnesium and calcium cations also varied depending on the herbal tea producers and types (Table 4). The differences observed for cations were bigger than those observed for anions in the same tea group. The following cation contents were established: for Na^+ - from 0.18 to 5.92 mg/p.p. (max. for calendula tea R-CA1), for K^+ - from 7.05 to 73.69 mg/p.p. (max. for violet tricolor tea P-VT2), for Mg^{2+} - from 0.62 to 10.11 mg/p.p. (max. for wild rose tea P-WR1) and for Ca^{2+} - from 1.13 to 26.80 mg/p.p. (max. for herbal mixture tea P-HM8).

Sodium and potassium are the basic electrolytes in the human biological system. Although no UL value has been established for these elements, it has been noted strong evidence for the contribution of sodium and potassium to high blood pressure in European populations. It is leading to the development of renal and cardiovascular diseases and stroke. The European Food Safety Authority (EFSA) Panels (EFSA 2019, 2016) point out that 2.0 g sodium/day and 3.5 g potassium/day is a safe and AI for the

general European population of adults. The sodium content in the studied teas was relatively low and did not usually exceed 2 mg/p.p., except for calendula teas and three mint teas. The Na⁺ content in Romanian calendula tea was more than twice as high as that found in the Polish tea. Potassium was the major cation present in all the teas (Table 4). Its content in the Polish violet tricolor as well as in melissa and in wild rose teas exceeded 60 and 50 mg/p.p., respectively. The Romanian violet color infusion had a three times lower K⁺ content in comparison with the mean content of these ions in the Polish teas (19.89 mg/p.p.). A nearly four times lower amount of the K⁺ ions was also observed in the Romanian wild rose infusion (14.14 mg/p.p.).

Magnesium ions belong to many enzyme activators and participate in the metabolic processes. According to EFSA Panel (2015), AI for magnesium is set at 350 mg/day for adult men and 300 mg/day for adult women. The calcium deficiency leads to hypocalcemia whose symptoms include seizures, nervous system weakness, general weakness, muscle weakness, and skin problems (VAN DRONKELAAR et al. 2018). A UL of 2,500 mg calcium/day from all sources was proposed for adults, and for pregnant and lactating women by EFSA Panel (2012). Besides herbal mixtures, the highest magnesium and calcium contents were observed in the Polish wild rose teas: the mean contents of those ions were four and two times higher than the contents in the Romanian teas, respectively. However, these values do not significantly affect the total intake of these elements in the daily diet. When comparing the mean contents of specific analytes in the Polish tea infusions with the values obtained for the Romanian teas, important differences were observed in few cases. They concern e.g. fluorides in the mint and violet tricolor teas, nitrates in the chamomile and mint teas. The differences observed for cations were bigger than those discerned for anions in the same tea group.

There are few studies on the metal content in herbal infusions (ALTINTIG et al. 2014, JURANOVIĆ et al. 2013, ÖZCAN et al. 2008, SCHUNK et al. 2016).

The content of sodium, potassium, calcium and magnesium in infusions of herbal teas from Polish producers was investigated by PYTLAKOWSKA et al. (2012): the highest content of Na (389 µg/g) they have found in the peppermint infusion, K (2650 µg/g – in melissa), Ca (114 µg/g) – in the infusion with nettle, and Mg (271 µg/g) – in sage herbal tea. These values are much lower than those obtained in this work, which may indicate the effect on the result of the analysis caused by the method of brewing, the origin of herbs, environmental conditions, etc. However, so far no one has examined and compared the content of these metals in Polish and Romanian herbal tea infusions.

Trace metal content

Figure 2 displays the mean concentrations of selected metals in the studied herbal teas; additional statistical information regarding the metal concentrations is summarized in Table 5.

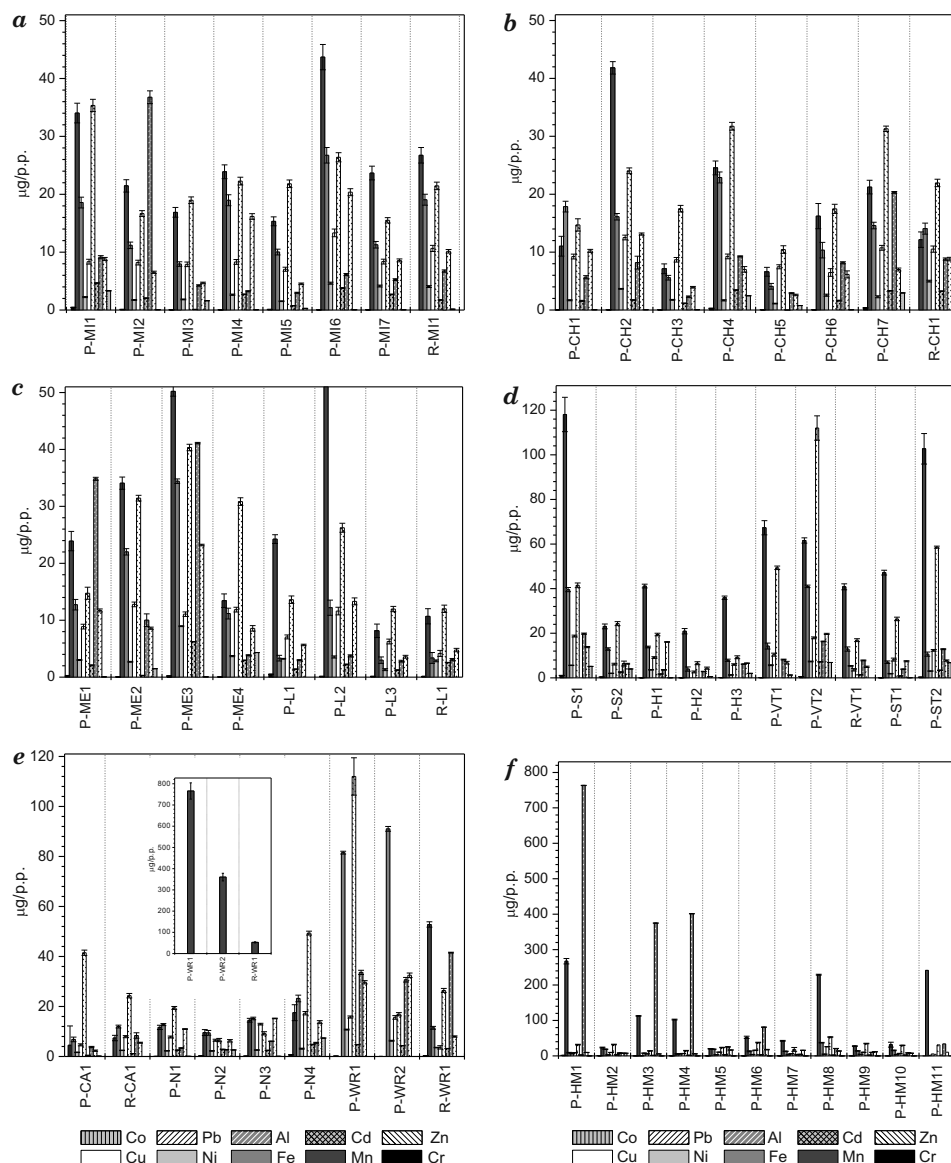


Fig. 2. Mean values of determined metals in particular tea types: a – mint; b – chamomile; c – melissa and linden; d – sage, horsetail, violet tricolor and St. John’s wort; e – calendula, nettle and wild rose; f – herbal mixture

Table 5

Trace metal content in the studied herbal teas [$\mu\text{g/p.p.}$]

Metal	Minimum	Maximum	Median	Mean
Cr	< LOQ	1.38	0.13	0.22
Mn	4.51	765.9	29.71	74.04
Fe	0.47	91.04	12.84	17.01
Ni	< LOQ	10.76	3.23	3.86
Cu	2.64	25.99	8.9	9.66
Zn	6.57	112.0	28.12	31.57
Cd	< LOQ	7.20	2.34	2.34
Al	2.30	763.35	8.30	47.94
Pb	2.32	32.49	7.87	10.34
Co	< LOQ	7.42	0.06	1.32

Cadmium is a highly toxic metal that belongs to the so-called *priority substances* with mutagenic and carcinogenic effects; it can be responsible for anemia and hypertension. Lead is usually transported into the human and animal organisms through consumption of contaminated plants. Zinc is a metal participating in the protein metabolism; its deficiency disturbs the functioning of the immune, nervous and reproductive systems. Copper is among elements necessary in the hemoglobin synthesis and in the iron absorption by the living organisms; the clinical research showed that the organism is more susceptible to diseases and infections if the copper deficiency is severe (SCHULZKI et al. 2017). From the toxicological point of view, chromium (VI) has a harmful influence on the organism; it is quickly absorbed and demonstrates carcinogenic and mutagenic characteristics (POHL et al. 2016). The nickel compounds have low toxicity, but they cause allergic symptoms if the organism is at a constant exposure. Iron is counted among microelements that have an important influence on the functioning of the living organisms. The soluble aluminum compounds have a negative impact on the human nervous system; it may be one of the factors influencing the development of the Alzheimer's and Parkinson's diseases (MALIK et al. 2013).

The contents of metals in the studied infusions were largely diverse. Among 10 tested metals, manganese and zinc demonstrated the highest share of the total contents of this group of elements in most infusions (Table 5). The analysis of 11 herbal mixtures also demonstrated large differences in the aluminum and manganese contents in the prepared infusions: the aluminum and manganese content were between 4.47 and 763.35 $\mu\text{g/p.p.}$ and

between 20.02 and 267.27 $\mu\text{g/p.p.}$, respectively, with definitely lower differences in the contents of the remaining metals. EFSA Panel (EFSA 2013) proposed an AI value of 3 mg Mn/day for adults, including pregnant and lactating women, whilst Regulation No. 1169/2011 of the European Parliament and of the Council (REGULATION (EU) NO 1169... 2011) indicates 2 mg of Mn as a reference value for beverages which means that not more than 150 $\mu\text{g/p.p.}$ may be present in 100 mL of the beverage. The manganese contents in the studied Polish wild rose teas, and three herbal mixtures exceeded this value. High manganese content observed in the infusions of Polish wild rose teas (765.92 and 360.27 $\mu\text{g/p.p.}$, respectively for P-WR1 and P-WR2) can be due to the fact that the manufacturer added hibiscus to these teas. 1 g of rose hips of the *Rosa canin* variety contains an average of 35 to 67 $\mu\text{g/g}$ of manganese (LEVENT *et al.* 2010) while 1 g of hibiscus contains about 390 μg of this metal (WRÓBEL *et al.* 2000). In turn, high content of manganese in herbal mixtures may be due to the presence of green tea (P-HM1), hibiscus (P-HM8) or birch leaves (P-HM11) in them. Both the results obtained in this paper for green teas (977 $\mu\text{g/g}$) and those presented in the literature (WRÓBEL *et al.* 2000) confirmed the high content of manganese and aluminum in this type of tea, and REIMANN *et al.* (2007) stated high content of Mn in birch leaves. The average AI concentration of 976.66 $\mu\text{g/p.p.}$ for the studied infusions is in the concentration range obtained for Brazilian herbal teas (SCHUNK *et al.* 2016). In the case of chamomile infusion ca. 5 times lower Al content was found in the Polish herbs (11.29 $\mu\text{g/g}$; this paper); 10.9–15.0 $\mu\text{g/g}$ (PYTLAKOWSKA *et al.* 2012) in comparison to the Brazilian herbs (51.62 $\mu\text{g/g}$).

The chromium content in nearly half the cases and cobalt content in 16 cases were lower than LOQ (0.10 $\mu\text{g/p.p.}$). According to the REGULATION (EU) NO 1169... 2011, not more than 7.5% of the daily reference intake for chromium (40 μg , i.e. max. 3 $\mu\text{g/p.p.}$) should be present in 100 mL of the beverage. None of the studied infusions demonstrated a value higher than 1.40 $\mu\text{g Cr/p.p.}$, which means that the chromium doses ingested with infusions are safe for the human organism. Cr concentration in Polish chamomile infusion is the same as in the case of the infusion prepared on the base the Brazilian chamomile (ca. 0.1 $\mu\text{g/g}$) but lower than that coming from Spain (0.61 $\mu\text{g/g}$) (MARTÍN-DOMINGO *et al.* 2017) or Turkey (1.22 $\mu\text{g/g}$) (BAŞGEL and ERDEMOĞLU 2005).

The iron contents ranged from below the LOQ level (<0.17, P-HM3, P-HM11) to 91.04 $\mu\text{g/p.p.}$ (P-WR2). For zinc, it was from 6.57 (P-H2) to 112.02 $\mu\text{g/p.p.}$ (P-VT2). The nickel contents ranged between below the LOQ value (0.11; P-H2) and 10.76 $\mu\text{g/p.p.}$ (P-WR1); for copper, the values ranged from 2.64 (P-H2) to 25.99 $\mu\text{g/p.p.}$ (P-HM8). Regulation (EU)

No 1169... 2011 defines daily reference intake values not only for manganese and chromium, but also for iron, zinc and copper at the levels of 14 mg (1050 µg/100 mL of the beverage), 10 mg (750 µg/100 mL of the beverage) and 1 mg (75 µg/100 mL of the beverage), respectively. When relating the reference values to the contents of particular elements in the studied infusions, it may be said that no recommended standards were exceeded in any case. However, the increased lead contents observed for the Polish wild rose tea infusions may be distressing from the health point of view. The values were approx. four times higher than the ones observed for the Romanian tea (Table 5). These differences may result, as in the case of manganese and aluminum, from the presence of hibiscus in infusions of Polish teas. Cadmium content varied between the below LOQ level (<0.10 µg/p.p; 9 infusions) and 7.20 µg/p.p (P-VT2), whereas the lead content ranged from 2.32 (P-CA1) to 32.49 µg/p.p. (P-WR2). Both of these metals are characterized by the highest toxicity among the tested metals. EFSA Panel on Contaminants in the Food Chain established tolerable weekly intake (TWI) of 2.5 µg/kg body weight to ensure sufficient protection of all consumers (EFSA 2012). In turn, adult exposure for lead was estimated at 0.50 µg/kg body weight per day (EFSA 2012).

The obtained mean concentrations of selected heavy metals were compared with the data obtained by other authors for sage infusions and presented in Table 6.

Table 6
Comparison of the obtained and literature data for selected elements in sage infusions

Reference	PYTŁAKOWSKA et al. (2012)	ÖZCAN et al. (2008)	MARTÍN-DOMINGO et al. (2017)	JURANOVIĆ CINDRIĆ, et al (2013)	This work
Element	the average content of the element [mg/g]				
Cr	–	20	2.02	< LOD	0.40
Mn	4.31	44	38.3	0.369	39.35
Fe	4.81	668	934	0.009	14.23
Ni	–	3	–	0.006	2.06
Cu	1.14	17	5.88	0.082	6.74
Zn	8.71	53	19.8	0.585	17.07
Cd	–	–	0.02	0.4	0.52
Al	35.5	14	–	< LOD	7.11
Pb	–	0.28	1.50	0.150	5.32
Co	–	2	–	–	2.31

The studied medicinal plants revealed diverse ionic and elemental profiles that can provide evidences for uses for certain purposes. It was established that in some instances, several herbal teas exhibit higher concentrations of hazardous analytes, such as cadmium, lead, or chromium; in such cases, the expected therapeutic effect can suffer a drawback due to the unwanted contaminants, with potentially negative effects. The variability of the contaminants' contents from the studied medicinal herbal teas can be due to the factors like differences between the plants species, geographical areas and exposure to different pollution sources or conditions during drying process. To assure the quality of the medicinal plants, good manufacturing practice shall provide a proper control of the raw material, with a special emphasis on a strict survey of the harvest areas and on processing in order to maintain the heavy metals' content at the lowest possible value. Since the medicinal plants and their extracts are used in traditional medicine, there is a possible hazard of heavy metal poisoning, if they originate from polluted areas; therefore, these raw materials should be collected from unpolluted regions and they should be analyzed for heavy metal content in order to avoid their cumulative toxicities in long-term use. Due to their hazard, the content of heavy metals has to be one of the main criteria for the use of medicinal plants as raw materials in the production of traditional remedies; hence it is essential to have a proper quality control to ensure safety and efficacy of herbal products.

Chemometric data analysis

Principal component analysis (PCA) of the full data set led to a model built on nine variables (the concentrations of the major studied ions), which reveals a fuzzy data structure, the model explaining only 54.9% of the variance (Figure 3a); the corresponding loading plot reveals a close correlation for the concentrations of chloride and sulfate/fluoride, nitrate and phosphate/potassium, calcium and magnesium (Figure 3b).

More suggestive is the PCA for anions' data set; the model corresponding for the scores' plot from Figure 4a and dendrogram from Figure 4b was built using five variables (the concentrations of the major studied anions), explains 77.22% variance and reveals three clusters: one containing the outliers P-HM4, P-HM1 and P-HM5 (with the highest concentrations of sulfate and chloride), another corresponding to the highest concentrations of fluoride, nitrate and phosphate and a major one, including most of the samples. This model emphasizes the herbal teas that can pose a risk on human health due the presence of hazardous nitrate anion.

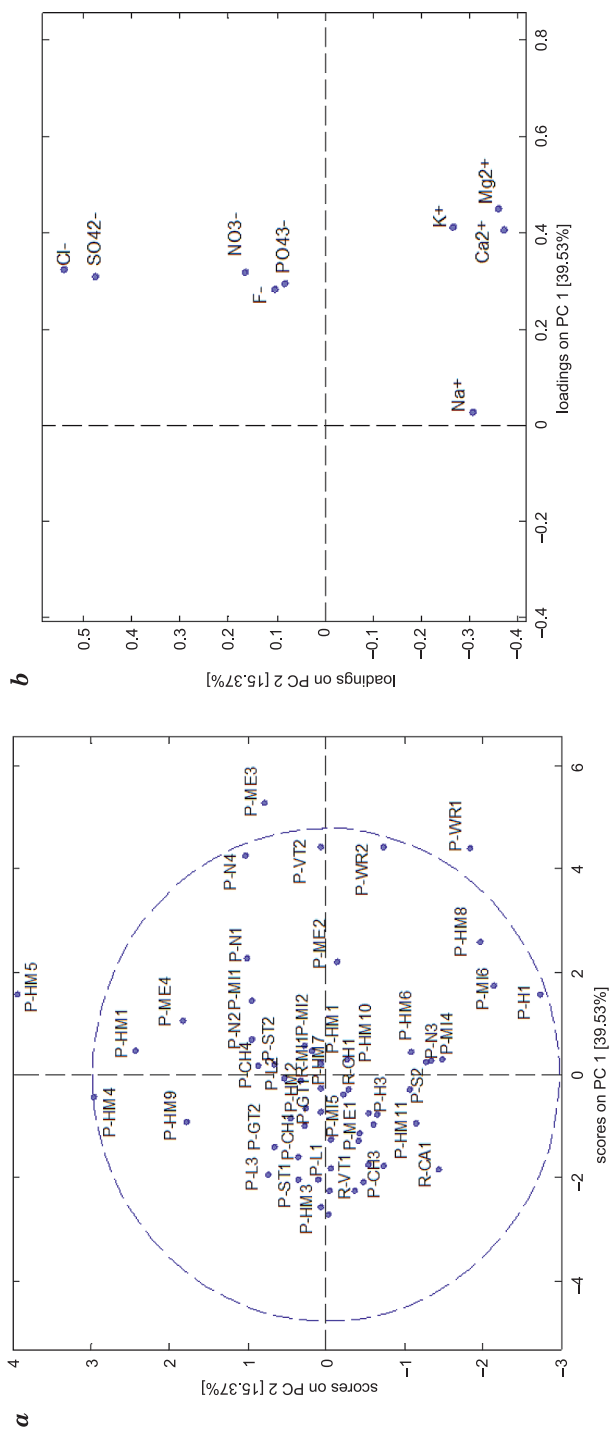


Fig. 3. The scores' plot (a) and the loadings' plot (b) for the full dataset

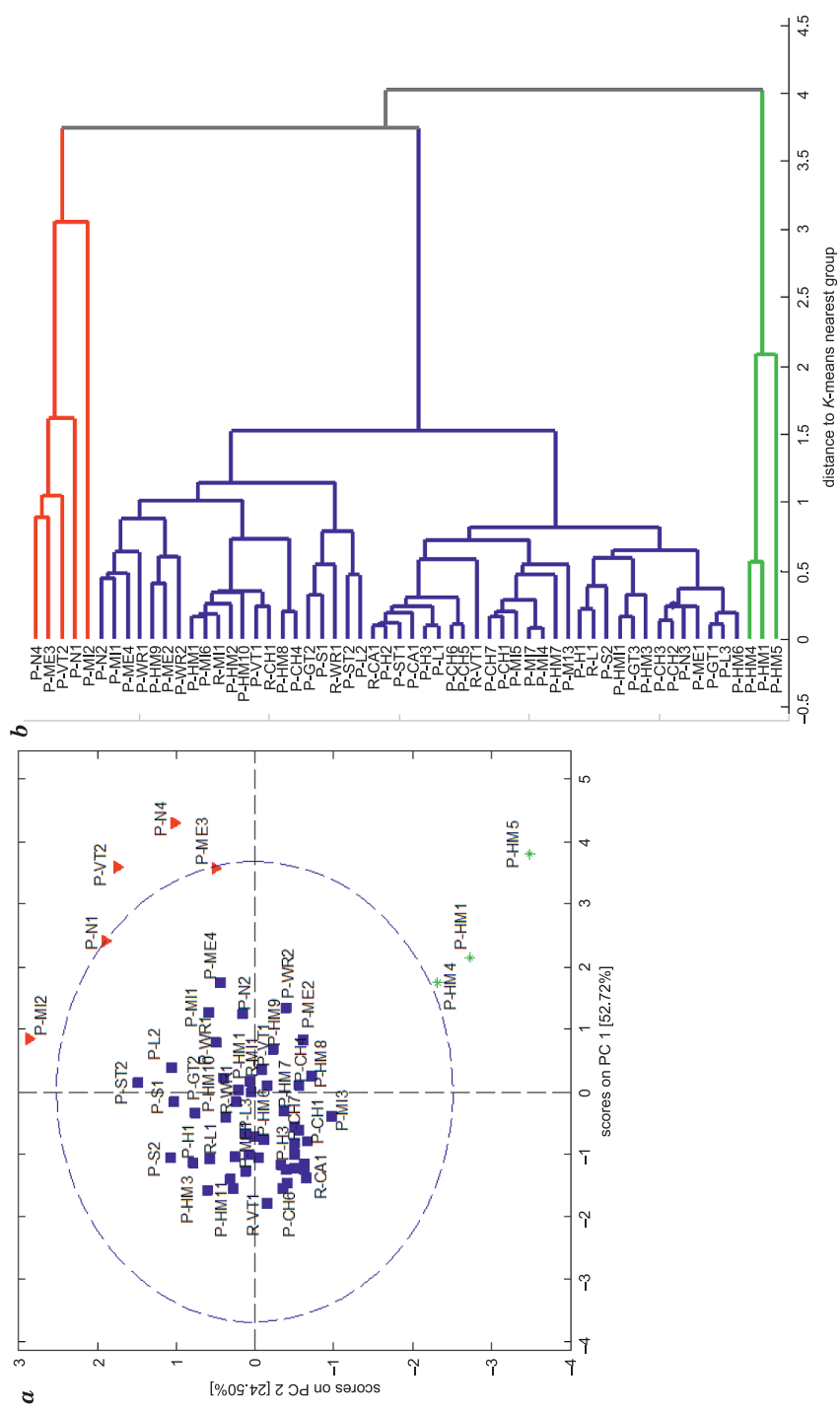


Fig. 4. The scores' plot for the anions' data set (a) and the dendrogram resulted from cluster analysis (K – means method) (b)

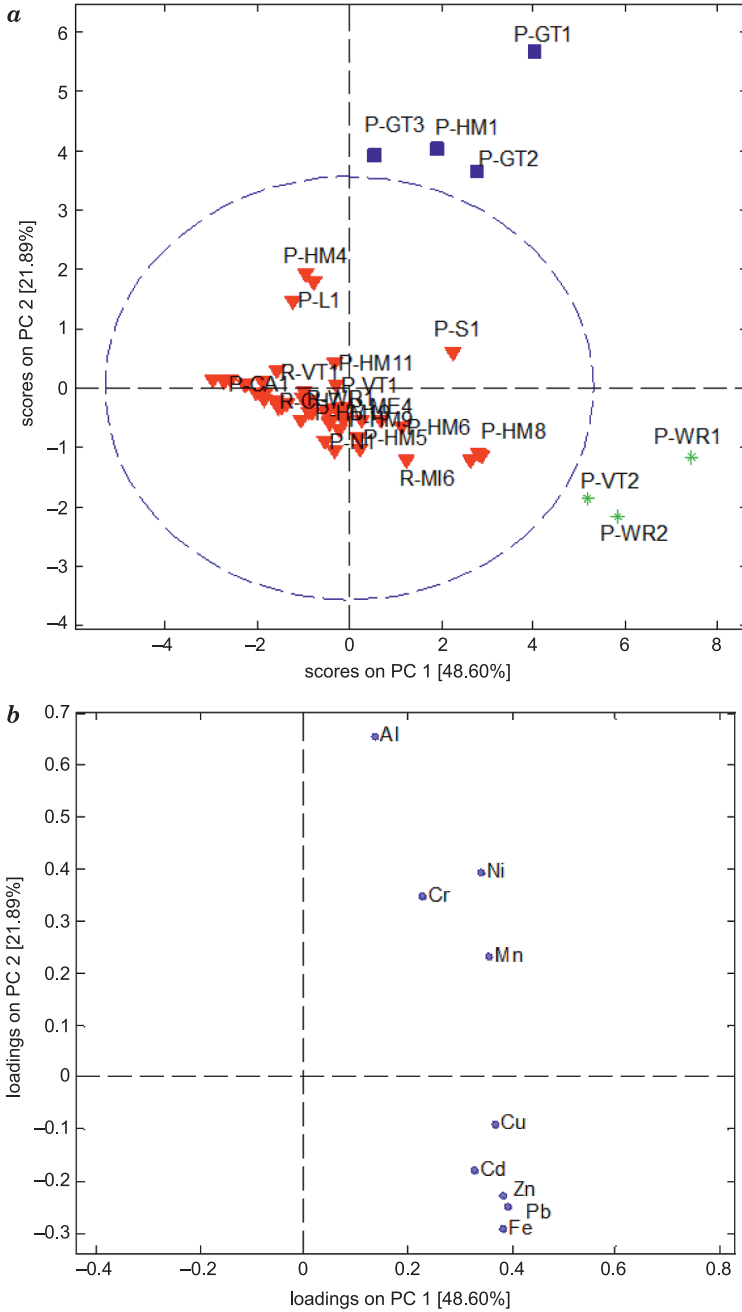


Fig.5. The scores' plot (a) and the loadings' plot (b) for the metal dataset

For the studied metals, PCA of the full dataset led to a model built on ten variables (the concentrations of the studied metals), which reveals four clusters, the model explaining 70.49% of the variance (Figure 5); the corresponding loading plot reveals a close correlation for the content of copper, cadmium, zinc, lead and iron (Figure 5b). Two from those three clusters contain the outliers green teas (P-GT) and PHM1 (in dark blue, with the highest concentrations of aluminum, chromium and nickel), another containing the samples with the highest concentrations of iron (PWR1, PWR2 and PVT2), a third one containing the samples high in nickel and copper (P-HM8, P-S1, P-N4, PME3) and a major one, corresponding to the most of the samples. This model emphasizes also several herbal teas that can pose a risk on human health due the presence of heavy metals of concern.

Conclusions

When comparing the mean contents of specific analytes in the Polish tea infusions with the values obtained for the Romanian teas, important differences were observed in few cases. These may have resulted from the country of origin, herb cultivation and harvest methods, as well as its processing, including additions of other herbs, aroma substances or spices that may have had an important impact on the effectiveness of extraction of particular ions into water in the herb steeping/brewing process. This may be crucial in terms of the health risks associated with drinking such teas. Consumers should be accurately informed about these parameters, given the popularity and easy access to herbal teas.

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Appendix 1

Table 1.1

Preparation method recommended by the manufacturer, according to the information provided on the product label

Code	Amount of tea [g] (p.p.)	Amount of water [mL]	Code	Amount of tea [g] (p.p.)	Amount of water [mL]	Code	Amount of tea [g] (p.p.)	Amount of water [mL]
P-MI1	1 tea bag (2.3)	200	P-MIE3	1 tea bag (1.7)	200	P-N1	1 tea bag (2.1)	200
P-MI2	1 tea bag (1.6)	200	P-MIE4	2 teaspoons (2.1)	150	P-N2	1 tea bag (1.7)	200
P-MI3	1 teaspoon (1.6)	150	P-L1	1 tea bag (1.5)	150	P-N3	1 tea bag (1.9)	200
P-MI4	1 tea bag (2.2)	200	P-L2	1 tea bag (2.0)	200	P-N4	2 tea bags (3.3)	200
P-MI5	1 teaspoon (1.1)	200	P-L3	1 teaspoon (1.3)	150	P-WR1	1 tea bag (4.1)	150
P-MI6	1 tea bag (2.5)	150	R-L1	1 teaspoon (1.1)	200	P-WR2	1 tea bag (3.6)	150
P-MI7	1 tea bag (2.2)	150	P-S1	1 tea bag (1.7)	150	R-WR1	1 teaspoon (2.3)	150
R-MI1	1 teaspoon (2.1)	200	P-S2	1.5 teaspoons (2.5)	100	P-HM1	1 tea bag (2.1)	200
P-CH1	1 tea bag (1.5)	200	P-H1	1 tea bag (2.5)	100	P-HM2	1 tea bag (2.2)	200
P-CH2	1 tea bag (2.0)	200	P-H2	1 teaspoon (1.0)	250	P-HM3	1 teaspoon (1.8)	200
P-CH3	1-2 teaspoons (2.1)	150	P-H3	1 teaspoon (2.0)	250	P-HM4	1 teaspoon (1.7)	200
P-CH4	2 tea bags (1.7)	200	P-VT1	2 teaspoons (3.0)	150	P-HM5	2 tea bags (2.2)	200
P-CH5	2 teaspoons (1.7)	150	P-VT2	1 tea bag (4.4)	200	P-HM6	1 tea bag (2.2)	200
P-CH6	1 tea bag (1.6)	150	R-VT1	1 teaspoon (1.0)	200	P-HM7	1 tea bag (2.2)	200
P-CH7	1 tea bag (1.8)	200	P-ST1	1 teaspoon (1.1)	200	P-HM8	1 tea bag (2.7)	200
R-CH1	1 teaspoon (1.4)	200	P-ST2	1 tea bag (2.3)	200	P-HM9	2 tea bags (2.2)	200
P-ME1	1 teaspoon (2.0)	200	P-CA1	2 teaspoons (1.0)	200	P-HM10	1 tea bag (2.0)	200
P-ME2	1 tea bag (1.4)	200	R-CA1	1 teaspoon (0.5)	200	P-HM11	2 tea bags (2.0)	200

