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Original article

Is it safe to consume medicinal plants in mined areas? Investigating possible effects caused by a metal-contaminated plant in southern Brazil

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ABSTRACT

Mineral extraction areas are a significant environmental concern due to soil, water, and plant food resources contamination. Some medicinal plant species, such as those of the genus *Baccharis*, potentially bioaccumulate toxic elements. We evaluate the metal content from coal mining activity present in *Baccharis sagittalis* and in the soil; and whether this plant consumption represents a risk to human health. Cd and Pb presented levels that exceed those recommended by three global health agencies. Cd and Pb showed high levels in the projections of the daily intake recommended by international health agencies. After interviewing local residents close to mining areas, we found that 53.8 % of the interviewees mentioned the consumption of *Baccharis sagittalis* as infusion. These results indicate that the consumption of metal-contaminated *Baccharis sagittalis* can cause health problems as those metals accumulate in the human body. However, studies on Al, Ba, Cr, Cu, Mn, Ni, and Zn acceptable levels in plants consumed by humans are scarce. The contamination of plant species with associated traditional use close to mining areas can increase food security vulnerability of people who live near those areas and are constantly exposed to these agents, using plants gathered in the region.

Keywords: mining, medicinal plants, traditional communities, metals, human health.

Introduction

Mineral extraction has a vital role in the economy, especially in developing countries, yet it is responsible for environmental stress such as soil and water resources contamination (Weiler *et al*. 2015; Brisbois *et al*. 2019). Investment in mining and the search for new extraction areas have grown in recent years. Studies point that by 2050, mineral extraction fields and the number of metal-contaminated locations will have doubled (Schrecker *et al*. 2018; Candeias *et al*. 2019; Farjana *et al*. 2019). In countries such as China, South Africa, and Brazil, the extraction of mineral resources presents environmental concerns due to an increase in contaminated areas (Kemper *et al*. 1994; Quinn *et al*. 2011; Maitland *et al*. 2016; Elyamine

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et al. 2018; Zeng *et al*. 2019). Environmental impacts caused by mining range from changes in the landscape (*e.g.*, removal of plant species and soil layers) to changes in ecological interactions dynamics (*e.g.*, favoring of species that manage to survive in the impacted area and the removal of essential vegetable species for the local fauna), and contamination from different metals (Blanco *et al*. 2020a; Feng *et al*. 2020). However, even though these countries are increasing economic investments in the mineral sector, there is insufficient knowledge about the levels at which these elements are available in the environment, whether plants absorb them, and whether they represent a risk to human health (Brisbois *et al*. 2019).

Some metals are released into the environment as byproducts during extraction (Della Bosca & Gillespie 2018). In coal extractions, for example, pyrite release generates acid drainage that intensifies mineral weathering, producing high quantities of these minerals in the water and soil. Some of these metals, such as copper (Cu), manganese (Mn), and zinc (Zn) (Duffus 2002; Campos *et al*. 2003; Ashraf *et al*. 2019; Li *et al*. 2020), contribute to biologically essential functions like nitrogen availability in the soil for plants' growth and development. When available in low quantities, they do not present a risk to the functioning of ecosystem dynamics and human health (Licina *et al*. 2007; Oti 2015). However, at high levels (*i.e.*, Cu above 29 $mg.kg^{-1}$ and Zn above 39 $mg.kg^{-1}$ in soil) (Ashraf *et al*. 2019) or in unexploitable forms (*i.e.*, lacking a biological function in the system) such as cadmium (Cd) and lead (Pb), these metals can cause toxic effects to the ecosystem and human health (Duffus 2002). This toxicity is directly related to the exposure time and considers levels that exceed those recommended by health agencies (Li *et al*. 2020).

A few plant species that grow in mining areas can accumulate metals at levels greater than those recommended by national and international surveillance centers (Ashraf *et al*. 2019). Species of the genus *Baccharis*, with occurrence in North and South America, have been studied for their medicinal qualities and their capacity to grow naturally in mining areas due to their ability to accumulate metals (Carreira 2007; Souza *et al*. 2007; Haque *et al*. 2008; Menezes *et al*. 2013; Paula *et al*. 2016). In coal mining areas in southern Brazil, *B. trimera* contained high levels of Mn (*i.e.*, above 2.3 mg in 200 mL) and Zn (*i.e.*, above 11 mg in 200 mL) in their leaves, compared to these metals' availability in the soil (Souza *et al.* 2007). The aqueous extract of *B. trimera* from coal mining areas displayed a mutagenic effect in animal cells, with high levels of cellular damage (Menezes *et al*. 2015), along with genotoxic effects in *in vitro* blood cells (Menezes *et al*. 2015; Paula *et al*. 2016). In *B. sarothroides* leaves, present in copper mining areas in the United States, scientists observed hyperaccumulation of Cu, Pb, chromium (Cr), Zn, arsenic (As), and nickel (Ni) (Haque *et al*. 2008). In addition to this situation, some species of the genus *Baccharis* are known widely and used for medicinal purposes in Brazil, being recognized as such by the National Health Surveillance Agency (ANVISA) and presented in the National List of Medicinal Plants of Interest to the Unified Health System (RENISUS) from Brazil (Marmitt *et al*. 2015).

The proximity increase of mining areas to human populations, including indigenous peoples and traditional and local communities, has grown in the last 20 years, generating territorial conflicts and food insecurity (Alonso *et al*. 2018; Horowitz *et al*. 2018; Vega *et al*. 2018). Ensuring food security is a challenge in the 21st century since, in addition to providing food for everyone, guaranteeing its safety is also needed (Marrugo-Negrete *et al*. 2020). However, due to the proximity increase to contaminated areas, indigenous peoples and traditional and local communities' health and food security are threatened; these populations are three-four times more vulnerable to diseases from unhealthy or unsafe food (Brisbois *et al*. 2019; Marrugo-Negrete *et al*. 2020). This vulnerability is partially due to food contamination from metals produced by mineral extraction (Brisbois *et al*. 2019). In countries such as China, which accounts for one of the largest volumes of coal mining worldwide, solutions to ensure food safety (Juric *et al*. 2018; Sun *et al*. 2019) include an environmental safety law and a resolution for acceptable levels of metals in food and tea (Ghose 2014; Yi *et al*. 2018). However, few studies have analyzed food security and the contamination of food and medicinal plants consumed by local communities living close to mined areas (Brisbois *et al*. 2019; Blanco *et al*. 2020a).

In coal extraction areas in southern Brazil, there is a mosaic of mining areas within human communities that use vegetable species for food and medicine (Blanco *et al*. 2020a). Many of these rural and urban neighborhoods established themselves specifically to support mining activity, occupying regions extremely close to the mines (Blanco *et al*. 2020a, b). In these areas, many *Baccharis* species known as *carqueja* are found, including *Baccharis sagittalis*, *B. trimera*, and *B. sarothroides,* all traditionally used as medicine (Santos *et al*. 2008; Karam *et al*. 2013; Stolz *et al*. 2014) *B. sagittalis*, commonly consumed as an infusion, is abundant and occurs spontaneously in these areas (Blanco *et al*. 2020a). Thus, this study aimed to investigate the contents of aluminum (Al), barium (Ba), Cd, Cr, Cu, Mn, Ni, Pb, and Zn in *B. sagittalis*, the frequency of this species' consumption by local people, and whether it represents a risk. The tested hypothesis is that in mining areas, *B. sagittalis* contains high levels of those metals, and its consumption can endanger human health.

Materials and methods

Data collection

Data was acquired in the Santa Catarina coal basin, the second biggest coal-mining region in Brazil. Six mined areas with *Baccharis sagittalis* (Less.) DC (Asteraceae) (voucher EAFM16711) occurrence were selected; three of these areas (S 28°32'34.6'' W 049°29'34.8'' Lauro Muller, S 27°31'56.9'' W 048°30'44.4'' Urussanga, and S 28°29'54.1'' W 049°22'57.9'' Treviso) experienced extensive coal mining activity (until coal depletion). In these areas, Al, Ba, Cd, Cr, Cu, Pb, Mn, and Zn presence in the soil had already been detected (Campos *et al*. 2010; Hugen *et al*. 2013; Souza *et al*. 2016), with no record of other anthropic activities affecting soil composition. The other three areas (S 28°32'33.2'' W 049°20'53.6'' Lauro Muller, S 28°32'34.5'' W 049°29'34.8'' Urussanga, and S 28°29'54.7'' W 049°22'57.5'' Treviso) were close to the mining areas ones but sustained no record of mineral exploration or anthropogenic activity (Fig. 1). The distance between mined and unmined areas spanned a maximum of 10 Km. Two transects were covered in each of the six collection areas, comprising ten whole individuals collected 15 m to 20 m apart. For each plant, the soil was obtained through three sub-samples, collected with the aid of an auger, at a depth of 20 cm. These subsamples were combined into a single sample for each individual of *B. sagittalis* and then air dried. To avoid contamination of samples from one collection to another, the auger was cleaned after each collection. In total, 60 individuals and 60 soil samples were obtained (*i.e.*, 30 samples from mined areas and 30 samples from unmined areas).

Preparation and analysis of soil samples

After pulverizing the samples to a fine powder in an agate mortar and straining them in a 0.149 mm sieve, they were subjected to acid digestion following the USEPA 3050 B method (USEPA 1996). Method reliability measurement was performed using a reference soil material CRM-Agro E2002a (EMBRAPA 2015) and cell samples for the Qualitative Limit of Detection (QLD) (Tab. 1). Then, The Instrument Detection Limits (IDL) according to APHA was calculated (APHA 2017). All analyses were performed in duplicate.

At last, Al, Ba, Cd, Cr, Cu, Pb, Mn, and Zn contents were quantified in an inductively coupled plasma optical emission spectrometer (ICP-OES); and Ni content in an air-acetylene flame atomic absorption spectrometer (F-AAS).

Preparation and analysis of plant material

Analysis in *B. sagittalis* samples comprised the same metals quantified in soil samples. The plants were weighed and dried in a greenhouse at 45° C for 42 hours, and dried again in 12 hours intervals to constant weight. The samples were macerated and stored for subsequent opening via the USEPA 3050 B method (USEPA 1996). The reliability of the analytical method was assessed using a reference sample from the CRM-Agro E1001a - *Brachiaria Brizantha* leaves (EMBRAPA 2013).

Interviews and daily ingestion

After getting their informed consent, semi-structured interviews were conducted with residents of the communities found at a maximum distance of 300 m from the mined areas, individually, between February and March 2018. A total of

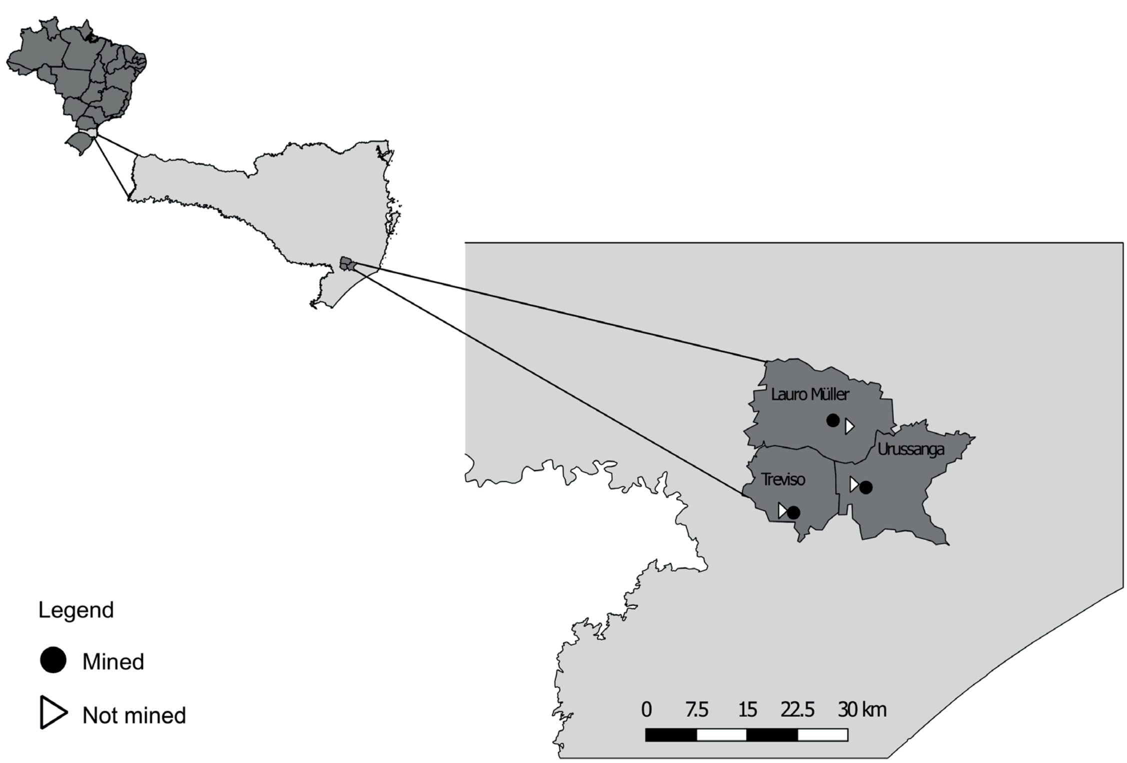

Figure 1. Map of samples areas in southern Brazil. Ten samples of *B. sagittalis* and soil were collected in each of the six locations identified on the map. Triangles represent unmined areas and circles represent mined ones.

14 local communities (Vila Funil, Rio Carvão, Barreiros, Guaitá, Cidade Alta, Vila Visconde, São Sebastião Alto, Vila São Jorge, Rio Fiorita, Volta Redonda, Campo Morozini, Santa Luzia, Santa Augusta, and São Sebastião), belonging to 6 municipalities (Criciúma, Forquilhinha, Siderópolis, Treviso, Urussanga, and Lauro Müller) were selected to take part in the study, given their historical background in mining activity. Within each community, house visits were paid only once, and interviews were conducted exclusively with those interested in participating in the research. Interviewees were questioned if they knew and consumed *B. sagittalis*, where they usually collected it, and asked about its consumption frequency. For measurement of *B. sagittalis* consumption frequency, each interviewee answered how many cups (200 mL) they ingested per week (see Text S1 for the questionnaire used). Three levels (De Godoy *et al*. 2013) were used to classify residents' frequency of *B. sagittalis* ingestion: small (*i.e.*, once a week or less), medium (*i.e.*, two-three times over the week), and frequent (*i.e.*, four-seven times a week or more). Based on worldwide information on the consumption of one tea bag (on average each bag is 2 g) per preparation. A consumption projection of 2 g per cup (200 ml) was performed and the estimate was calculated, as a rule of three, of the intake associated with Cd, Cr, Cu and Pb (based on the values indicated by international and national agencies (ANVISA 2013; Soliman 2016; Westman 2018).

Statistical Analysis

Generalized linear models (GLMs) were used to compare the differences in metal concentrations in *B. sagittalis* leaves from mined and unmined areas. All models met the premises of normality and homoscedasticity, and for each metal, a model was created using a gamma distribution family. For graphical representation, boxplots were generated. Parameters followed ANVISA RDC resolution nº 42 of August 29, 2013, for Brazil (ANVISA 2013), as well as maximum contamination levels for China (Westman 2018) and the European Union (Soliman 2016), given that both provide reference values for acceptable levels of many metals in herbs and infusions (ANVISA 2013; Soliman 2016; Westman 2018).

Results

The reference values of the elements Al, Ba, Cd, Cr, Cu, Mn, Ni, Pb and Zn for the soil and *Baccharis sagittalis* are reported in Tab. 1 and 2, respectively. The values obtained were compared with a reference base, with the values of CRM-Agro E2002a for the soil and for *B. sagittalis* the values of CRM-Agro E1001a.

The average contents of metals quantified in the soil samples are reported in Table 3. Al, Mn, and Pb were present in higher concentrations in soil from mined areas (16453.97 ± 2235.53 mg.kg^{-1}, 308.22 ± 224.96 mg.kg^{-1}, and 15.80 ± 9.69 mg.kg^{-1}, respectively) when compared to unmined areas (14594.98 ± 5132.04 mg.kg^{-1}, 87.92 ± 69.49 mg.kg^{-1}, and 11.86 ± 5.51 mg.kg^{-1}, respectively) ($p < 0.05$). The samples from unmined areas showed higher contents of Cr, Cu, and Ni (10.46 ± 2.98 mg.kg^{-1}, 41.63 ± 12.11 mg.kg^{-1}, and 18.05 ± 5.50 mg.kg^{-1}, respectively) in comparison to samples from mined ones (8.18 ± 3.48 mg.kg^{-1}, 22.13 ± 9.91 mg.kg^{-1}, and 13.86 ± 4.23 mg.kg^{-1}, respectively) ($p < 0.05$). The contents of Ba, Cd, and Zn showed nonsignificant variations between the two sampled groups ($p \geq 0.05$).

Table 1. Reference obtained values of the elements. Al, Ba, Cd, Cr, Cu, Mn, Ni, Pb and Zn of the reference sample CRM-Agro E2002a (EMBRAPA 2015) and Instrumental Detection Limit (IDL) (APHA 2017). Metals were quantified by ICP-OES, except for Ni, which was quantified by F-AAS.

Metal	Obtained Contents (mg.kg^{-1})	Reference Contents (mg.kg^{-1})	IDL
Al	58.20 (g.kg^{-1})	†	0.61
Ba	7.12	†	0.04
Cd	86.02	94.0 ± 11.4	0.03
Cr	77.49	120.0 ± 30	0.03
Cu	8.56	8.8 ± 4.0	0.09
Mn	76.28	130.0 ± 20	1.18
Ni	6.87	†	0.12
Pb	104.50	173.8 ± 18.8	0.51
Zn	6.09	†	0.19

Note: The symbol (†) means that the metal was not presented in the CRM-Agro E2002a reference material Brazilian Agricultural Research Company (EMBRAPA 2015).

Table 2. Reference obtained values of the elements. Al, Ba, Cd, Cr, Cu, Mn, Ni, Pb and Zn of the reference sample CRM-Agro E1001a - *Brachiaria Brizantha* leaves (EMBRAPA 2013) and Instrumental Detection Limit (IDL) (APHA 2017). Metals were quantified by ICP-OES, except for Ni, which was quantified by F-AAS.

Metal	Obtained Contents (mg.kg^{-1})	Certified Contents (mg.kg^{-1})	IDL
Al	25.3	†	0.61
Ba	3.24	†	0.08
Cd	10.91	19.9 ± 5.1	0.03
Cr	1.41	3.3 ± 1.66	0.03
Cu	3.18	4.00 ± 0.7	0.09
Mn	53.34	70.19 ± 18.00	0.95
Ni	0.59	†	0.47
Pb	2.41	4.00 ± 1.80	0.11
Zn	6.13	9.90 ± 1.60	0.26

Note: The symbol (†) means that the metal was not presented in the CRM-Agro E1001a reference material l Brazilian Agricultural Research Company (EMBRAPA 2013).

Al and Mn had the highest mean concentration in mined soil with a high standard deviation (16453.97 ± 2235.53 mg.kg^{-1} and 308.22 ± 224.96 mg.kg^{-1}, respectively). Considering reference levels from Santa Catarina state, Cr and Cd showed values above the acceptable level (5 mg.kg^{-1} and 0.12 mg.kg^{-1}, respectively) (Hugen *et al*. 2013; Souza *et al*. 2016), both in mined and unmined soil. In addition, Cu also presented concentration levels above the allowed, in unmined areas (29 $mg.kg^{-1}$) (Hugen *et al.* 2013) (Tab. 3).

In plant specimens, Ba, Cd, Mn, and Zn showed higher concentrations in leaves of individuals collected in mined areas (11.69 ± 10.25 $mg.kg^{-1}$, 0.30 ± 0.09 $mg.kg^{-1}$, 335.8 ± 212.60 $mg.kg^{-1}$, and 23.34 ± 9.51 $mg.kg^{-1}$, respectively) when compared to those obtained from unmined ones (3.64 ± 1.84 $mg.kg^{-1}$, 0.25 ± 0.04 $mg.kg^{-1}$, 263.87 ± 79.58 $mg.kg^{-1}$, and 17.83 ± 3.81 $mg.kg^{-1}$, respectively) ($p < 0.05$). Al and Cr were the only metals that showed a higher content in plant leaves collected from unmined areas in comparison to samples from mined areas (Tab. 4).

Concerning maximum concentrations of Pb (0.60 $mg.kg^{-1}$) allowed for herbs and teas in Brazil (ANVISA 2013), these metals presented showed levels higher than recommended for consumption (Tab. 5). The maximum concentrations of Cd and Pb presented levels higher than recommended for consumption for China and the European Union (Cd: above 1 $mg.kg^{-1}$ for China and 0.05 $mg.kg^{-1}$ for European Union; Pb: above 5 $mg.kg^{-1}$ for China and 1 $mg.kg^{-1}$ for European Union) (Soliman 2016; Westman 2018) (Tab. 5). References for Al, Ba, Cr, Cu, Mn, Ni, and Zn maximum contents in teas and infusions were not found throughout the literature. Both metals with a significant difference in concentration between mined and unmined areas and metals with concentrations higher than recommended (Soliman 2016; Westman 2018) can be observed in Fig. 2. Pb presented values above the allowed limits in Brazil and European Union (above 0.60 $mg.kg^{-1}$ and 1 $mg.kg^{-1}$, respectively) (Soliman 2016; Westman 2018). Similar to the observed in soil, Mn and Pb contents were higher in leaf samples from mined areas, while leaf samples from unmined ones showed higher values of Al (Tab. 4).

Table 3. Average contents ($mg.kg^{-1}$) and standard deviation. Al, Ba, Cd, Cr, Cu, Pb, Mn and Zn in the soil of mined and unmined areas of Santa Catarina. Both P and T values of comparisons between metal concentrations in mined and unmined areas are presented, as well as total metal concentrations allowed for the state of Santa Catarina. SD: Standard Deviation; VRQ SC: Quality reference values for Santa Catarina.

Metals in soil	Unmined area Soil ($mg.kg^{-1}$)	SD	Mined area Soil ($mg.kg^{-1}$)	SD	P Value	T Value	VRQ SC ($mg.kg^{-1}$)
Al	14594.98	5132.04	16453.97	2235.53	0.03	-3.7	†
Ba	68.80	36.78	88.85	68.09	0.15	1.43	106.5‡
Cd	0.29	0.08	0.23	0.02	0.10	-1.65	0.12‡
Cr	10.46	2.98	8.18	3.48	0.01	-2.5	5§
Cu	41.63	12.11	22.13	9.91	0.03	-2.2	29§
Mn	87.92	69.49	308.22	224.96	0.03	3.0	†
Ni	18.05	5.50	13.86	4.23	0.01	-3.29	23.48‡
Pb	11.86	5.51	15.80	9.69	0.04	2.09	†
Zn	33.88	8.12	27.41	14.79	0.05	-1.93	39§

Note: The symbol (†) means not reported. (‡) Souza *et al*. (2016); (§) Hugen *et al*. (2013).

Table 4. Average contents ($mg.kg^{-1}$). Al, Ba, Cd, Cr, Cu, Pb, Mn and Zn in *B. sagittalis* leaves from unmined and mined areas of Santa Catarina. Both P and T values of comparisons between metal concentrations in mined and unmined areas are presented, SD: Standard Deviation.

Metals in plant samples	Unmined areas ($mg.kg^{-1}$)	SD	Mined areas ($mg.kg^{-1}$)	SD	P value	T value
Al	55.25	60.67	29.25	17.36	0.00	0.24
Ba	3.64	1.84	11.69	10.25	0.00	4.45
Cd	0.25	0.04	0.30	0.09	0.00	2.82
Cr	0.57	0.20	0.54	0.21	0.80	-3.6
Cu	6.18	1.79	6.45	1.45	0,52	0.64
Mn	263.87	79.58	335.8	212.60	0.05	1.88
Ni	3.55	1.24	3.99	1.11	0.15	1.44
Pb	3.80	1.02	4.00	1.64	0.56	0.57
Zn	17.83	3.81	23.34	9.51	0.00	3.209

Table 5. Average contents ($mg.kg^{-1}$). Cd, Cr, Cu, and Pb found in *B. sagittalis* leaves and their maximum recommended values in herbs consumption in Brazil, China, and the European Union.

Metals in plant samples	Sample site		Maximum contents recommended in tea/infusion consumption		
	Unmined areas ($mg.kg^{-1}$)	Mined areas ($mg.kg^{-1}$)	Brazil ($mg.kg^{-1}$)†	China ($mg.kg^{-1}$)‡	European Union ($mg.kg^{-1}$)§
Cd	0.25	0.30	0.40	1.00	0.05
Cr	0.57	0.54	--	5.00	--
Cu	6.18	6.45	--	30.00	15.00
Pb	3.80	4.00	0.60	5.00	1.00

Note: (†) ANVISA (2013); (‡) Westman (2018); (§) Soliman (2016).

In total, 195 residents were interviewed, with an average of 14 residents (± 5.4) per location. Their age ranged from 15 to 86 years old, with a mean of 53 years (± 17.8). Among the interviewees, 130 were women (68 %), and 66 were men (32 %). Of all, 105 (53.8 %), who were mainly over 50 years old (74.2 %) and women (70.4 %), claimed to consume *B. sagittalis* as tea (infusion). Within this group, 96 interviewees (91.4 %) mentioned a small consumption frequency (once a week), 1.9 % reported an average one (two-three times a week), and 6.6 % of respondents mentioned a frequent *B. sagittalis* infusion consumption (four-seven times a week or more). Intake amount varied from one to two cups for most of the interviewees (95.7 %), with the intake of one cup (200 mL) being the most mentioned (46.6 %), followed by one-two cups (200 mL each) (44.7 %). Among interviewees who declared to consume *B. sagittalis*, 8.5 % do it with the infusion of yerba mate or "chimarrão" (*Ilex paraguariensis*).

Using published information on the use of 2 g of *B. sagittalis* per infusion preparation and based on indicators (ANVISA 2013; Soliman 2016; Westman 2018), estimates of Cd, Cr, Cu, and Pb consumption per respondent were calculated (Tab. 6). Cd value was higher than the limit recommended by the European Union (< 0.10×10^{-3} mg.kg^{-1} in 2 g of vegetable dry matter) (Soliman 2016) (Tab. 6). Pb value was higher than those recommended by both Brazil (< 1.12×10^{-3} mg.kg^{-1} in 2 g of vegetable dry matter) (ANVISA 2013) and the European Union (< 2×10^{-3} mg.kg^{-1} in 2 g of vegetable dry matter) (Soliman 2016) (Tab. 6). However, all projections (Tab. 6) considered values of *B. sagittalis* dry leaves instead of its leaf infusion; therefore, depending on the method of infusion preparation, these results can vary.

Discussion

Individuals of *B. sagittalis* from both mined and unmined areas in Santa Catarina presented high contents of metals that can be toxic to human health if consumed. However,

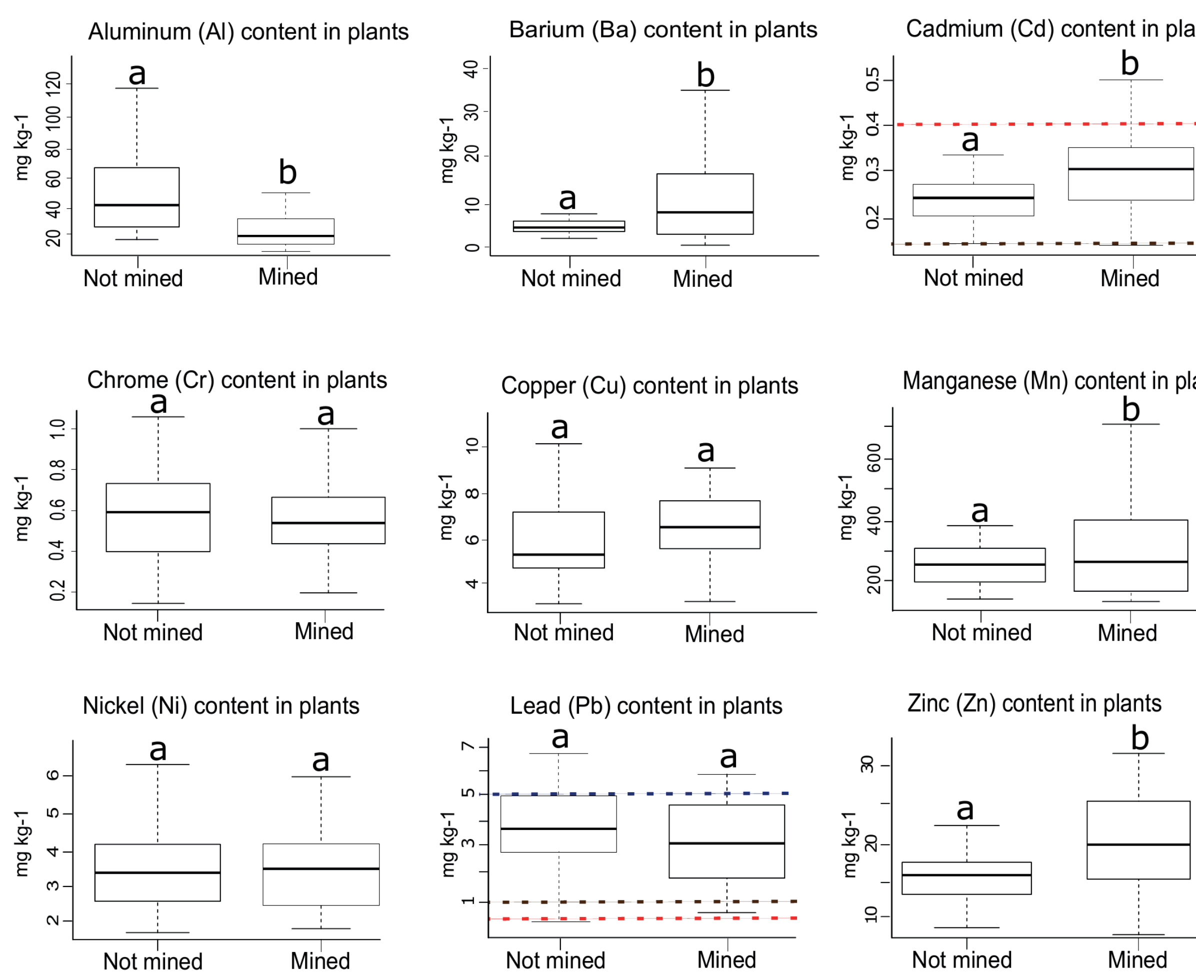

Figure 2. Boxplot of Al, Ba, Cd, Cr, Cu, Mn, Ni, Pb and Zn contents (mg. Kg^{-1}) in plants from mined and unmined areas in Santa Catarina. The red dotted line indicates the maximum reference values allowed for Cd and Pb in Brazil, according to ANVISA resolution[45] (2013); the blue dotted line indicates the maximum reference values allowed for Cd and Pb for China, according to Westman[46] resolution (2018); and the brown dotted line indicates the maximum allowable reference levels for Pb in the European Union according to the resolution of Soliman[47] (2016). Different letters indicate significant difference ($p < 0.05$) of the element concentration between plants from unmined and mined areas.

Table 6. Estimates of daily intake in mg of Cd, Cr, Cu, and Pb in 2 g of *B. sagittalis* dry matter, and acceptable metal concentration values in herbs consumption in Brazil, China, and the European Union. Based on the values in Table 5, a rule of three was used to predict the amount of intake of elements per preparation.

Metals	Acceptable metal concentration within tea			Estimates of metals daily intake within 2 g of *B. sagittalis* dry matter	
	Brazil (mg in 2 g of vegetable dry matter)†	China (mg in 2 g of vegetable dry matter)‡	European Union (mg in 2 g of vegetable dry matter)§	From unmined areas (mg in 2 g of vegetable dry matter)	From mined areas (mg in 2 g of vegetable dry matter)
Cd	0.8×10^{-3}	2×10^{-3}	0.10×10^{-3}	0.5×10^{-3}	0.6×10^{-3}
Cr	--	10×10^{-3}	--	1.14×10^{-3}	1.08×10^{-3}
Cu	--	60×10^{-3}	30×10^{-3}	12.36×10^{-3}	12.90×10^{-3}
Pb	1.2×10^{-3}	10×10^{-3}	2×10^{-3}	7.6×10^{-3}	8×10^{-3}

Note: (†) ANVISA (2013); (‡) Westman (2018); (§) Soliman (2016).

the concentration of analyzed metals differed between soil and plant samples from mined and unmined areas. Al, Mn, and Pb had higher percentages in the mined area soil, while Ba, Cd, Mn, and Zn prevailed in plant samples in the mined area. Meanwhile, Cr, Cu, Ni and Zn have higher soil concentrations in unmined areas, in the plant of these same areas only Al showed higher concentrations. According to reference values for Santa Catarina, Cd and Cr concentrations in soil were above the recommended in both mined and unmined areas (Souza *et al*. 2016) (above 0.12 $mg.kg^{-1}$ and 5 $mg.kg^{-1}$, respectively). Cu concentration in the soil was also above the recommended levels in the state, but in unmined areas only (above 29 $mg.kg^{-1}$) (Hugen *et al*. 2013). Also, in estimates considering consumption of 2 g of *B. sagittalis* dry matter, Cd and Pb, also had values above the recommended in the European Union (ANVISA 2013) (*i.e.*, Cd < 0.10×10^{-3} $mg.kg^{-1}$ (Soliman 2016) and Pb < 2×10^{-3} $mg.kg^{-1}$ (Soliman 2016)), and Pb above the recommended in the Brazil (*i.e.*, Pb < 1.2×10^{-3} $mg.kg^{-1}$ [47] (ANVISA 2013)) (Tab. 6). These figures are worrisome, as 53.8 % of respondents said they consume this species as infusion in the region.

Among the metals with elevated concentrations in the soil and *B. sagittalis* leaves, Al and Mn stood out. Aluminum results may be due to Santa Catarina's soil traits, with naturally higher availability of this element, a feature observed in other soils as well (Quitaes 2000; Echart & Cavalli-Molina 2001; Cunha 2018; Suppi *et al*. 2018). In addition, anthropogenic actions may alter the ways in which Al is available in the environment, ultimately changing plants' capacity to absorb this metal (Echart & Cavalli-Molina 2001). Such an effect could be accountable for the high concentrations of Al found in plants from unmined areas, even though levels of this metal were higher in the soil samples from mined areas (Cunha 2018; Suppi *et al*. 2018). Al consumption may be related to the development of kidney problems and early stages of Alzheimer's disease (ATSDR 2008a; Walton 2011). Still, the effects of daily Al consumption are yet to be determined (ATSDR 2008a). In the United States, estimates point that an average adult ingests around 7 to 9 mg of Al every day through food, which can be contaminated in numerous ways, such as mining activity (ATSDR 2008a; Landry 2014).

The human body is unable to use Al. On the other hand, Mn is an important element for many physiological functions in humans, such as mitochondria oxidative stress prevention, digestion, and immune response (Röllin & Nogueira 2011). However, when in elevated concentrations, it can cause several problems, including damage to the nervous system in prenatal stages and early childhood (Röllin & Nogueira 2011; Miah *et al*. 2020). Since Mn was the element with the highest concentration found in *B. sagittalis* (263.87 $mg.kg^{-1}$ in unmined areas and 335.8 $mg.kg^{-1}$ in mined ones), the people living near these locations, who are consuming this plant, are being subjected to a vulnerable health situation.

As Al, Ba is a non-essential metal for the human body and can be toxic on some occasions; however, the lack of known parameters for Ba contaminated food ingestion complicates the comprehension of its effects on human health (Lu *et al*. 2019; Pi *et al*. 2019). Studies conducted in China have shown that people who consumed rice from areas with Ba contamination presented low contamination levels (Lu *et al*. 2019). This result suggests that there might be some biochemical pathways (in plants, human beings or both) that can lower Ba concentration and reduce its possible harmful effects (Lu *et al*. 2019). Still, to answer this question, further investigation is required. For instance, can the consumption of leaf infusion from bioaccumulating plants grown in Ba tainted areas also present the same results?

In acidic and altered soils (product of mining activities, for example), Cd content and mobility rise, affecting soil microbiota functioning and enhancing its absorption by plants (Oti 2015; Feng *et al*. 2019). In the study region, soil measurements of pH ranged from 3.5 to 5.5 in mined areas and 4.5 to 6.5 in unmined ones, revealing its acidity. Indeed, high Cd concentration in soil was found (029-0.23 $mg.kg^{-1}$), as well as in *B. sagittalis* leaves (0.25-0.30 $mg.kg^{-1}$) and in consumption estimates projected considering ingestion of 2 g of *B. sagittalis* dry matter (Cd < 0.10×10^{-3} $mg.kg^{-1}$ (Soliman 2016). Such results were observed in mined and unmined areas, possibly related to soil origin in the region, with naturally higher Cd availability (Guo *et al*. 2013). However, high levels of Cd, already observed in rice and corn species consumed by the Chinese, may present a risk to food security, considering its harmful effects on human health, like nutrient absorption and kidney function alterations (Staessen *et al*.

1994; Sun *et al*. 2008; Baye & Hymete 2010; Ata-Ul-Karim *et al*. 2020). Cadmium is the most abundant metal found in coal mining areas, and its high concentration is a concern for food security (Qi *et al*. 2014; Zhang *et al*. 2015).

Levels of Cu, Ni, and Zn in unmined soil were higher than in mined soil (22.13 $mg.kg^{-1}$, 13.86 $mg.kg^{-1}$ and 27.41 $mg.kg^{-1}$, respectively). This result is possibly due to soil leaching and increased mobility in soils with a pH lower than 4.0 (Krämer & Clemens 2008; Elbana & Selim 2011). Plants use Cu, Ni, and Zn for numerous metabolic activities. For example, photosynthesis and respiration require Cu, Ni is essential for the metabolic process of plant defense, and Zn is crucial to protein binding (Yruela 2005; Krämer & Clemens 2008; Fabiano *et al*. 2015). As observed for plants, these metals also play a major role in human beings' health. Copper assists in neurological formation, while Ni is crucial for proper muscular development (Plum *et al*. 2010; Bost *et al*. 2016). However, when at high concentrations, Cu and Ni may be toxic to humans; in fact, Cu can cause kidney diseases, and Ni can induce pulmonary fibrosis (Krämer & Clemens 2008; Bost *et al*. 2016). Additionally, Zinc is an important metal for the immune system and has positive effects in cellular apoptosis control. Besides, it has a low intoxication rate and requires a high dosage to cause harm, such as digestive intoxication or stomach wall damage (Plum *et al*. 2010).

Even though projections for daily consumption were based on dry *B. sagittalis* leaves and not on its infusion, Cd and Pb presented concentrations far above the recommended levels for consumption (Cd < 0.10×10^{-3} $mg.kg^{-1}$ (Soliman 2016), Pb < 1.2×10^{-3} $mg.kg^{-1}$ (ANVISA 2013) and < 2×10^{-3} $mg.kg^{-1}$ (Soliman 2016)) and should be further investigated. Just as Cd, Cr (0.57 $mg.kg^{-1}$) has been found in high concentrations in mining areas and in food items grown in former coal mining areas, such as rice (*Oryza* spp.) in China rice plantations (Achmad & Budiawan 2017; Sun *et al*. 2018). Cr can be found in different forms: Cr(III), Cr(IV), Cr(VI), and despite its importance for lipid and protein metabolism in humans, this metal can cause severe damage to human health, increasing chances of uterine cancer development, and causing severe respiratory symptoms (ATSDR 2008b; Achmad & Budiawan 2017). Considering its ability to accumulate in the food chain, added to its high absorption by plants, Cr presence in high concentrations is concerning (ATSDR 2008b), given that estimated levels of Cd, Cr, and Pb ingestion within 2 g of *B. sagittalis* infusion were already higher than recommended (Cd < 0.10×10^{-3} $mg.kg^{-1}$ (Soliman 2016), Pb < 1.2×10^{-3} $mg.kg^{-1}$ (ANVISA 2013) and < 2×10^{-3} $mg.kg^{-1}$ (Soliman 2016)).

Along with that, reports on *Baccharis* species increasing availability in the region within the last 10-15 years also call for attention to potential growth in residents' consumption. An increase in the *Baccharis* population can be related to the fact that it is a pioneer species (Heiden 2006) that can easily develop in contaminated areas (Menezes *et al*. 2013; 2016). With rising numbers of abandoned and unrecovered coal mining areas in the Santa Catarina coal region (Rocha-Nicoleite *et al*. 2017; Blanco *et al*. 2020a), an increase in this species availability can be expected. Consumption of *B. sagittalis* was reported mainly by women (*i.e.*, 70.4 % of respondents who claimed to use this plant), which reveals women's greater vulnerability in the region. Overall, this result acknowledges the worldwide panorama that shows that women are the most affected by contaminated environments and have a higher food security vulnerability (Lutomia *et al*. 2019).

Food safety and human health risks related to food grown near mining areas are rather recent concerns; in China, the focus has been on *Oryza* spp. (rice) and *Camellia sinensis* (green tea), both of which may present bioaccumulative potential and be harmful to human health (Zhang *et al*. 2015; Huang *et al*. 2017; Liu *et al*. 2017). In India, Japan, and Europe, research regarding *C. sinensis* bioaccumulative potential has shown that it can gather high levels of heavy metals (Soliman 2016). In Canada and the United States, toxic metals from mining have been stocking up in some moose and sheep species that feed on bioaccumulating plants, being subsequently consumed by people (Schuster *et al*. 2011; Loring & Whitely 2019). Research in northern Brazil has shown that the consumption of fish contaminated by gold mining activities has caused mercury to accumulate in indigenous women's breast milk (Carvalho *et al*. 2009; Spurway & Soldatic 2016).

Since these metals are invisible contaminants (without odor, taste or physical alteration) that cannot be detected by human senses (Vyner 1988; Spurway & Soldatic 2016), their perception by human communities is challenging (Vyner 1988). The lack of parameters concerning food contamination by mining activity and the possible health effects resulting from their consumption only aggravates this difficulty (Vyner 1988). Given that metal content in the human body rises at a slow rate, requiring a long period of exposure to accumulate, relating human health issues to high metal concentrations is an arduous task (Gifford 2011; Candeias *et al*. 2019). Also, there is a considerable amount of diseases these metals can cause, including abdominal pain, headaches, and slowly developing cancers, increasing difficulties to diagnose diseases' sources (Vyner 1988; Gifford 2011; Candeias *et al*. 2019). Altogether, these complexities make it hard for people to be aware of metal contamination and food insecurity to which they are exposed.

The impact of mining activity on edible plants is evident, yet only a few studies provide data on safe ingestion rates for humans (Brisbois *et al*. 2019). A recent review regarding mining impacts on human health revealed that most research focuses on direct exposure to toxic agents in developed countries (Brisbois *et al*. 2019). Still, most mining areas are in developing or undeveloped countries, but literature is scarce on food safety, especially associated to indigenous people and local communities (Vyner 1988; Blanco *et al*. 2020a).

Baccharis is widely known and consumed in Brazil (Schripsema *et al*. 2019), but its traditional consumption is also frequently reported in Uruguay (Abad & Bermejo 2007), Argentina (Abad & Bermejo 2007), Chile (Morales *et al*. 2008), Colombia (Abad & Bermejo 2007), Mexico (Abad & Bermejo 2007), United States (Haque *et al*. 2008), and Canada (Freire *et al*. 2007). Likewise, *Baccharis* occurrence, as well as its heavy metal bioaccumulation have been reported in mining areas (Carreira 2007; Haque 2008; Oti 2015). For instance, in the United States, Pb absorption and translocation to leaves of another species of the genus *Baccharis* (*B. sarothroides*) were observed (Haque 2008). In 2020, medicinal plants use had a significant increase worldwide since the COVID-19 pandemic caused people to turn to medicinal plants more than in the former period (Nugraha *et al*. 2020).

All described situations increase food insecurity and human health vulnerability in traditional and local communities that live close to mining areas and contaminated environments. A study limitation was that, since the analyzed plants were not in a controlled environment, it was not possible to determine metals' original soil concentrations and compare them to the values absorbed by the plant. However, Cd and Pb levels (0.25-0.3 $mg.kg^{-1}$ and 3.8-4 $mg.kg^{-1}$) were above the reference levels (ANVISA 2013; Soliman 2016; Westman 2018) in plants from both mined and unmined areas. Additionally, Cd and Pb estimate consumption (Cd < 0.10×10^{-3} $mg.kg^{-1}$ (Soliman 2016), Pb < 1.2×10^{-3} $mg.kg^{-1}$ (ANVISA 2013) and < 2×10^{-3} $mg.kg^{-1}$ (Soliman 2016), are higher than recommended. Thus, *B. sagittalis* medicinal consumption in this area is not safe, as it poses a threat to food security and local communities' health.

Conclusion

Studies on food and medicinal plants contamination through mining activities are scarce and require greater attention. This study results revealed contamination in a plant of native occurrence in mined areas and in surrounding areas without mining activity, which is traditionally used as medicine. High levels of Al, Cd, Mn, and Pb were observed in *B. sagittalis*, showing that collecting this species from mined regions for consumption is not safe. It also adds concerns regarding the lack of knowledge on safe consumption levels of contaminated plants. The importance of studies like this must be highlighted, since they are crucial to provide a better understanding of mining impacts on food security and human health of populations living close to mined regions.

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