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Radioactive and stable elements' concentration in medicinal plants from Brazil

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Abstract Since the early days of mankind, plants have been used as food and for medicinal purposes. Still, little information exists in literature about the activity concentration of ^{238}U and ^{232}Th decay products, as well as stable element concentrations in Brazilian plants. Activity concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb, and chemical concentrations of As, Ba, Br, Cs, Co, Cr, Cu, Eu, Fe, Hf, La, Lu, Rb, Sb, Sc, Sm, Ta, Tb, Yb, Zn and Zr were determined in ten samples commonly used in Brazilian medicinal plants.

Keywords Medicinal plants · Radioactive elements · Stable elements

Introduction

Since the early days of mankind, plants have been used as food and medicinal source [1]. Observing animals feeding themselves, men started to taste plants and realized that they were sometimes relieved of headaches, stomachaches and muscular aches after external use of medicinal plants [2]. Generally, the studies related with therapeutic plants aim at characterizing the active component of the plant for scientific evidence of its therapeutic properties [3–5]. Therefore, little information exists in the literature about the activity concentration of natural radionuclides that belong to natural radioactive series of ^{238}U and ^{232}Th, as well as about the stable elements concentration in plants used for this purpose in Brazil. The knowledge of the elemental concentration of these elements can be useful to verify possible interferences in therapeutic activity; depending on their concentration they can also represent some threat for human being [6].

Instrumental neutron activation analysis is one of the most used methods for elemental characterization and was applied in this work to determine As, Ba, Br, Cs, Co, Cr, Cu, Eu, Fe, Hf, La, Lu, Rb, Sb, Sc, Sm, Ta, Tb, Yb, Zn and Zr concentrations [7, 8]. The major amount of Ra and Pb enters the human body via ingestion. Approximately 20% of Ra and 10–15% of the Pb ingested reaches the blood stream, is distributed for the whole body and follows the same metabolism of Ca. The objective of this work it to determine the activity concentration of ^{226}Ra, ^{228}Ra and ^{210}Pb and the elemental concentration in samples used as common medicinal plants; as well as to determine the elemental concentration in alcoholic extracts. The plants analyzed were: *Allium sativum* L., *Aloe vera*, *Portulaca oleracea* L., *Peumus boldus*, *Matricaria chamomilla* L, *Rhamnus purshiana* D.C., *Camellia sinensis* L., *Ginko biloba* L., *Panax ginseng* C. A. Meyer and *Bixa orellana* L.

Experimental

Sample preparation

Medicinal plants used in this work were obtained from drugstores and informal street commerce and are listed in Table 1, along with the part of the plant with therapeutical function. Species identification was made by comparison with specialized literature [9, 10]. Since a lot of impurities are present in most of the samples, like plant parts other

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Table 1 Herb samples analyzed, their botanical names, corresponding part that presents therapeutical functions, and their medicinal uses

Sample		Botanical name	Analyzed part of the sample	Medicinal uses
A1	Garlic	*Allium sativum*	Bulb	Atherosclerosis, high cholesterol, circulation problems and respiratory tract infections
A2	Aloe Vera	*Aloe vera*	Leaf	Topically to heal wounds and skin conditions, orally as a laxative
A3	Pigweed	*Portulaca oleracea*	Whole plant	Treatment for parasites, blood-cleanser, refresh digestive system
A4	Boldo	*Peumus boldus*	Leaf	Balance liver function; upper digestive tract disorders, intestinal worms and liver flukes
A5	Chamomile	*Matricaria chamomilla*	Flower	Anti-inflammatory, antispasmodic, antidiarrheal anxiolytic
B1	Cascara Sagrada	*Rhamnus purshiana*	Bark of the stem	Purgative and laxative
B2	Tea plant	*Camellia sinensis*	Leaf and flower	Antitoxic, diuretic, expectorant, stimulant and stomachic
B3	Maidenhair	*Ginko biloba*	Leaf	Cerebrovascular insufficiency, memory deficit, depressive emotional condition, arterial disease
B4	Ginseng	*Panax ginseng*	Leaf	Central nervous system stimulation, analgesic and anti-inflammatory
B5	Lipstick tree	*Bixa orellana*	Root	Skin diseases
C1	Ginseng[a]	*Panax ginseng*	Root	Enhancement of mental and physical capacities

[a] Industrialized sample

than those of pharmaceutical interest and a variety of other materials, the samples were cleaned with the help of a magnifying glass [11]. After impurities had been eliminated, samples were dried at constant temperature of 60 °C during seven days and grounded using ceramic mortar and pestle. For extraction procedure, samples were softened by soaking them in 70% ethanol during 7 days at room temperature.

Instrumental neutron activation analysis

The elemental determination was performed by instrumental neutron activation analysis (INAA) in samples and in extracts. Two types of samples were analyzed: bulk and extract of the samples. Approximately 5 g of the dry samples were accurately weighted and treated as described above to obtain the extract, which was dropped in filter papers. The bulk samples were also ground and accurately weighted. Both were sealed in plastic bags and in aluminum foils for neutron irradiation. Irradiated samples were counted in a hyper pure germanium detector during 5,000 s. Certified reference materials, MAG-1 (USGS) and San Joaquin Soil (NIST SRM 2709), were also irradiated together with the samples, for elemental concentration calculation.

Radiochemical separation

The radionuclide analysis was performed by radiochemical separation and total alpha and beta counting. Two grams of samples were used for extraction procedure and for bulk sample analysis. Samples were dissolved in hot nitric acid and oxygen peroxide till total elimination of organic matter. Carrier of Ba^{2+} and Pb^{2+} were added before the dissolution. The solution was treated with citric acid for Fe and Pb complexation in pH of 4.5–5.0. Sulfuric acid was added for sulfate precipitation of Ra^{2+}, co-precipitation as $Ba(Ra)SO_4$, and $PbSO_4$. The precipitate was dissolved with NTA and 6 M NaOH was added to achieve a basic medium. The addition of $(NH_4)_2SO_4$ (25 mg mL^{-1}) and glacial acetic acid precipitates $Ba(Ra)SO_4$ leaving Pb^{2+} in solution. The precipitate was separated in two steps of centrifugation and washing, dissolved with EDTA and precipitated in a Millipore filter and separated for counting.

The solution containing Pb^{2+} was treated with 1 M Na_2S to precipitate PbS. The precipitate was centrifuged, dissolved in nitric acid and filtered for sulfur separation. The addition of 30% Na_2CrO_4 precipitated $PbCrO_4$ that was filtered in Millipore filter and separated for counting. Counts were made in a gas flow proportional detector of low background, Berthold, model Lb 770, during 200 min. The procedure was from MOREIRA [12] and OLIVEIRA [13].

Results and discussion

Results for total alpha and beta measurements are shown in the Table 2 for the bulk and extract samples. In the bulk sample, lead concentration ranged from 32 to 76 mBq g^{-1}.

Table 2 Activity concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb, in Bq kg^{-1}, in the plant samples and in the extract (*)

Sample	^{226}Ra	^{226}Ra*	^{228}Ra	^{228}Ra*	^{210}Pb	^{210}Pb*
A1	<2.2	<2.2	33 ± 3	39 ± 4	51 ± 5	52 ± 5
A2	18.4 ± 0.2	<2.2	65 ± 4	42 ± 4	61 ± 6	44 ± 4
A3	4.5 ± 0.5	<2.2	44 ± 4	46 ± 5	76 ± 8	52 ± 5
A4	<2.2	<2.2	36 ± 4	37 ± 4	68 ± 7	56 ± 6
A5	<2.2	<2.2	41 ± 4	43 ± 4	52 ± 5	47 ± 5
B1	<2.2	2.3 ± 0.2	38 ± 4	33 ± 3	70 ± 7	50 ± 5
B2	13.2 ± 0.1	10 ± 1	40 ± 4	41 ± 4	73 ± 7	63 ± 6
B3	4.1 ± 0.4	<2.2	53 ± 5	55 ± 6	32 ± 3	74 ± 7
B4	5.1 ± 0.5	5,2 ± 0.5	37 ± 4	43 ± 4	58 ± 6	48 ± 5
B5	5.1 ± 0.5	3.4 ± 0.3	40 ± 4	36 ± 4	68 ± 7	57 ± 6
C1	<2.2	3.6 ± 0.4	36 ± 4	29 ± 3	35 ± 4	47 ± 5

Lead occurs naturally in plants as a result of uptake, mainly in places with high concentration due to atmospheric fall-out. Sample C1 showed the lowest concentration and was the only sample obtained in industrialized powder form. Activity concentrations of ^{226}Ra were lower than ^{228}Ra in all samples. As the former belong to ^{238}U and the last to ^{232}Th decay series, these results were in accordance with those obtained for ^{238}U, which was measured in just one sample (0.2 ± 0.1 in sample B3); the concentration of other samples was smaller than the detection limit. It can be observed that the extracts contain almost all the radium and lead contents present in the plant.

Radium in nature exists in soil, rock, surface water, groundwater, plants and animals, at low concentrations, generally lower than 37 Bq kg^{-1}. According to ANL [14] the average concentration of ^{226}Ra lies in the range of 0.4–1 Bq kg^{-1} in food; the observed values for ^{226}Ra are higher than this range for A2, A3, B2, B4 and B5. Activity concentrations of ^{228}Ra were one order of magnitude higher than those of ^{226}Ra in all samples. Average concentrations for ^{232}Th in plants [14] were of the order of 0.04 µg g^{-1}, one order of magnitude lower than those found in the studied samples, except for Lipstick tree (B5). Uranium activity concentrations were below the detection limits (0.9 Bq kg^{-1}) [15] in all samples, except for Maidenhair (B3). Committed effective equivalent doses were evaluated considering the ingestion of 5 kg per year of the plant ranged from 0.3 to 0.6 mSv year^{-1}, due to the ingestion of the analyzed samples. The higher value was obtained for Pigweed.

The difference in the concentration of elements is due to peculiarities in the absorption by different botanic structures of the plants as leaf, root, bark, flowers and the soil composition in which they were cultivated [16]. Besides that, the use of fertilizer, agricultural protections and irrigation water, climatic conditions and industrial pollution can contribute to variations in the concentrations normally observed. The concentration of elements determined by INAA in the bulk plant and in the extract is shown in Tables 3 and 4, respectively. Measurement of the standard material IAEA-336 (lichen) allowed for quality control of the results, which are seen in Table 2.

Macronutrients as K ranged from 0.8% to 3.3% for the bulk plant and from 0.06% to 0.6% for the extract; and for Na, from 0.003 to 0.03 µg g^{-1} for the bulk plant and from 0.003 to 0.01 µg g^{-1} for the extract. Potassium and iron are the most abundant elements in the analyzed plant. The use of *Camellia sinensis* and *Panax ginseng* that are rich in potassium could provide this element in deficiency cases. Micronutrients like Fe, Cr, Zn and Co are essential for the organism. Iron possesses a unique function in the metabolic process associated with hemoglobin and oxygen transport. Iron deficiency is the main nutritional deficiency in humans and is associated with insufficient diet, excessive menstruation and multiple births [3]. The need of iron for an adult is 20 mg day^{-1} and for a children, 10 mg day^{-1}. The variation of iron in the analyzed samples was from 0.003% to 0.3% for the bulk plant and from 3 to 9 µg g^{-1} for the extract. The use of *Ginkgo biloba* and *Panax ginseng* can be indicated for compensating iron deficiency.

Another essential element, Cr, acts a co-factor in the insulin synthesis and in the cholesterol and blood triglycerides control. The daily-recommended ingestion of chromium is 50–250 µg day^{-1} [17]. Amounts of Cr ranged from 0.5 to 5.5 µg g^{-1} in the bulk sample and from 0.001 to 0.1 µg g^{-1} in the extract. The analyzed drugs are not suitable to supply chromium deficiencies. Zinc concentration ranged from 7.2 to 68 µg g^{-1} in bulk plants and from 0.48 to 7.2 µg g^{-1} in extracts. This element is necessary for the cellular growth and multiplication, once it participates in the DNA and RNA synthesis and bone metabolism [16]. Low concentrations of zinc are related to infertility. High Zn concentration was observed in tea plant (68 µg g^{-1}), which is indicated in cases of zinc deficiency.

Cobalt is essential for the B12 vitamin and the thyroid metabolism; the daily ingestion must be around 3 µg [16]. The Co concentration varied from 0.085 to 1.16 µg g^{-1} in bulk plants and from 0.01 a 0.26 µg g^{-1} in the extract. Only in the tea plant (B2), it was observed a measurable amount of Se (0.3 µg g^{-1}) that is an essential element involved in antitumoral, immune and inflammatory metabolism and is used in degenerative disease treatment and neurologic disturbs [17]. It is well known that several elements are essential for human beings, although at high levels they can be toxic. More research is required on metal concentrations in medicinal plants used for therapeutic purposes, in order to take more advantage of their benefits and to prevent intoxications.

Table 3 Elemental concentration in μg g^{-1}, or otherwise indicated (%), of elements determined by neutron activation analysis

	Ba	Br	Ce	Co	Cr	Cs	K (%)
A1		3.2 ± 0.1					1 ± 0.2
A2	126 ± 32	46.2 ± 0.9	1 ± 0.3		1.8 ± 0.4		
A3	123 ± 32	72 ± 1	5.5 ± 0.4	0.3 ± 0.03	2.5 ± 0.4	0.25 ± 0.05	1 ± 0.7
A4	43 ± 14	25.8 ± 0.4	1.8 ± 0.2	0.11 ± 0.01			1.1 ± 0.2
A5	36 ± 5	24 ± 0.2	1.2 ± 0.2	0.18 ± 0.02	0.5 ± 0.2		0.7 ± 0.1
B1			3 ± 0.2	0.09 ± 0.01	4.7 ± 0.3	0.25 ± 0.02	0.8 ± 0.1
B2	105 ± 19		5.8 ± 0.4	0.47 ± 0.02	2.4 ± 0.4	0.29 ± 0.04	3.3 ± 0.6
B3	129 ± 25		5.3 ± 0.4	0.83 ± 0.04	5.5 ± 0.5	0.41 ± 0.05	1.4 ± 0.3
B4	159 ± 17	23 ± 0.2	11.5 ± 0.7	1.16 ± 0.05	4.2 ± 0.4	0.25 ± 0.05	1.6 ± 0.4
B5	119 ± 9	3.3 ± 0.1	1.2 ± 0.1	0.1 ± 0.01	1.1 ± 0.1		0.9 ± 0.1
C1	56 ± 5	23.3 ± 0.2	2.5 ± 0.2	0.81 ± 0.03	1.2 ± 0.2		1.9 ± 0.2
Md		14 ± 1	1.02 ± 0.3	0.25 ± 0.03	1.77 ± 0.4	0.09 ± 0.02	
VM	6.4	12.9	1.28	0.29	1.06		0.184

	Fe (%)	Eu	Sc	Sm	Na	Lu
A1	0.0030 ± 0.0003		0.0018 ± 0.0001		0.030 ± 0.001	
A2	0.030 ± 0.002	0.024 ± 0.007	0.080 ± 0.004	0.036 ± 0.007	0.006 ± 0.001	
A3	0.071 ± 0.003	0.072 ± 0.008	0.242 ± 0.009	0.07 ± 0.02	0.026 ± 0.002	
A4	0.034 ± 0.002	0.11 ± 0.01	0.11 ± 0.01	0.08 ± 0.01	0.029 ± 0.001	0.019 ± 0.004
A5	0.034 ± 0.002	0.066 ± 0.006	0.113 ± 0.004	0.21 ± 0.01	0.028 ± 0.001	0.009 ± 0.003
B1	0.034 ± 0.001	0.027 ± 0.003	0.069 ± 0.003	0.015 ± 0.002	0.003 ± 0.001	
B2	0.056 ± 0.002	0.088 ± 0.006	0.105 ± 0.004	0.35 ± 0.02	0.016 ± 0.001	0.004 ± 0.002
B3	0.220 ± 0.004	0.094 ± 0.007	0.72 ± 0.02	0.35 ± 0.02	0.09 ± 0.003	0.027 ± 0.004
B4	0.29 ± 0.01	0.19 ± 0.01	0.55 ± 0.02	0.92 ± 0.05	0.016 ± 0.002	0.020 ± 0.004
B5	0.012 ± 0.001	0.018 ± 0.003	0.026 ± 0.001	0.048 ± 0.003	0.003 ± 0.001	
C1	0.082 ± 0.003	0.079 ± 0.005	0.189 ± 0.006	0.25 ± 0.01	0.003 ± 0.001	0.006 ± 0.002
Md	0.042 ± 0.002	0.025 ± 0.01	0.17 ± 0.02	0.11 ± 0.02	0.032 ± 0.003	0.006 ± 0.002
VM	0.043	0.023	0.17	0.11	0.032	0.007

	Rb	Sb	Hf	La	Th	Yb	Zn
A1	2.0 ± 0.2			0.02 ± 0.01			2.8 ± 0.2
A2		0.11 ± 0.03		0.34 ± 0.05			18 ± 1
A3	49 ± 4		0.20 ± 0.03	4.6 ± 0.2	0.26 ± 0.04		48 ± 3
A4	11 ± 1			1.2 ± 0.1			11 ± 1
A5	9.8 ± 0.7	0.02 ± 0.01		1.29 ± 0.05	0.12 ± 0.02	0.23 ± 0.04	11 ± 1
B1	49 ± 4	0.03 ± 0.01	0.06 ± 0.01	0.19 ± 0.02	0.16 ± 0.02		7.2 ± 0.5
B2	22 ± 1	0.11 ± 0.01	0.11 ± 0.03	5.0 ± 0.3	0.4 ± 0.04	0.21 ± 0.04	68 ± 3
B3	85 ± 4	0.25 ± 0.02	0.35 ± 0.03	2.5 ± 0.2	0.92 ± 0.08		14 ± 1
B4	31 ± 2	0.07 ± 0.02	0.77 ± 0.05	8.5 ± 0.5	0.92 ± 0.06	0.17 ± 0.04	31 ± 2
B5	4.8 ± 0.3			0.56 ± 0.03	0.05 ± 0.01		10 ± 1
C1	46 ± 2	0.02 ± 0.01	0.38 ± 0.02	2.9 ± 0.1	0.15 ± 0.02	0.07 ± 0.02	18 ± 1
Md	1.75 ± 0.2	0.09 ± 0.05	0.04 ± 0.01	0.62 ± 0.09	0.14 ± 0.04	0.08 ± 0.02	25 ± 5
VM	1.76	0.07		0.66	0.14	0.037	30.4

Blank indicates values not measured

Md, Mean value obtained in the measurement of standard reference material IAEA-336, n = 4

VM, Certificated values for the standard reference material IAEA-336

Table 4 Elemental concentration in ng g^{-1}, or otherwise indicated (% or μg g^{-1}) (*), in the extract samples

	Br	Co	Cr	Cs	Fe*	K (%)	La
EA1	160 ± 10	18 ± 1			9.3 ± 0.6	0.6 ± 0.1	6 ± 1
EA2	320 ± 5	9 ± 1			5.3 ± 0.4	0.06 ± 0.01	5.2 ± 0.7
EA3	730 ± 20	28 ± 2	230 ± 20	24 ± 3	3.3 ± 0.5	0.30 ± 0.04	7.2 ± 0.5
EA4	510 ± 10	23 ± 1			3.5 ± 0.5	0.19 ± 0.03	2.9 ± 0.6
EA5	330 ± 10	49 ± 4	570 ± 30	17 ± 2	4.8 ± 0.4	0.6 ± 0.1	3.7 ± 0.5
EB1	61 ± 3	10 ± 1	410 ± 20	60 ± 4	4.4 ± 0.4	0.27 ± 0.06	3.7 ± 0.4
EB2	53 ± 4	44 ± 2	360 ± 20	30 ± 2	3.1 ± 0.3	0.6 ± 0.1	3.0 ± 0.4
EB3	230 ± 10	33 ± 3		3 ± 2	5.4 ± 0.4	0.22 ± 0.05	2.5 ± 0.4
EB4	400 ± 10	48 ± 2	300 ± 20	24 ± 2	6.3 ± 0.5	0.29 ± 0.06	4.3 ± 0.5
EB5	1.61 ± 10		37 ± 2	8 ± 2	4.0 ± 0.1	0.6 ± 0.1	11 ± 1
EC1	1.61 ± 10	260 ± 10	80 ± 20	6 ± 2	3.9 ± 0.6	0.6 ± 0.1	11 ± 1

	Na	Rb*	Sb	Sc	Sm	Zn*
EA1	10.40 ± 0.02	1.5 ± 0.1	2.8 ± 0.4			7.2 ± 0.4
EA2	4.16 ± 0.01	0.63 ± 0.05	1.9 ± 0.3	0.33 ± 0.06		0.7 ± 0.1
EA3	3.34 ± 0.01	6.0 ± 0.4	1.8 ± 0.3	0.57 ± 0.09	0.34 ± 0.06	1.5 ± 0.1
EA4	7.10 ± 0.02	1.5 ± 0.1	1.7 ± 0.2		0.7 ± 0.1	1.4 ± 0.1
EA5	7.60 ± 0.02	18.9 ± 0.7	5.9 ± 0.9	0.32 ± 0.07		2.2 ± 0.1
EB1	6.40 ± 0.01	6.9 ± 0.3	3.8 ± 0.5	0.50 ± 0.05	0.24 ± 0.08	0.74 ± 0.03
EB2	4.40 ± 0.01	9.9 ± 0.4	2.8 ± 0.5	0.43 ± 0.07	0.5 ± 0.1	1.8 ± 0.1
EB3	8.70 ± 0.02	1.15 ± 0.06	3.9 ± 0.8	0.27 ± 0.06	0.6 ± 0.2	0.56 ± 0.04
EB4	6.10 ± 0.02	4.9 ± 0.3	4 ± 1	0.67 ± 0.09		0.48 ± 0.03
EB5	6.30 ± 0.02	2.0 ± 0.1		0.4 ± 0.1		1.32 ± 0.06
EC1	6.30 ± 0.02	15.9 ± 0.7	0.40 ± 0.01		6.3 ± 0.2	1.57 ± 0.07

Blank indicates values not measured

Conclusions

The concentrations of major, minor and trace elements were determined in ten usual medicinal plants and in alcoholic extracts by INAA and total alpha and beta counting. The elemental concentrations varied in a wide range, both for bulk plant and extract. It can be highlighted the use of *Ginkgo biloba* and *Panax ginseng* as a source of iron; high Zn concentration in tea plant could be used for zinc deficiency. Only in tea plant, it was observed measurable amount of Se, an essential element. Also, the results showed that the day-by-day use of these plants could greatly contribute for the elemental needs of the body. Nevertheless, it must be noted that although a great part of the world population uses medicinal plants, their use should be accompanied by the assurance of their quality, efficacy and safety.

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