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Characterization of a group of medicinal herbs commercialized in Rio de Janeiro state through XRF spectroscopy

Caracterização de um grupo de ervas medicinais comercializadas no estado do Rio de Janeiro através de espectroscopia por XRF

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Abstract
The use of medicinal plants by the Brazilian population is a traditional practice, and is often the only resource used in basic health care. Within the context of the new instrumental analytical techniques, X-ray fluorescence spectroscopy stands out, being used mainly for areas where the rapid profile of constituents is indispensable. The medicinal plants used in this study were: Brazilian arnica (***Solidago microglossa*** D. C.), chamomile (***Chamomilla recutita*** (L.) Rauschert), Chilean boldo (***Peumus boldus*** Molina), lemongrass (***Cymbopogon citratus***) and Brazilian ginseng (***Pfaffia Paniculata*** (Martius) Kuntze). This work aims to qualitatively analyze, through X ray fluorescence, the elemental composition of selected medicinal plants that are commonly marketed in the state of Rio de Janeiro. By means of the generated energy spectra, Ca, K, Fe,

Zn, Ni, among others, were observed. The Chilean boldo, camomila and arnica samples presented a large number of photon counts for Ca and K, while the lemongrass showed a high number of photons for Fe and Ni. However, one of Chilean boldo samples was highlighted as worrying, due to peak in the spectrum, referring to energy that corresponds to a heavy metal, lead (Pb).

Keywords: Fluorescence. X Rays. Phytotherapy.

Resumo

O uso de plantas medicinais pela população brasileira é uma prática tradicional, sendo muitas vezes o único recurso utilizado na atenção básica de saúde. No contexto das novas técnicas analíticas instrumentais, a espectroscopia por fluorescência de raios X se destaca, sendo utilizada principalmente para áreas em que a obtenção de rápido perfil de constituintes é indispensável. As amostras incluíram plantas medicinais de uso comum, a saber: arnica brasileira (***Solidago microglossa*** D. C.), camomila (***Chamomilla recutita*** (L.) Rauschert), boldo do Chile (***Peumus boldus*** Molina), capim cidreira (***Cymbopogon citratus*** D. C. (Stapf)) e ginseng brasileiro (***Pfaffia Paniculata*** (Martius) Kuntze). Este trabalho tem como objetivo analisar qualitativamente, por meio de fluorescência de raios X, a composição elementar das plantas medicinais selecionadas que são comumente comercializadas no Estado do Rio de Janeiro. Por meio dos espectros de energia gerados, foram observados Ca, K, Fe, Zn, Ni, entre outros. As amostras de boldo do Chile, camomila e arnica apresentaram grande número de contagens de fótons para Ca e K; já o capim cidreira apresentou elevado número de fótons para Fe e Ni. No entanto, uma das amostras de boldo do Chile se destacou como preocupante, devido a um pico, no espectro, referente à energia que corresponde ao metal pesado, chumbo (Pb).

Palavras-chave: Fluorescência. Raios X. Fitoterapia.

INTRODUCTION

Since the earliest days, humans seek in nature ways to improve their living conditions. Firstly, they noticed that plants could be used as foods; over the years, they began to use plants also for healing purposes in natural medicine, administered in the form of teas, tincture, ointment, powder, drops, tablets, bringing benefits to the body and recovering it from illnesses.[1]

The World Health Organization (WHO) defines medicinal plant as every and any vegetable that has in one or more organs substances that can be used for therapeutic purposes or are precursors of semi-synthetic drugs.[2]

The use of medicinal plants by the Brazilian population is a traditional practice,[3,4] many times being the only resource in primary health care.[2] The National Policy of Integrative and Complementary Practices of the Unified Health Service (in Portuguese, ***Política Nacional de Práticas Integrativas e Complementares no Sistema Único de Saúde – SUS***) accepts the premise that common knowledge about medicinal plants should not be underestimated. It also warns that such knowledge should only be passed on after confirmation of the alleged properties and a safe use of the plants.[5]

Medicinal herbs are usually sold in pharmacies and natural product stores, where plant preparations receive an industrial label. In general, these preparations do not possess a quality certificate and are produced from cultivated plants, altering the character of traditional medicine, which uses native plants.[2] Most of these plants are rich in metals that may or may not be beneficial to health, some of them more utilized by industries and, for this reason, are more investigated from the toxicological point of view.[6]

In the context of new instrumental analytical techniques, X-ray fluorescence (XRF) spectroscopy is an outstanding technology, especially for areas that require a rapid profile of metal and non-mental constituents.[7] XRF is based on the characteristic X-rays, and by identifying and measuring them, the chemical element of its origin is identified.[8]

This study aims at analyzing qualitatively and identifying, within the detection limits of the equipment, elements ranging from sodium (Na) to uranium (U), in the following herbs: ***Solidago microglossa*** D.C (Brazilian arnica), ***Chamomilla recutita*** (L.) Rauschert (chamomile), ***Peumus boldus*** Molina (Chilean Boldo), ***Cymbopogon citratus*** (D.C) Stapf (lemongrass) and Brazilian ginseng (***Pfaffia Paniculata*** (Martius) Kuntze), marketed in Rio de Janeiro. Thus, the aim is to provide more information to consumers and the society in general about the chemical composition of these plants, by means of scientific arguments, within the detection limit of the equipment.

METHODOLOGY

To choose the samples, we conducted a literature survey on scientific databases (Biblioteca Virtual em Saúde, PubMed/Medline, Scielo, Periódico Capes, Web of Science, Scholar), in the period of August 2014 to March 2015, with readings of publications on medicinal herbs often used for treatment of the most common diseases. We verified the diversity of these plants available in the marketplace. They are:

Brazilian Arnica (***Solidago Microglossa*** D. C.): it is commonly known as arnica, Brazilian arnica, mountain arnica, and in Brazil also as ***erva-lanceta***, ***arnica silvestre***, ***espiga de ouro***, ***lanceta***, ***macela miúda***, ***marcela miúda***, ***rabo de rojão***, ***sapé macho***.[9,10] In Brazilian popular medicine, it has been used as a diuretic, also having anti-inflammatory, analgesic, stomachic, astringent, healing and vulnerary properties.[11] Topically, it is used for the treatment of wounds, injuries, traumas and bruises.[10] In its chemical composition, in the plant shoots, it is found quercetin, which is a glycosidic flavonoid, in addition to tannins, saponins, resins and essential oil. In the roots, we find diterpenes such as inulin and rutin, quinic acid, rhamnosides, as well as caffeic, chlorogenic, hydrocinnamic acids and derivatives.[11] There are few studies on the toxicity of this plant, which indicates the need for carrying out toxicological assays to determine whether its use is safe.[12]

Chamomile (***Chamomilla recutita*** (L.) Rauschert): it is the medicinal plant most used in the world, largely consumed in daily life as a tea. It has sedative, anti-inflammatory and analgesic effects, helps ease stomach and abdominal cramps, and stimulates menstruation. It is also widely used in cosmetics. The scientific name of chamomile is ***Chamomilla recutita*** (L.) Rauschert, and it is distinguished by the pharmacological properties of its flower, especially those related to the chemical constituents contained in its essential oil, such as anti-inflammatory and sedative activities.[13-16] The chemical composition of the essential oil of the flowers, which are the most used part of the plant, consists of chamazulene, alpha-bisabolol, choline, flavonoid, coumarin, and mineral salts.[17]

Boldo (***Peumus boldus*** Molina): it is commonly found in gardens of Brazilian houses and is mainly used as a tea for stomach and liver problems. There are diverse kinds of boldos: common boldo or Indian coleus, Coleus forskohlii (***Plectranthus barbatus***), blackroot and, in Brazil also as ***boldo-do-campo*** or ***doce-amargo-do-campo*** (***Pterocaulon polystachium***), Chilean boldo (***Peumus boldus***). The ***Peumus boldus*** (boldo) trees contain essential oils (ascaridole, cineole, esters, aldehydes, ketones and hydrocarbons), alkaloids (boldine, isoboldine and others), glycosides and others (flavonoids, citric acid, gum, sugars, tannins, minerals, lipids, etc.). The outer covering of the stem is richer in alkaloids. Concentrations of essential oils and alkaloids vary according to the region where it is grown, and in some regions its cultivation is not viable.[18] The

Brazilian plants do not produce sufficient active principles (boldine) to justify their therapeutic use, and the part of the plant that is used are leaves [18]

Lemongrass (***Cymbopogon citratus*** (D.C) Stapf): it is also known in Brazil as ***capim-limão***, ***capim-santo*** or ***capim-cidrão***.[19] It has pharmacological activities for various disorders, such as insomnia, nervousness, maldigestion, flatulence, as well as antispasmodic of uterine and intestinal tissues, diaphoretic, antipyretic, diuretic, antiallergic and analgesic properties.[20,21] The plant also has reported insecticide properties, especially larvicidal and insect repellent. Its main component is citral (essential oil), a mixture of neral (cis-citral) and geranial (trans- citral) isomers. Studies have reported that the leaves have sedative, central nervous system depressant, analgesic, antimicrobial and antifungal activities.[22-25]

Brazilian Ginseng (***Pfaffia Paniculata*** (Martius) KuntzeJ: it belongs to the family ***Amarathanceae*** and has three species more widely known, ***Pfaffia glomerata*** (Spreng) Pedersen, ***Pfaffia iresinoides*** (Sprengel) and ***Pfaffia paniculata*** (Brazilian ginseng). The part of the plant that is used is the root, and the most marketed species is Pfaffia paniculata, because besides being Brazilian it is usually confused with other type of ginseng, the imported one. The constituents of the active principle of Pfaffia paniculata are pfaffic acid (noriterpenoid); saponins: pfafosides A, B, C, D, E, F, which have antitumor potential; allantoin, sitosterol and stigmasterol phytosterols, natural salts such as phosphorus, calcium, iron and potassium; amino acids; mucilage indicated for treatment of the blood circulatory system, stress, anemia, diabetes and asthenia. Some studies confirm the activity of pfaffic acid and pfafosides in inhibiting the growth of cultured tumor cells, such as melanoma B-16. It stimulates and tones the body, eliminating physical and mental fatigue, alleviating stress and depression. Its pharmacological action contributes to strengthening the heart, improving the circulatory process and increasing the number of red cells and the level of hemoglobin. It also has hypoglycemic and insulin production actions.

The samples were obtained at different neighborhoods in Rio de Janeiro capital (Campo Grande, Bangu, Tijuca, Barra da Tijuca, Santa Cruz), and nearby cities (Niterói and Paracambi). They were collected following the criterion of not repeating the same suppliers, and then were prepared at the ***Laboratório de Farmacotécnica***, ***Faculdade Bezerra de Araújo*** (Pharmacotechnics Laboratory, Bezerra de Araújo Faculty), Rio de Janeiro-RJ.

Firstly, they were oven-dehydrated and then carefully ground and crushed. After preparation of each sample, the entire equipment was thoroughly cleaned and decontaminated to prevent any residue to contaminate the next sample. Once the sample was in powder, it was taken to the ***Laboratório de Instrumentação e Simulação Computacional Científica Aplicada*** (LISComp, ***Instituto Federal de Educação Ciência e Tecnologia*** (IFRJ) (Laboratory of Instrumentation and Applied Scientific Computer Simulation, Federal Institute of

Science and Technology Education) where three pellets of each sample were produced. A compactor and a hydraulic press (*SPECAC*) were used. Each sample was subjected to eight tons of pressure during three minutes.

An X-ray fluorescence spectroscopy equipment (Bruker Tracer IV SD), installed at LIS-Comp/IFRJ, in Paracambi, was used to identify the chemical elements present in the samples. This analysis enables to obtain information on the chemical elements of the sample by means of detection of characteristic X-rays emitted by the same.

Each sample was placed on the top of the equipment; then, a dome was used to serve as a shield against occupational exposure from the secondary X-rays beam. The radiographic parameters were then dosed during 300 seconds, with 40 kV and 10 mA, with the samples being irradiated at three different points. The energy (in kg electron-volt, keV) and counts (in photons per second) were then obtained.

Once data were obtained, the average of the three points analyzed in each sample was calculated to build the spectrum. For the spectral analysis, the ***OriginPro software, version 8***. was used. Then, each element (Si, Cl, K, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, As, Pb,0 Kr,) was defined by letters of spectral lines (K*, Kp, La and Lp) provided by the equipment.

RESULTS AND DISCUSSION

The figures below show the XRF spectra corresponding to each sample analyzed.

Figure 1. Energy spectrum of a sample of arnica (*Solidago microglossa* D.C)

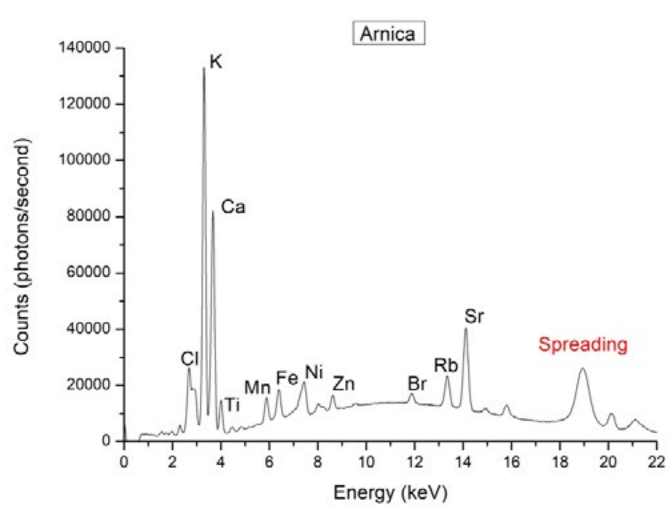

Figure 2. Energy spectrum of a sample of boldo (*Peumus boldus* Molina)

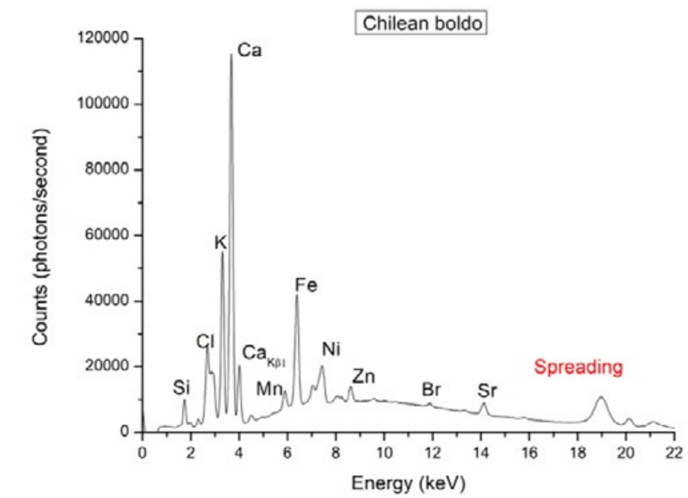

Figure 3. Energy spectrum of a sample of chamomile (*Chamomilla recutita* (L.) Rauschert)

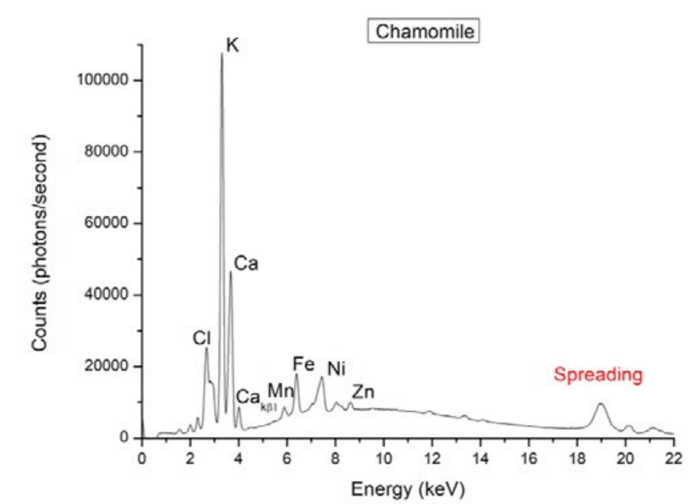

Figure 4. Energy spectrum of a sample of lemongrass (*Cymbopogon citratus* (D.C) Stapf)

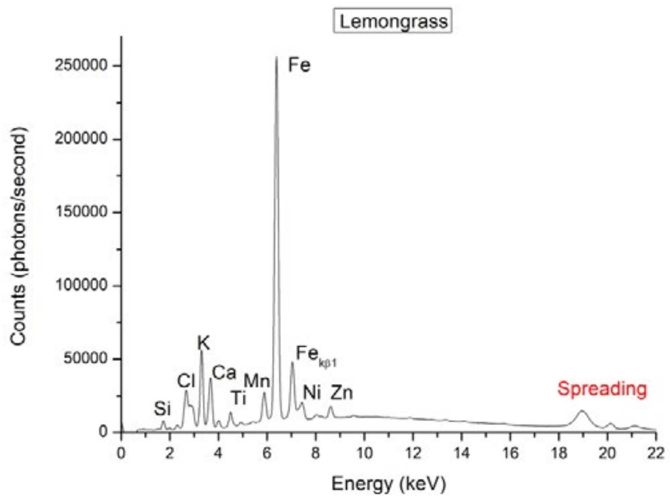

Figure 5. Energy spectrum of a worrying sample of boldo (*Peumus boldus* Molina) collected in the city of Rio de Janeiro

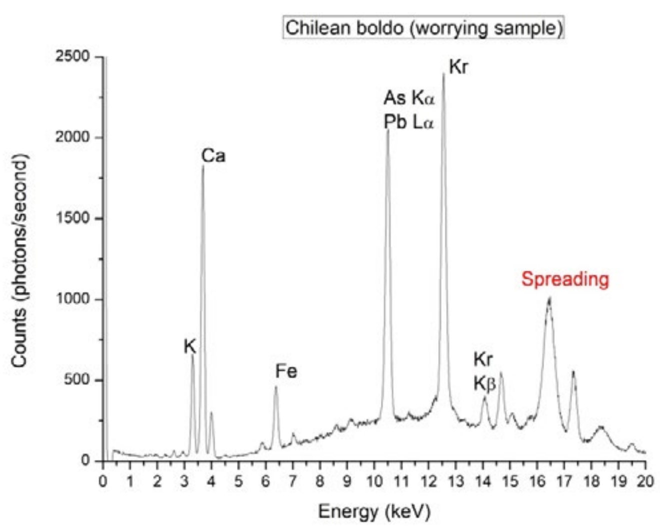

Figure 6. Energy spectrum of a sample of Brazilian ginseng

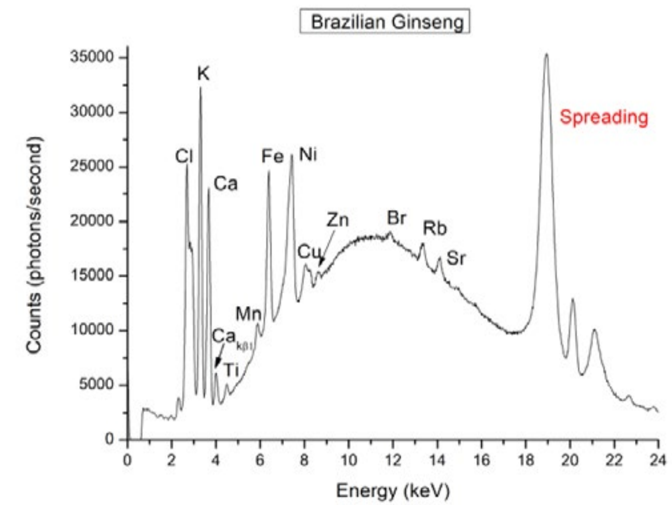

Figure 5 represents the XRF energy spectrum of one of the samples collected at one of the stores of Rio de Janeiro metropolitan region. This sample was of very high concern due to the peak of energy indicating a heavy metal, lead (Pb). This sample was prepared with caution in order to avoid contamination and irradiated again under the same conditions. It was then irradiated under different conditions, i.e., changing the technique (kV, mA and time). Yet, the peak relating to energy of 10.51 keV was observed again.

According to the spectral lines, this energy is very close to Pb on line La (10.54 keV). However, the Ka line of arsenic (As) also refers to the same energy value, i.e., 10.54. Confirmation of the element would be achieved by lines Kp and Lp,, but these spectra were overlapped and their identification was unavailable.

The sample was irradiated in other equipment, using other analytical technique, the electronic microarray method with energy dispersive spectroscopy (EMA/EDS)[26] of IFRJ/Paracambi, and the same spectral lines were identified. Considering that other pellets of the same sample were prepared at different locations and by different personnel, but following the same preparation conditions, the hypothesis of contamination during the preparation and handling process was disregarded. Soil is a very important factor in metals absorption,[27] since plants may transfer to the food chain the metals that accumulate in all plant tissues.[28]

Table 1 shows the elements found in each analyzed sample.

Table 1. Analyzed samples of medicinal plants with their respective chemical elements found within the detection limit of the equipment. Rio de Janeiro/2015.

Samples	Elements
Arnica	Cl, K, Ca, Ti, Mn, Fe, Ni, Zn, Br, Rb, Sr
Chamomile	Cl, K, Ca, Mn, Fe, Ni, Zn
Boldo	Si, Cl, K, Ca, Mn, Fe, Ni, Zn, Br, Sr
Lemongrass	Si, Cl, K, Ca, Ti, Mn, Fe, Ni, Zn
Ginseng	Cl, K, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr
Boldo*	K, Ca, Fe, As, Pb, Kr

*worrying sample

Calcium was found in all samples, particularly in Chilean boldo and arnica, an expected result considering that medicinal plants transfer to the food chain essential elements that accumulate in their tissues, providing benefits to human health. However, although essential, these elements must be balanced in the body so not to be harmful to health.

Calcium may be a great ally for individuals who have low levels of this element in the body and an option for those with lactose intolerance and need to replace this milk element. But in individuals with high levels of calcium, this element may cause kidney stones.

In the worrying Chilean boldo sample, the XRF analytical method detected the presence of heavy metal (Pb) in its composition, showing the efficiency and quickness of this technique, as well as possible utilization in the quality control of this kind of food.

It was found that all samples have in their composition elements that play a major role in the human body, such as Ca, Fe, Zn, etc.

CONCLUSION

The present study achieved its goal of identifying, by means of an accurate, rapid and reliable technique, the chemical composition of herbs and medicinal plants commonly consumed. Using XRF, it was possible to verify a variety of elements, including the presence of heavy metal (Pb) in the worrying sample, which was confirmed by EMA.

As the analysis was qualitative, identifying only the chemical element and not its concentration, it cannot be affirmed that the detected Pb is or is not within the maximum limit allowed by legislation for teas (0.60 mg/kg), according to Resolution RDC no. 42, of August 29, 2013.[29]

Thus, this study was limited to identifying the chemical element only, and so it is suggested complementary studies designed to conduct quantitative analysis to obtain the respective concentrations.

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Contributors

Luiz LC worked in every stage of the study, from conception to final revision. Costa AS participated in the preparation of the sample and data analysis and interpretation. Demarque DDM participated in the preparation of the samples and data analysis and interpretation. Batista RT participated in the conduction of measures and data analysis; and Freitas, RP, of data analysis an interpretation.

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