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Toxicological Aspect of Some Selected Medicinal Plant Samples Collected from Djelfa, Algeria Region

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Abstract

Instrumental neutron activation analysis (INAA) has been used to determine the concentration of some toxic chemical elements in a variety of aromatic plants samples collected from Djelfa region. In the present work, eight medicinal plants were examined, such as *Artemisia herba-alba* Asso., *Artemisia compestris* L., *Laurus nobilis* L., *Origanum vulgare* L., *Mentha spicata* L., *Rosmarinus officinalis* L., *Mentha pulegium* L., and *Pistacia lentiscus* L. The levels of toxic elements were compared to their daily total intake; Arsenic was present in all plant species examined, with a concentration ranging from 0.18 to 5.44 μg g^{-1}. Bromine was also detected in all the medicinal plant species, with high concentrations, compared to arsenic except in the case of *Laurus nobilis* that has the highest concentration of arsenic. Cerium, cobalt, chromium, and antimony were presented in all plant species. The exactitude of the results was assessed by analyzing the certified reference material of SRM-NIST 1573a and CRM GB07605 (GSV4). These data analysis for this medicinal plant can be useful for therapeutics and pharmaceutical purposes.

Keywords Toxic elements · Arsenic · Bromine · Antimony · INAA method · Medicinal plants · Daily total intake

Introduction

According to the World Health Organization report, there is an estimated 65 to 80% of the world's population who use traditional medicine for their primary health [1]. However, the use of herbal medicines has come under scrutiny due to their perceived long-term toxicity among other considerations; in other words, there is usually a small window between nutrition and toxicity [2–4]. The causes of the toxicities in medicinal plants are their individual properties: concentrations of metals in soil, air, and water; climatic factors; plant species; and other environmental factors [5, 6].

Monitoring the content of potentially toxic elements is one of the most important aspects of controlling food safety, in order to establish tolerable intakes of different contaminants that exhibit thresholds of toxicity [7–9]. The most often used term regarding toxic element intake is tolerable daily intakes (TDIs) allocated by the Joint FAO/WHO Expert Committee on Food Additives (JECFA); the TDIs are the levels of intake of toxic elements that on the basis of scientific knowledge, they are refereed by the food and nutrition international scientific committees, to maintain the body safe [10, 11].

There are many analytical techniques for determining trace element concentration in different biological matrices like particle induced X-ray emission, inductively coupled plasma mass spectrometry, atomic absorption spectrometry, and instrumental neutron activation analysis [12–14]. The signification of quality control and assurance (QC/QA) to internal and external validation is an indispensable step in the analytical procedure; in this context, it requires that all steps must be checked. The INAA technique has been the most used in biological samples due to its multi-elemental character and high sensitivity [15–18].

The interest of most research was paid to study the organic part of the medicinal plants, including antioxidant and antibacterial, while not much study attention was focused on their elemental contents [19–28], despite the importance of toxic chemical elements, which contribute to the valuable properties, chemical, biological, or nutritional, of medicinal plants [29–33]. Nowadays, there is an increasing interest back to nature and medicinal

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plants; rural communities rely on herbal remedies and traditional recipes for treating diseases [21, 34].

The aim of the present work was to contribute by using nuclear analytical techniques, to determine concentrations of some toxic chemical elements (As, Br, Ce, Co, Cr and Sb) in eight aromatic plants that are largely used in traditional Algerian medicine such as *Artemisia herba-alba* Asso., *Artemisia compestris* L., *Laurus nobilis* L., *Origanum vulgare* L., *Mentha spicata* L., *Rosmarinus officinalis* L., *Mentha pulegium* L., and *Pistacia lentiscus* L. Our selection of plants studied for this work corresponds perfectly to the scientific needs because these samples are widely used by the Algerian population in their health treatments. The results of the present work may be useful and will be used as a database for the researchers and specialists.

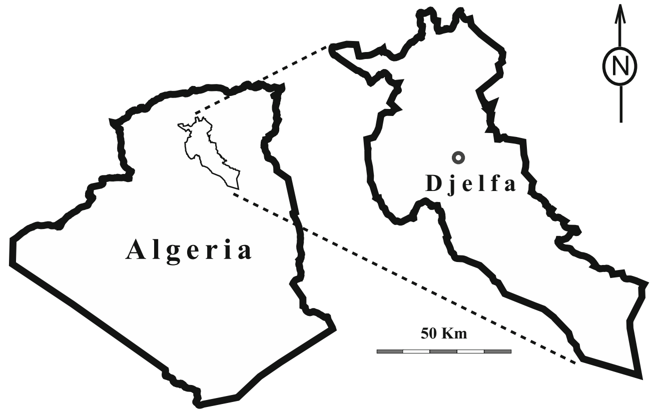

Fig. 1 Map of Algeria with the sampling localization of Djelfa province

Materials and Methods

Plants Collection and Sample Preparation

The eight studied taxa (Table 1) were collected during the year 2012 from the full steppe zone of Algeria, Djelfa province, distanced from Algiers by 300 km (Fig. 1); the plant samples were prepared at the radiochemical laboratory. At first, the samples were washed many times with deionized water to remove soil particles, then dried for 3 weeks at room temperature. Next, the samples were ground to a fine powder; three samples of each plant weighing about 120 mg were stored in precleaned polyethylene capped bottles. In this work, the standard reference materials were used to determine the toxic elemental concentration, which is NIST-SRM 1573a (tomato leaves) come from the National Institute of Standard and Technology (NIST) and Chinese tea leaves (GBW 07605, National Research Center for CRM, Lang fang, China). Finally, all samples and the standards were placed in an aluminum irradiation capsule.

Irradiation and Counting

All samples and the standards were placed in an aluminum irradiation capsule, which was irradiated for 6 h at a thermal neutron flux of 4.5×10^{13} cm^{-2} s^{-1}. After a decay of 2 days, the irradiated samples together with the standard were measured at different decay times using a coaxial HPGe detector, Canberra Genie 2000 gamma-spectrometric system [28], the system has a resolution of 1.8 keV for 1332.5 keV gamma-peak of ^{60}Co. The first cooling time for middle-lived radionuclides was in the range of 2–5 days and counted about 7200 s for each sample and each standard. The second measurements for the long-lived radionuclides were measured after 20 days for a collection time of 10,800 s (see Fig. 2).

The gamma-ray spectrometry of the irradiated samples was carried out by using the nuclear parameters listed in (Table 2) [35].

Table 1 The collected plants and its traditional uses in Algeria

Binomial name	Family/species	Local name	Traditional uses
Artemisia herba-alba Asso.	Asteraceae	Shih	Enteritis, intestinal disturbances, antiseptic, antispasmodic properties [19]
Artemisia compestris L.	Asteraceae	Dgouft	Febrifuge, against digestive troubles, menstrual pain [20, 21]
Laurus nobilis L.	Lauraceae	Rend	Rashes caused by poison ivy, astringent and salve for open wounds [22]
Origanum vulgare L.	Lamiaceae	Zaâtar	Anti-aspergillus property providing an inhibition of all assayed mold strains Antimicrobial uses in food conservation systems [23]
Mentha spicata L.	Lamiaceae	Naânaâ	Treatment of stomach ache, chest pains, irritable bowel syndrome [24].
Rosmarinus officinalis L.	Lamiaceae	Iklil Aljabal	Antibacterial, anticancer, antidiabetic, anti-inflammatory, antinociceptive [25]
Mentha pulegium L.	Lamiaceae	Flio	Antiseptic, antispasmodic, carminative, diaphoretic, emmenagogue, sedative, stimulant, uterine tonic [24].
Pistacia lentiscus L.	Anacardiaceae	Darou	Ointments for skin disorders, treatment of digestive system, easing high blood pressure, and reducing the risk of heart attacks [26, 27]

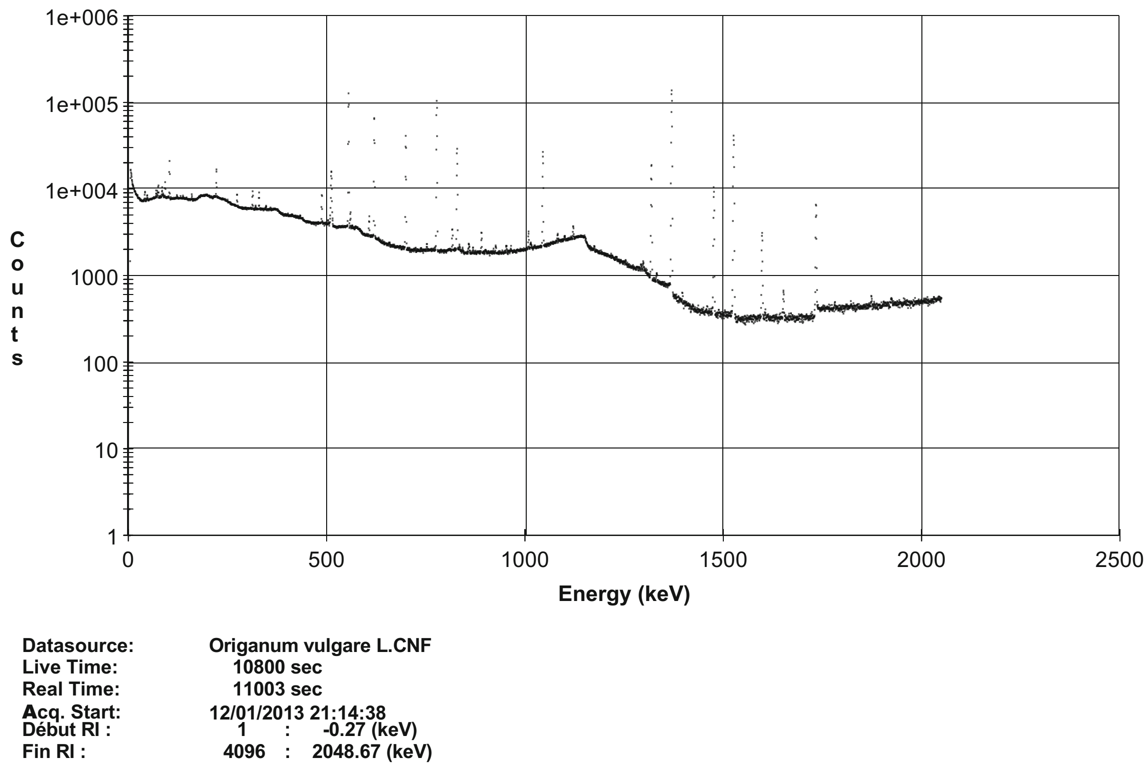

Fig. 2 Typical gamma-ray spectrum of the irradiated *Origanum vulgare* L. sample obtained using Genie-2k software

Results and Discussion

Concentrations of Toxic Elements

Six toxic elements were determined in different types of plants studied; the concentration values obtained for all samples analyzed are given in the Table 3.

In this study, six toxic chemical elements (As, Br, Ce, Co, Cr, and Sb) were identified by the INAA method. The main sources of arsenic contamination are found worldwide in the soil, water, air, and all living matter, and the problem of arsenic pollution is presently a major health concern worldwide [36]. Arsenic is a carcinogen and can cause cancer of the lungs, skin, bladder, and liver; on the other hand, intake of inorganic arsenic in lower concentrations may cause abnormal heart rhythm and decreased production of red and white blood cells [37]; from this study, arsenic was present in all plant species examined with almost the same concentration (between 0.18 μg/g to 2.91 μg/g) except arsenic in *Laurus nobilis* which is the highest with 5.44 μg/g. Elemental bromine is toxic and affects badly on human health, Br contents of different samples of plants were found to have a wide variation ranging from 3 to 63 μg/g. This variation may be due to the use of bromine-containing agricultural chemicals and pesticides such as methyl bromide and bromopropylate.

Table 2 Optimum experimental conditions and nuclear data employed in this study

Elements	Radionuclide	Half-life	γ-ray peaks (keV)	Decay time
Arsenic	^{76}As	25.86 h	559.1, 657.04	2–5 d
Bromine	^{82}Br	35.30 h	554.3, 698.08, 776.5	2–5 d
Cerium	^{141}Ce	32.508 d	145.4	20 d
Cobalt	^{60}Co	5.2714 y	1173.2, 1332.5	20 d
Chromium	^{51}Cr	27.7025 d	320.1	20 d
Antimony	^{124}Sb	60.20 d	169, 602.7	20 d

d days, *h* hours, *y* years

Table 3 Concentration of toxic elements (μg/g) in all samples

	As (μg/g)	Br (μg/g)	Ce (μg/g)	Co (μg/g)	Cr (μg/g)	Sb (μg/g)
Artemisia herba-alba	0.88 ± 0.10	63.32 ± 7.48	1.62 ± 0.17	0.27 ± 0.03	0.74 ± 0.02	0.04 ± 0.01
Artemisia compestris	0.46 ± 0.05	36.02 ± 4.26	1.58 ± 0.17	0.30 ± 0.04	1.50 ± 0.05	0.03 ± 0.01
Laurus nobilis	5.44 ± 0,60	2.58 ± 0.31	0.73 ± 0.08	0.13 ± 0.01	1.75 ± 0.05	0.05 ± 0.01
origanum vulgare	0.82 ± 0.09	4.76 ± 0.56	1.94 ± 0.20	0.48 ± 0.06	3.42 ± 0.11	0.06 ± 0.01
Mentha spicata L.	2.91 ± 0.32	12.14 ± 1.44	0.82 ± 0.09	0.35 ± 0.04	1.10 ± 0.03	0.04 ± 0.01
Rosmarinus officinalis	0.71 ± 0.08	3.90 ± 0.46	2.34 ± 0.25	0.40 ± 0.05	2.39 ± 0.08	0.04 ± 0.01
Mentha pulegium	0.97 ± 0.11	7.14 ± 0.85	3.28 ± 0.35	0.70 ± 0.08	2.78 ± 0.09	0.06 ± 0.01
Pistacia lentiscus fruits	1.82 ± 0.20	4.76 ± 0.57	0.23 ± 0.03	0.10 ± 0.01	2.18 ± 0.07	0.02 ± 0.001
Pistacia lentiscus leaves	0.18 ± 0.02	7.93 ± 0.94	0.56 ± 0.06	0.09 ± 0.01	1.60 ± 0.05	0.02 ± 0.001

The highest amount of bromine found in the samples is much lower than the permissible level of 250 μg/g [38]. Cobalt is considered an essential component of vitamin B12, but can accumulate in high levels in the liver, kidney, pancreas, and heart; cobalt has been found to produce tumors in animals and is likely a human carcinogen as well [39]; the concentration of Co is almost similar in range from 0.1 to 0.7 μg/g.

Deficiencies of the element of chromium may cause heart conditions, but the uptake of too much chromium can cause adverse health effects as well, for instance, skin rashes; for this work, the content of Cr ranged between 0.7 and 3.4 μg/g [39].

The cerium was found in all samples examined, with concentration ranging from 0.23 to 3.28 μg/g. Antimony is a highly toxic element; the Agency for Toxic Substances and Disease Registry has composed a comprehensive account of the public health effects of antimony [40]. The concentrations of antimony in our samples were ranged from 0.02 to 0.06 μg/g.

Quality Control and Evaluation of Results

The quality assurance procedures are described for individual steps in neutron activation analysis such as preparation, calibration, irradiation, counting and measurement of gamma-ray spectrum, and internal quality control [41]. Table 4 gives a comparison of our results for the reference material to its certified values and the statistical parameters relative bias (RB), *Z*-score, and *U*-score. The results are almost in good agreement with the certified values of two CRMs of NIST-SRM 1573a (tomato leaves) [42], and GBW 07605 (tea leaves).

The statistical parameters are calculated according to the following equation (Eqs. (1)–(3)):

$$U_{\text{score}} = \frac{|X_{\text{lab}} - X_{\text{ref}}|}{\sqrt{\mu_{\text{lab}}^2 + \sigma_{\text{ref}}^2}} \tag{1}$$

$$Z_{\text{score}} = \frac{X_{\text{lab}} - X_{\text{ref}}}{\sigma_{\text{ref}}} \tag{2}$$

$$\text{Relative bias (RB)} = \frac{X_{\text{lab}} - X_{\text{ref}}}{X_{\text{ref}}} \times 100\% \tag{3}$$

where X_{lab}, μ_{lab}, X_{ref}, and σ_{Ref} are the laboratory results, standard deviation, and the recommended and standard uncertainties respectively.

The laboratory performance is evaluated as satisfactory if *Z*-score ≤ 2, questionable for 2 < *Z*-score < 3, and unsatisfactory for *Z*-score ≥ 3; (result and certified value are not in agreement) [43].

Table 4 Quality assessment of the analytical results based on CRMs (NIST1573a) and GBW 07605

Element	NIST-1573a					GSV4 (GBW 07605)				
	Calculated values	Certified Values	*Z*-score	*U*-score	Bias	Certified values	Calculated values	*Z*-score	*U*-score	Bias
Br	1271 ± 150	1300	–	0.19	2.21	3.4 ± 0.4	3.48 ± 0.04	0.20	0.20	2.35
Ce	1.69 ± 0.18	2	–	1.72	15.5	1 ± 0.1	1.18 ± 0.04	1.80	1.67	18.0
Co	0.56 ± 0.02	0.57 ± 0.02	0.50	0.35	1.75	0.18 ± 0.02	0.19 ± 0.02	0.50	0.30	5.56
Cr	2.07 ± 0.07	1.99 ± 0.06	1.33	0.87	4.02	0.77 ± 0.03	0.80 ± 0.02	1.51	0.83	3.75
Sb	0.05 ± 0.01	0.063 ± 0.006	2.17	1.11	20.6	0.056 ± 0.005	0.07 ± 0.01	2.80	1.25	25.0

Table 5 Intake values (in mg/day, person) of some toxic elements FAO/WHO [11]. (All values are expressed on dry weight)

	Potential toxic chemical elements TDI (adult with 70 kg (BW))				
Elements	As (μg/day/person)	Br (mg/day/person)	Co* (mg/day/person)	Cr* (mg/day/person)	Sb (μg/day/person)
Artemisia herba-alba	8.8	0.63	0.003	0.007	0.4
Artemisia compestris	4.6	0.36	0.003	0.015	0.3
Laurus nobilis	54.4	0.02	0.001	0.017	0.5
Origanum vulgare	8.2	0.04	0.005	0.034	0.6
Mentha spp.	29.1	0.12	0.0035	0.011	0.4
Rosmarinus officinalis	7.1	0.04	0.004	0.024	0. 4
Mentha pulegium	9.7	0.07	0.007	0.028	0.6
Pistacia lentiscus fruits	18.2	0.05	0.001	0.029	0.2
Pistacia lentiscus leaves	1.8	0.08	0.0009	0.016	0.2
Males 19–50 years	150	70	0.2	1.7	420

TDI tolerable daily intake of adult (TDI/70 kg) of some toxic elements

*Recommended daily allowance expressed in mg/day, person for adult men

Estimation of the Dietary Intake

Studies that include the human food are very important to know and control the potential nutritional deficiencies or excess of toxic elements [11].

Medicinal plants are one of the most important sources of essential nutrients and toxic elements; for this reason, consumption must be under strict control.

In this study, we have assumed an intake of 10 g of plants leaves, and the average intake values per day and per person for toxic elements were determined; the results are given in Table 5, with the suggested daily tolerance limits. The estimation of probable intake values through the consumption of these samples shows that the contents of toxic elements are well below toxicological reference values and were found to satisfy nutritional recommendations.

Conclusion

This work was undertaken with the aim of investigating some toxic elements (As, Br, Ce, Co, Cr, and Sb) contained in the most widely used medicinal plants in Algerian traditional medicine. The content of antimony and cobalt are the lowest, whereas the content of bromine was found the highest; the results were found to vary depending on the plants. The study has allowed us to know that the toxic element concentrations in all the selected plants were well under the estimated permissible limits set by FAO/WHO for human consumption. The results obtained are informative and can support therapeutic usage, and also can let us to understand the mechanisms and interactions of some chemical compounds related to pharmacology and traditional medicine.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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