SEVEN publicações acadêmicas

Metallic species in yerba-mate and their infusions: Interrelationship of its distribution with phenolic compounds and melanoidins

Crossref doi
https://doi.org/10.56238/globalhealthprespesc-061

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ABSTRACT
Yerba mate (Ilex paraguariensisis), also known as mate, is a plant traditionally cultivated and consumed in different South America countries, where its green or roasted, dry leaves are usually employed to prepare chimarrão or mate tea, respectively. Mate presents a very complex chemical composition, rich in bioactive compounds, including polyphenols and melanoidins, wich in turn can have several several health effects, mainly due to their antioxidant capacity and their role in the prevention of multiple diseases, such as cancer, diabetes, and cardiovascular diseases. In addition, mate also tends to accumulate essential and potentially toxic trace metals, and their presence in beverages prepared as infusion from the dried leaves of the plant may be influenced by factors associated with the preparation of those infusions. The extraction of metallic ionic species to the infusions it dependent of the nature of phenolic compounds and melanoidins present in mate, since this group of bioactive compounds are capable of associate with metal ions through complexation. This type of association can lead to the formation of compounds with different solubilities in aqueous media, consequently impacting the solubility of metal ions in the infusions. Moreover, the presence of different chemical forms of metal ions in infusions may influence their absorption by the human body. Thus, it is essential to carry out studies to evaluate and understand how the essential and potentially toxic trace elements present in yerba mate may be related to potential impacts caused by these minerals on the general health of consumers of its hot infusions. The present review aims to establish a connection between the chemical composition of yerba mate and the presence of metallic species in its infusions.

Keywords: *Ilex paraguariensis*, infusions, trace metals, bioactive compounds.

1 INTRODUCTION

The yerba mate *(Ilex paraguariensis*) is a plant native to subtropical regions, produced mainly in southern Brazil, northern Argentina, Paraguay and Uruguay (BASTOS et al., 2007). Traditionally, the green or toasted yerba mate is used as an input for the preparation of tonic and stimulant drinks, such as tereré, mate and mate tea (DANIEL, 2009).

The yerba mate has several biological activities beneficial to health, resulting from the action of bioactive compounds, such as polyphenols, xanthines, caffeine derivatives, saponins and minerals (BASTOS et al., 2007; HECK and MEJIA, 2007; JUNIOR and MORAND, 2016; CAHUÊ et al., 2019).

Polyphenols – including chlorogenic acids, flavonoids, stilbenes and lignins – are responsible for part of the bioactive properties of yerba mate, which have relatively strong antioxidant activity and even act in the prevention of cancer, diabetes and cardiovascular diseases (ROBBINS, 2003; SOTO et al., 2011; ZHANG et al., 2019). Another pertinent characteristic of this class of compounds is the ability to complexe to ionic metallic species and change their chemical form and, consequently, their availability or distribution, in the leaves of mate and its infusions (STELMACH et al., 2013; MUSCCHINO and MUSCI, 2014).

In addition, it becomes increasingly important to conduct studies with yerba mate in order to better understand its therapeutic benefits and possible side effects associated with its content of metal ions, since this plant is a very rich source of minerals considered essential to the body. Such metallic species are naturally absorbed and accumulated in the leaves of mate, but can also be a result of soil and water contamination due to anthropogenic activities (SAIDELLES et al., 2010).

Currently, studies involving the determination of metal ions in mate and its infusions essentially address the determination of the total concentrations of elements considered essential or potentially toxic, such as Al, As, Ba, Ca, Cd, Cr, Cu, Fe, K, Mg Mn, Ni, Pb and Zn (ALBERTI et al., 2005; BRAGANÇA, MELNIKOV and ZANONI, 2011; Giulian et al., 2009; MARCELO et al, 2014; Pozebon et al, 2015; SAIDELLES et al., 2010).

In infusions of mate or mate tea, the metallic species present in yerba mate may be distributed in the form of free ions or complexed to bioligands, such as polyphenols, melanoids, among others (MUCCHINO and MUSCI, 2014). It is important to emphasize that there are several factors that influence the distribution of these elements both in the dried leaves of green or toasted yerba mate, as well as in their infusions. Thus, the present work seeks to review how the chemical composition of mate can be affected by conditions dependent on agronomic variables associated with its cultivation, as well as dependent on the storage process and its industrial processing. In addition, we also review how the availability of essential and potentially toxic trace metals present in the dried green and toasted leaves of mate, and in their respective infusions, may be affected by such conditions.

1.1 LITERATURE REVIEW

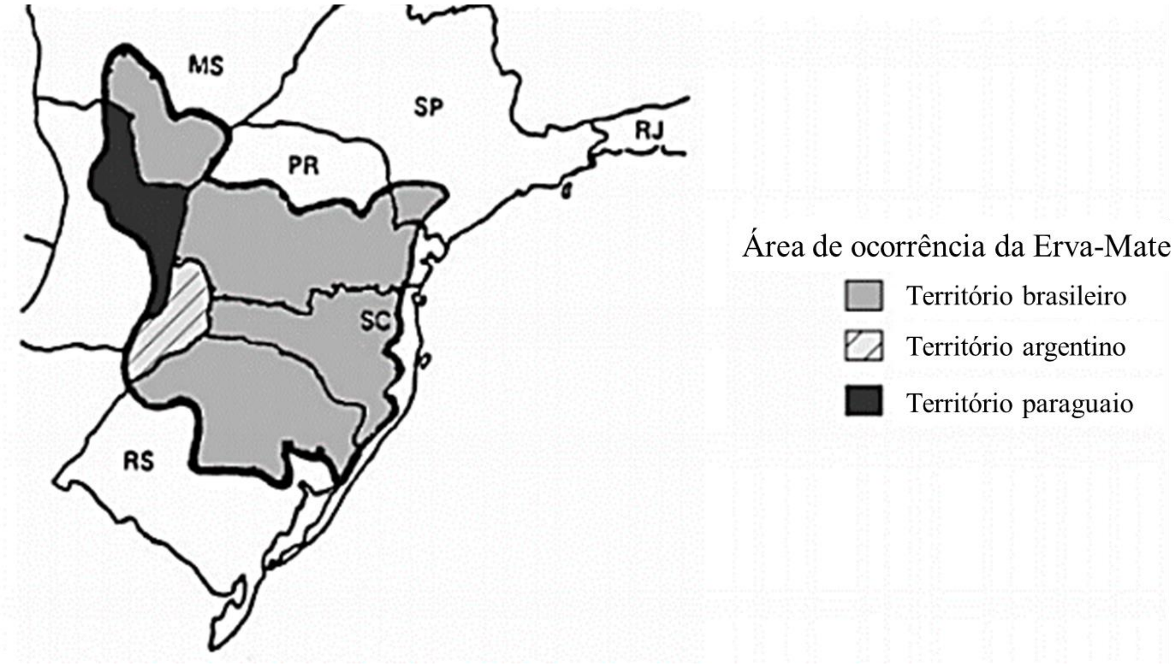
1.1.1 Yerba Mate (Ilex paraguariensis)

Mate tea, mate and tereré are stimulant drinks derived from yerba mate and are very popular in South American countries such as Brazil, Argentina, Paraguay and Uruguay. These drinks are usually prepared from the infusions (hot or cold) of the dried leaves of *Ilex paraguariensis* (Figure 1**), a plant native to South America** (**Figure 2**) and belonging to the Aquifoliaceae family (GAWRON-GZELLA, CHANAJ-KACZMAREK and CIELECKA-PIONTEK, 2021; HECK and DE MEJIA, 2007).

According to data provided by Embrapa (Brazilian Agricultural Research Corporation), the area of natural occurrence of yerba mate in Brazil corresponds to approximately 540,000 km2 (EMBRAPA, 2022). Historically, mate has been of great importance for the economy of several municipalities in southern Brazil, figuring as the main non-timber product of the region. The largest producers in the country are the states of Rio Grande do Sul, Santa Catarina, Paraná and Mato Grosso do Sul, the latter representing less than 1% of national production (IBGE, 2019).

Figure 1: Leaves of yerba mate (*Ilex paraguariensis*) (HECK and MEJIA, 2007).

Figure 2: Map of the area of natural occurrence of *Ilex paraguariensis* in some South American countries (adapted from OLIVEIRA and ROTTA, 1993).



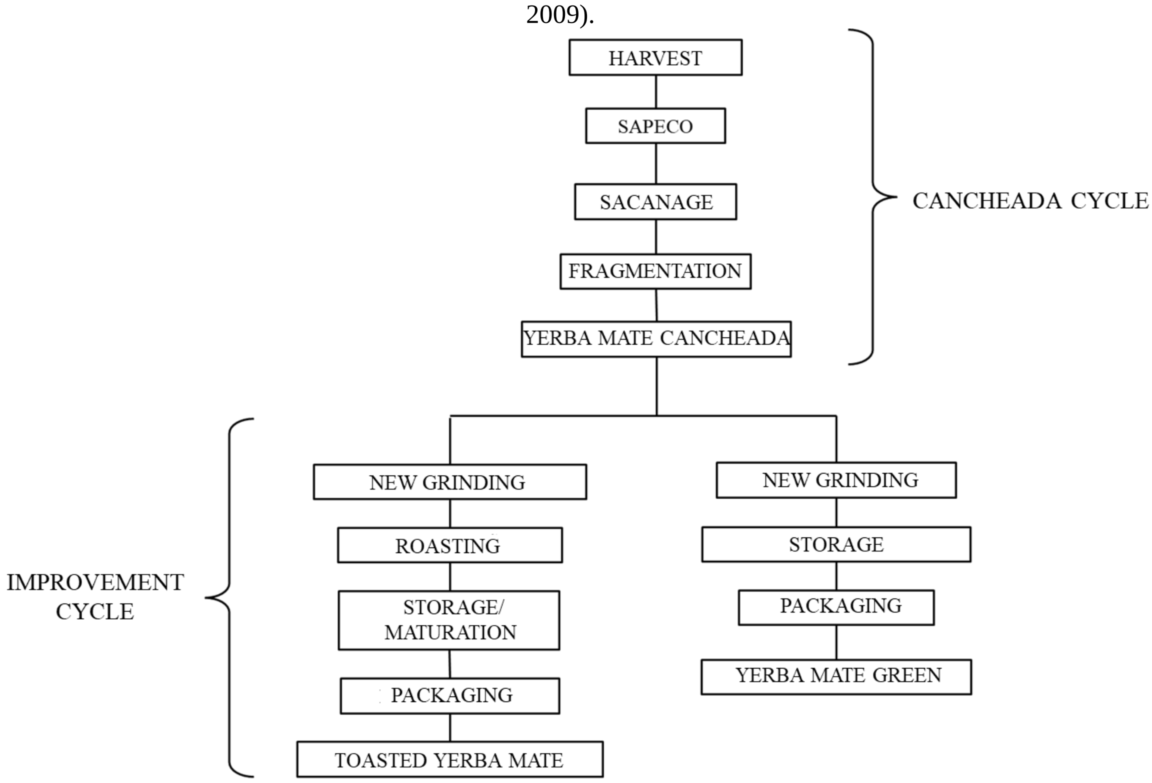
Between 2010 and 2017, there was a growing export of mate to the USA and African, Asian and European countries, signaling the expansion in the yerba mate market beyond the MERCOSUR countries (FAO, 2020). The commercial interest in yerba mate and its derivatives is mainly due to its numerous health benefits, which are linked to the presence of about 200 compounds with bioactive properties, such as saponins, alkaloids, polyphenols, essential oils, vitamins and minerals, among others (HECK and DE MEJIA, 2007).

There are several studies that attest to the positive effects that the regular consumption of mate has on the body (BASTOS et al., 2007; MIRANDA et al., 2008; Boaventura et al, 2012; KIM et al., 2015; JUNIOR and MORAND, 2016; RIACHI and DE MARIA, 2018; CAHUÊ et al., 2019), such as antioxidant activity, anti-inflammatory, stimulant to the central nervous system, diuretic action, hypocholesterolemic and hepatoprotective effects, in addition to providing good digestion, reducing the risk of cardiovascular diseases and acting in the control of diabetes and obesity (BASTOS et al., 2007; BRACESCO et al., 2011; RIACHI et al., 2017; GAWRON-GZELLA, CHANAJ-KACZMAREK and CIELECKA-PIONTEK, 2021).

Thus, in addition to the typical drinks prepared from yerba mate, its concentrated extracts can be used in the formulation of creams and face masks, which help in the reduction of free radicals of the skin, in the composition of anti-obesity drugs or thermogenic supplements. In the food industry, this herb is used for the preparation of functional foods, non-alcoholic energizing drinks, and its essential oil and chlorophyll can become an input in the composition of candies and gums or even as natural dyes and preservatives (MAZUCHOWSKI and RUCKER, 1997;

GAWRON-GZELLA, CHANAJ-KACZMAREK and CIELECKA-PIONTEK, 2021; Villanova, 2021). It is important to note that, despite the increasingly expressive export rates of yerba mate, the vast majority of this national product is still destined for domestic consumption, mainly in the forms of tereré, mate tea and mate (JUNIOR and MORAND, 2016).

Figure 3: Stages of the industrial processing of yerba mate (adapted from HECK and MEJIA, 2007; DANIEL, 2009).



As for the production of green and toasted yerba mate **(Figure 3)**, the processing of the leaves and branches of the plant can be divided between the cycles of cancheamento (or primary transformation) and processing.

In this way, the canching process includes the following steps:

a) Harvesting: the leaves and branches are harvested by pruning leafy branches for the preparation of the cancheada or fragmented grass;

b) Sapeco: consists of the rapid heating (10 s to 3 min) of the leaves at high temperatures (± 500 °C) to deactivate enzymes that promote oxidation (peroxidase and polyphenoloxidase), thus avoiding the blackening of the leaves of the herb after drying. In this process, the leaves lose approximately 20% of their moisture, acquiring the golden-green color, characteristic of a product with good quality;

c) Drying: the sapecadas leaves are introduced into heating chambers (±100 °C) for a period of 8 to 24 h. At this stage the process of dehydration of yerba mate is completed (the remaining moisture is of the order of 3 to 6 %), which is fundamental for the quality of the final product;

d) Fragmentation: the dried product passes through mechanical chippers and then is subjected to sieving to obtain the yerba mate cancheada.

As for the processing cycle, the industrial processing of mate can be carried out for the production of two different products: green yerba mate and toast:

a) green yerba mate: the grinding of yerba mate for the preparation of mate is carried out by a set of pestles until obtaining a very fine product, unlike the herb for the production of tereré, which is marketed in a fragmented form, but not ground. As for the storage time, the green yerba mate should be kept in stock for the shortest possible time, to ensure the maintenance of its green color;

b) Toasted yerba mate: used for the preparation of mate tea, the roasting of the product is carried out at relatively high temperatures (180 to 215 °C) for a period between 15 and 17 min. The maturation (aging) of mate, on the other hand, consists of increasing the concentration of compounds such as methylxanthines and polyphenols, and aims to develop and assign specific flavors to the drink, being preserved for up to 12 months.

The cultivation and harvesting of yerba mate can be conducted in different ways, depending on the producer and the region where they are carried out, as well as its industrial processing that may differ depending on the desired flavor for the mate, through the use of different times and temperatures in its different stages and also depending on the type of wood used for toasting and drying the product. It is worth mentioning that the way industrial processing is conducted can affect not only the quality of the product, but also the organoleptic characteristics and chemical composition of mate (ESMELINDRO et al., 2002; BASTOS et al., 2007; DANIEL, 2009; GAWRON-GZELLA, CHANAJ-KACZMAREK and CIELECKA-PIONTEK, 2021).

1.1.2 Chemical composition of yerba mate

The chemical composition of mate is quite complex and its varied amount of numerous organic compounds is directly associated with the bioactive properties that the regular consumption of yerba mate has on the human body. Are part of the composition of mate: essential oils, nutrients (carbohydrates, proteins and lipids), phenolic compounds, saponins, methylxanthines (caffeine, theophylline and theobromine), minerals and vitamins – mainly water-soluble, such as vitamins A, B1, B2, C and E (ESMELINDRO et al., 2002; HECK and DE MEJIA, 2007; BERTÉ et al., 2011).

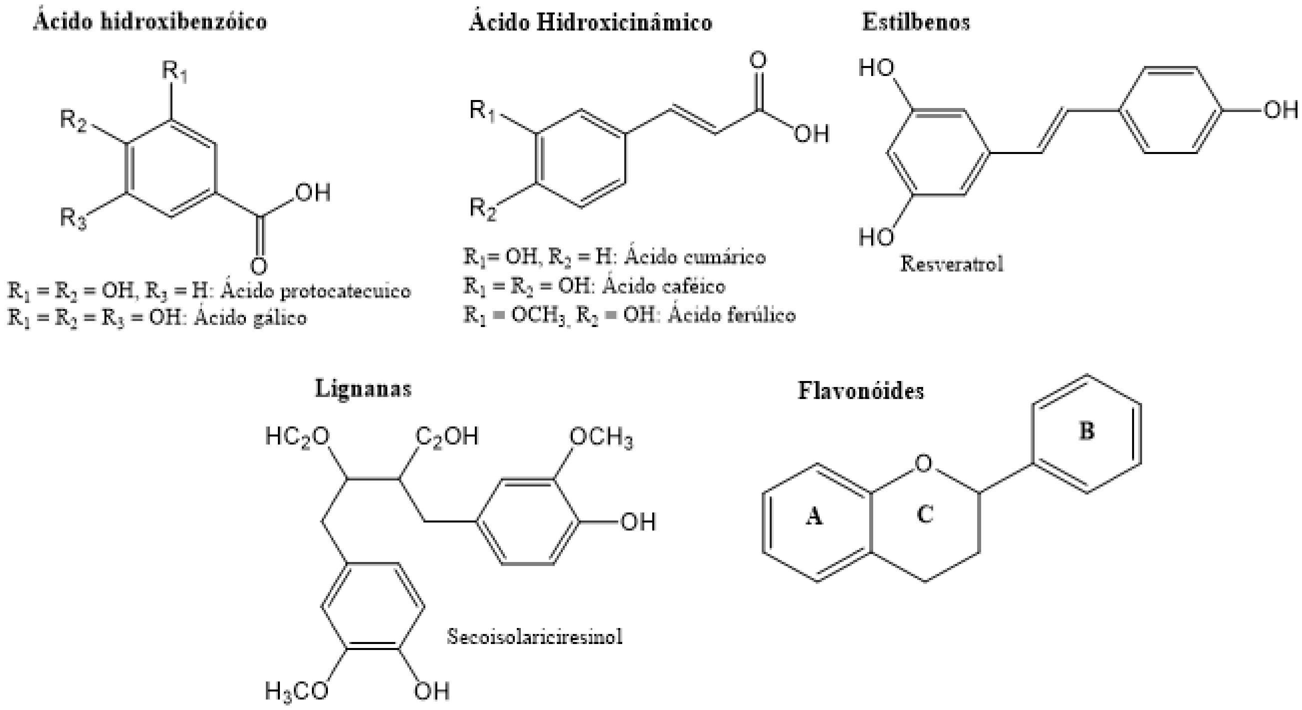
1.1.3 Phenolic compounds

During the development of a plant, its secondary metabolism produces compounds that aid in protecting against environmental stressors such as pests, solar radiation, and extreme temperatures. Among the secondary metabolites of yerba mate are phenolic compounds (or polyphenols), responsible for several bioactive properties, which also confer flavor, astringency and bitterness to fruits, vegetables, coffees, wines and teas (MANCH et al., 2004; VAUZOUR et al., 2010; ARCHELA and DALL'ANTONIA, 2013; ABBAS et al., 2017; RIACHI and DE MARIA, 2017).

Other external factors that accompany the development of yerba mate should impact on the presence of phenolics, such as conedaphoclimatic ditions (climate, air humidity, soil type, atmospheric composition, wind and river precipitation), fertilizer use, genetic variability and different culture media (MANACH et al., 2004; HECK, SCHMALKO and DE MEJIA, 2008; RIACHI and DE MARIA, 2017; RIACHI et al., 2018). Industrial processing, storage or maturation must also change the chemical composition of mate, since these compounds are easily oxidized (ESMELINDRO et al., 2002; ISOLABELLA et al., 2010; RIACHI et al., 2018).

Yerba mate is an important natural source of phenolic compounds (**Figure 4**), mainly chlorogenic acids (GCA) (caffeolquinic, dicaffeolquinic and ferulic acids), which are powerful antioxidants capable of chelating to metallic ionic species (BASTOS et al., 2007). In smaller quantities, flavonoids, stilbenes and lignans are also present, which should contribute to the bioactive properties of the plant (BASTOS et al., 2007; ARCHELA and DALL'ANTONIA, 2013; ABBAS et al., 2017; RIACHI and DE MARIA, 2017).

Figure 4: Chemical structure of phenolic compounds present in yerba mate.



Among the benefits to human health that a diet rich in polyphenols can provide, are listed the ability to combat reactive species involved in aging and / or chronic, autoimmune, inflammatory, osteoporosis, diabetes mellitus and the prevention of diseases associated with oxidative stress, such as cancer, neurodegenerative and cardiovascular diseases (MANACH et al., 2004; SOTO et al., 2011; Hardman, 2014; ABBAS et l., 2017).

As mentioned earlier, an important property of polyphenols is their ability to form complexes with metallic species. By acting as ligands of metal cations, polyphenols interfere directly in the chemical form with which these elements are distributed in yerba mate, and influence their absorption by the human body and establish, consequently, their degree of toxicity or essentiality (POHL, STELMACH and SZYMCZYCHA-MEDEJA, 2014; MUCCHINO and MUSCI, 2014).

1.1.4 Melanoidins

Melanoidins are nitrogenous polymeric compounds (macromolecules) of brown color, formed in the final stages of the Maillard reaction (RM), and consisting of reducing sugars and proteins (or amino acids) (BEKDAM, 2008), are associated with color, flavor and aroma of foods that undergo any type of heat treatment, such as baked goods, teas, coffees, wines or beers. In addition, they also have bioactivity, acting as antioxidants and antimicrobial, antihypertensive and anti-inflammatory agents (BEKEDAM, 2008; SHAHEEN et al., 2021; SINGH, TRIPATHI, CHANDRA, 2021; FENG et al., 2022).

Although their structures are not completely elucidated, it is well established that melanoidins are compounds of relatively high molar mass, with hydrophilic and anionic properties, capable of forming stable complexes with positive species and relatively low molar mass, such as metal cations (MORALES, FERNÁNDEZ-FRAGUAS, JIMÉNEZ-PÉREZ, 2005; COELHO et al., 2014; FENG et al., 2022).

1.1.5 Trace metals in yerba mate

Elements considered essential are essential to maintain the regular functioning of the human body, and are divided between micronutrients and macronutrients. In plants such as yerba mate, macronutrients are typically found in concentrations greater than 1000 mg kg^{-1} and these include Ca, K, Mg and Na; the micronutrients (or trace elements), such as Cu, Ni, Fe, V, Zn and Mn, represent a concentration lower than 100 mg kg^{-1} (GOLDHABER, 2003).

In contrast, potentially toxic metals such as inorganic As, Cd, Hg and Pb are of great concern from a toxicological point of view, since the chronic exposure of the body to these elements must cause serious damage to health, including cancer, damage to the neurological and cardiovascular systems, memory loss, damage to the kidney, among others. It is important to consider that even the elements considered as essential are likely to cause toxic effects to the body, when ingested in excess or when present in relatively high concentrations in food and beverage samples (CAO, et al., 2010; Saidelles, 2010; WHO, 1996).

The U.S. Food and Nutrition Board of the Institute of Medicine establishes Reference Daily Intake (DRI) levels for trace and potentially toxic metals, providing useful parameters to prevent nutritional deficiency or poisoning caused by some metal species. The DRIs (**Table 1**) include Allowable Daily Recommendation (RDA), Adequate Intake (AI) and Maximum Tolerable Intake Level (UL) and each of these values correspond to the average daily intake of a nutrient, and that may vary according to the age group of an individual (GOLDHABER, 2003).

Table 1: Recommended values of daily intake for essential and potentially toxic elements (GOLDHABER, 2003; NIH, 2022).

Element	RDA (mg/day)	AI (mg/day)	UL (mg/day)
Zn	11 (men) 8 (women)	ND	40
Cr	9	0.035 (men) 0.025 (women)	ND
Mn	ND	2.3 (men) 1.8 (women)	11
Fe	8 (men and women > age 51); 18 (women aged 19‒50 years)	ND	45
Se	0,055	ND	0,4
Ca	1000 (men 19 – 70 years old and women 19 – 50 years old) 1200 (men aged > 70 years and women aged > 51 years)	ND	2500
Mg	400 (men aged 19 – 30 years) and 420 (men aged > 31 years) 310 (women aged 19 – 30 years) and 320 (women aged > 31 years)	ND	350
Mo	0,045	ND	2

ND: not available; the values presented are established by the U.S. Food and Nutrition Board of the Institute of Medicine and are representative for men and women between 19‒70 years and > 70 years, unless otherwise indicated; RDA (allowable daily recommendation): average daily intake sufficient to meet the daily nutritional needs of almost all (97-98%) healthy individuals; AI (adequate intake): concentration of adequate daily intake, is used when scientific results are not sufficient to establish the RDA; UL: Maximum tolerable level of intake that would likely not expose an individual to side effects.

Yerba mate has a huge tendency to accumulate metals in its tissues, such as Al, As, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, V and Zn (HECK and MEJIA, 2007; KARAK and BHAGAT, 2010; SAIDELLES et al., 2010). The absorption of these elements by the human organism depends not only on their concentrations, but also on the physico-chemical forms with which they are found in the plant. Thus, it is of paramount importance to seek a more in-depth knowledge about the distribution and behavior of such elements in mate, aiming to establish the real therapeutic or toxicological effects that a regular consumption promotes.

It is important to emphasize that the concentration of trace and potentially toxic metals in yerba mate can vary due to external or internal factors, such as age of the leaves, presence of natural or synthetic fertilizers, soil type and pH, air pollution, and above all, as to the ways of processing and storage of mate (GIULIAN et al., 2009; KARAK and BHAGAT, 2010; SAIDELLES, 2010).

Jacques et al. (2007) studied how agronomic variables influence the concentration of metals in yerba mate leaves. In general, the mineral content was higher in plants grown in shaded areas, as well as in young leaves, where the concentration of some nutrients (K, Ca and Mn) progressively decreased with the age of the leaves. The use of nitrogen fertilizers increased the content of Ca and Mg, while fertilizers based on K and N, or only based on K, increased the levels of Mn and K, respectively. Thus, the study concluded that the metabolism of yerba mate and the absorption of certain metallic species are strongly influenced by their growing conditions, as well as the absorption of certain metallic species.

Another factor that should change the mineral content of the leaves of yerba mate are the different regions where the plant is grown. Marcelo et al. (2014) classified yerba mate samples according to their countries of origin, based on the determination of the total concentration of some metals in samples produced in Argentina, Brazil, Paraguay and Uruguay. From the data obtained, they concluded that the Brazilian samples were characterized by low concentrations of Ba, Ca, Mg and Al, while the Argentine samples had high levels of these same metals. In addition, Argentinean, Paraguayan and Uruguayan samples showed similar concentrations of Cu and P. Thus, the researchers were able to develop a new way to classify and characterize yerba mate according to the presence and content of metallic species.

However, the mineral content present in the leaves of yerba mate differs as to the mineral content of its infusions. Pozebon et al. (2015) studied this relationship and found that for most of the elements analyzed, the extraction rate for hot infusions ranged from 20 to 60%, for which Al, Ba, Ca and Sr were less leached when compared to Li, Rb and Pb, suggesting that some elements are more extracted than others. In addition, the authors emphasize that the extraction efficiencies are directly associated with the conditions used in the preparation of the infusions and should vary according to the water temperature, the extraction time, the degree of grinding of yerba mate (particle size), among other factors.

2 CONCLUSION

In the present review, we address how the influence of industrial processing, cultivation conditions of yerba mate, its genetic variability and the way its infusions are prepared can influence the chemical composition of its beverages and, in turn, also impact on its bioactive properties. It is important to emphasize the relationship between the presence of polyphenols and melanoidins and the distribution of metallic species, especially in infusions derived from green and toasted yerba mate, but also in their dry masses. Nevertheless, to better observe this relationship, it is extremely important to carry out more in-depth studies, which are also concerned with the bioavailability or bioaccessibility of these species in the infusions mentioned here, in order to deepen the knowledge about the true nutritional value of yerba mate in the different forms of consumption.

ACKNOWLEDGMENT

First, we thank the Coordination for the Improvement of Higher Education Personnel (CAPES) and the National Council for Scientific and Technological Development (CNPq) for the financial upport. We would also like to thank the APLE-A/CNPq Research Group and Prof. Dr. Edivaldo Egea Garcia, who work at the State University of Maringá (UEM).

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