

Short communication

Natural radioactivity determination in samples of *Peperomia pellucida* commonly used as a medicinal herbFábio V. Sussa^a, Sandra R. Damatto^b, Marcos M. Alencar^b, Barbara P. Mazzilli^b, Paulo S.C. Silva^{a,*}^aInstituto de Pesquisas Energéticas e Nucleares – Centro do Reator de Pesquisas, (IPEN – CRPq/CNEN – SP), Av. Professor Lineu Prestes 2242, Cidade Universitária, CEP 05508-000 São Paulo, SP, Brazil^bInstituto de Pesquisas Energéticas e Nucleares – Laboratório de Radiometria Ambiental, (IPEN – LRA/CNEN – SP), Av. Professor Lineu Prestes 2242, 05508-000 São Paulo, SP, Brazil

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ABSTRACT

The concentration of ²³⁸U, ²³²Th, ²³⁰Th, ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb were determined in samples of *Peperomia pellucida* and in the surrounding soil, by alpha spectrometry and gross alpha and beta counting. The radionuclide activity concentrations ranged from 4.3 to 38 Bq kg⁻¹, 1.7–124 Bq kg⁻¹, 2.1–38 Bq kg⁻¹, 8.5–37 Bq kg⁻¹, 3.2–46 Bq kg⁻¹, 39–93 Bq kg⁻¹, respectively. In the plant extractions and infusions as used for consumption, the mean recoveries were from 23% to 60% in maceration and 24–75% in infusion.

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1. Introduction

The therapeutic use of medicinal plants by man is as ancient practice (Alvim et al., 2006). In recent times, the study of medicinal plants has become the focus of ever more extensive research all over the world, due to the diversity and potential that medicinal plants have as source of medicinal products (Ahmad and Beg, 2001; Lee et al., 2008; Rokaya et al., 2010; Street, 2012; Sharma et al., 2012; Shanmugam et al., 2012). Generally, studies related to therapeutic plants aim to characterize the active compounds of the plant for scientific evidence of their therapeutic properties (Naidu et al., 1999; Balunas and Kinghorn, 2005; Gambari and Lampronti, 2006; Jäger et al., 2010; Nyila et al., 2012). However, studies of the distribution of ²³⁸U and ²³²Th natural radionuclides series in such plants are sparse.

An emerging problem in many development countries is industrial pollution that threatens not only the health of the local environment but can lead to contamination of vegetation with toxic metals, pesticides, or radioactivity (Desideri et al., 2010). There are several sources contributing to plant contamination which can result in direct deposition of radioactive particles from the

atmosphere onto the above-ground parts, indirect sorption of radionuclides from the soil by the root system, as well as the resuspension and deposition of radionuclides in the soil. The presence of radionuclides in plants constitutes a pathway for their transfer to man. The amount of radioactive nuclides from U and Th series in edible vegetables are well known since they have been the main concern of research conducted worldwide (Smith-Briggs and Bradley, 1984; Santos et al., 1993; Radhakrishna et al., 1996; Pietrzak-Flis et al., 2001; Santos et al., 2002; Al-Masri et al., 2008; Ele Abiama et al., 2012; Jha et al., 2012).

Medicinal plants, on the other hand, are not included in the group of edible plants that have been studied for their radioactive content, possibly because the ingestion of radioactive material through their consumption has not been recognized or was considered insignificant (Desideri et al., 2010). A medicinal plant containing high concentrations of natural radionuclides can cause health problems, since they are commonly used over long periods.

The medicinal plant *Peperomia pellucida* (L.) KUNT is used in Brazilian folk medicine to treat abscesses, furuncles, skin sores and eye inflammation. In northeastern Brazil, the plant is also used to lower the cholesterol levels (Santos et al., 2001), as a cough suppressant, emollient, and diuretic. Ethno-medicinal uses of this plant vary depending on the region and literature data indicates its use for treating abdominal pain, colic, fatigue, gout, headache, renal disorders, rheumatic joint pain and to stop bleeding (Khan et al.,

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2008; Arrigoni-Blank et al., 2004; Bayma et al., 2000). Its roots are used to treat fevers and aerial parts used for dressing wounds (Munhoz et al., 2000). Literature data confirms the species antimicrobial (Bojo et al., 1994) and analgesic (Aziba et al., 2001) effects while other activities, such as anti-inflammatory, were not yet studied. Beyond its medicinal uses *P. pellucida* leaves and stems may be consumed as a vegetable.

Despite its wide use as a medicinal herb, the radionuclide content on *P. pellucida* has never been determined. In this sense, in this study, concentration of naturally occurring radionuclides ^{238}U , ^{232}Th , ^{230}Th , ^{226}Ra , ^{228}Ra and ^{210}Pb were determined in samples of *P. pellucida* and in the surrounding soil.

2. Experimental

The samples of *P. pellucida* were collected in the Botanical Garden of the city of Rio de Janeiro, Brazil, in December 2010. The species was identified in the Research Institute Botanical Garden, RJ and the voucher specimen (N°. PMSP 13184) has been deposited at the Ibirapuera Park Herbarium of São Paulo City, Brazil.

The samples of *P. pellucida* were washed with ultrapure water to remove impurities and soil particles present in all plant structures, air-dried and separated into aerial parts and roots. After these processes, the samples and soil were dried at 60 °C and ground into a fine powder. For the analysis of the extraction by maceration, dried *P. pellucida* aerial parts were softened in 70% ethylic alcohol during seven days at room temperature. The extract was filtered through Wattman filter paper and the residue discarded after washing. The filtrate was then evaporated to near dryness and separated for analysis of radionuclides. The powdered material of *P. pellucida* aerial parts was stored in centrifuge tubes with subsequent addition of 50 ml of Milli-Q® water at 90 °C in the tube. This mixture was homogenized and left to stand for 5 min to prepare the extract by infusion. After this period, the solution was filtered and the residue again discarded after washing. The final solution, in both cases were transferred to beakers, placed on heated plate at 150 °C, evaporated to near dryness and separated for analysis. The soil sample was completely dissolved in acid mixture of HNO_3 , HCl and HF.

The content of U and Th isotopes in the aerial parts, roots and soil was determined by alpha spectrometry after radiochemical separation by ionic exchange resins and measurement with a silicon surface-barrier detector as described in Silva et al. (2011) and references therein. The radionuclides measurement of ^{226}Ra , ^{228}Ra and ^{210}Pb were carried out by gross alpha and beta counting after radiochemical separation according to Sussa et al. (2009) and references therein. The percentage of ^{226}Ra , ^{228}Ra and ^{210}Pb extraction from medicinal plant into maceration and infusion was also evaluated in this study. All the concentrations presented are on dry weight basis.

3. Results and discussion

Radionuclide concentrations were determined in reference materials IAEA-300 and IAEA-326 for quality control of the measurements. The results were in good agreement with the certified values (Fig. 1).

Table 1 shows the activity concentrations (Bq kg^{-1}) of the natural radionuclides ^{238}U , ^{232}Th and ^{230}Th , ^{226}Ra , ^{228}Ra and ^{210}Pb determined in the medicinal plant selected in this study and in the surrounding soil. For comparison in the same Table the activity concentration for vegetable foodstuffs, tea and herbal medicines are also presented.

The world average of ^{238}U and ^{232}Th content in soil is 35 Bq kg^{-1} and 30 Bq kg^{-1} , respectively (UNSCEAR, 2000). The level of ^{232}Th

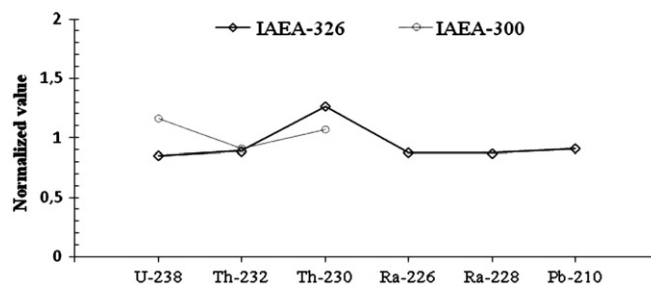


Fig. 1. Concentration values measured for reference materials IAEA-300 (radionuclides in Baltic sea sediment) and IAEA-326 (radionuclides in soil) normalized by the respective concentration certified values.

observed in this study was much higher than the worldwide average. The content of radionuclides in vegetation is dependent on the soil radionuclide concentrations. The uptake via root system is one of the main processes by which radionuclides are incorporated by plants. Another important pathway is the uptake of radionuclide directly from the atmosphere or from resuspended material (Vandenhove et al., 2009). It was observed in this study that the general trend of the radionuclide concentrations was the following: soil > roots > aerial parts.

From Table 1 it is possible to observe that the radionuclide concentrations in *P. pellucida* were in the same order of magnitude or slightly greater than that found in vegetables, tea and medicinal plants (leaves and roots) except for the values reported by Carvalho et al. (2009) for lettuce. The concentrations measured by Carvalho et al. (2009) are related to an area of high natural radioactivity, e.g. surrounding a uranium mine. In the same report, the values of ^{238}U , ^{232}Th , ^{230}Th and ^{226}Ra varied from $0.34\text{--}1.29 \text{ Bq kg}^{-1}$, $0.17\text{--}0.30 \text{ Bq kg}^{-1}$, $0.31\text{--}0.39 \text{ Bq kg}^{-1}$ and $1.3\text{--}1.2 \text{ Bq kg}^{-1}$, respectively, in lettuce grown in the reference control site and these values are lower than those reported here.

Activity concentrations of ^{226}Ra were higher than ^{228}Ra in the plant samples with the opposite occurring in the soil sample. As the former belongs to ^{238}U and the latter to the ^{232}Th decay series, these results are in accordance with those obtained for ^{238}U and ^{232}Th . Radium exist in nature in soil, rock, surface water, plants and animals at low concentrations generally lower than 37 Bq kg^{-1} in soil and rocks (ANL, 2005). As a member of the alkaline earth metals, radium isotopes present similar chemical characteristics as Ca and Mg that play important role in the plant growth and nutrition. This fact can explain the higher activity of ^{226}Ra and ^{228}Ra than their parents ^{238}U and ^{232}Th . Despite the fact that the analyzed soil had a relatively high concentration of ^{232}Th , the concentration of ^{228}Ra in the plant must be controlled by other factors than ^{232}Th soil concentration. According to Markose et al. (1983) and Rodriguez et al. (2002), soil-plant transfer of radium depends on the concentration of others elements such as iron in the plant and phosphorus and alkaline earth levels in the soil.

Although there are many uncertainties about the specificity of metals absorption mechanisms by plants, the content and the accumulation of the elements in plant tissue and roots usually depends on their availability in the soil solution. It has been observed that there is a greater accumulation of radionuclides in the roots and with higher concentration for ^{238}U . This accumulation could be related to immobilization of these elements in organic complexes present in the root tissue (Silva et al., 2007).

Incorporation of ^{210}Pb by above-ground plant-part occurs mainly via atmospheric deposition, and transfer via the root system is rather small (Bunzl and Trautmannsheimer, 1999). *P. pellucida* showed high concentrations of ^{210}Pb in all the analyzed plant parts. This can be explained by the fact that this radionuclide is a decay

Table 1
Activity concentration, mean and standard deviation of three replicates, in Bq kg⁻¹ dry weight of the radionuclides ²³⁸U, ²³²Th, ²³⁰Th, ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb determined in aerial parts and roots of *P. Pellucida*, soil and literature values.

	²³⁸ U	²³² Th	²³⁰ Th	²²⁶ Ra	²²⁸ Ra	²¹⁰ Pb	References
<i>P. Pellucida</i> aerial parts	4.3 ± 0.3	1.7 ± 0.2	2.1 ± 0.5	8.5 ± 0.2	3.2 ± 0.3	52 ± 3	This work
<i>P. Pellucida</i> roots	7.5 ± 0.2	7.0 ± 0.7	7.1 ± 0.3	23.2 ± 0.2	10 ± 1	58 ± 6	This work
Infusion				15 ± 1	28 ± 5	32 ± 7	This work
Maceration				12 ± 1	25 ± 7	30 ± 6	This work
Soil	38 ± 5	124 ± 2	38 ± 3	37 ± 1	46 ± 4	93 ± 3	This work
Lettuce	2.36–123	0.80–4.3	2.2–28–5	7.2–29.7			Carvalho et al., 2009
	0.42–0.91					0.37–0.38	Ekdal et al., 2006
	0.09–1.37			0.71–2.77	0.13–4.94		Anke et al., 2009
Cabbage						0.24–0.33	Lauria et al., 2009
						7.0–17.8	Ekdal et al., 2006
Herbal tea				0.7–4.9	2.3–8.0	4.0–36.8	Santos et al., 1993
Tea				1.6–4.5	2.5–9.0	<10–58.9	Desideri et al., 2011
		0.42–6.36		0.55–23.4			Desideri et al., 2011
	0.9	2.7					Görür et al., 2011
Medicinal plants		0.2–3.7		<2.2–18.4	33–65	32–76	Kılıç et al., 2009
Medicinal plant leaves	<0.1–0.97			<0.3–6.9		27.1–58.3	Sussa et al., 2009
	0.51–1.61	0.11–0.97					Desideri et al., 2010
Medicinal Plant roots	<0.1–0.39			<0.3–1.2		<0.3	Reddy et al., 1998
							Desideri et al., 2010

product of ²²²Rn, a noble gas which is continuously emanated from the soil to the atmosphere, and can be deposited on vegetation and incorporated by foliar or root absorption (Moreira, 1993). It is interesting to note (Table 1) that high concentrations of ²¹⁰Pb were also observed in other Brazilian plants (Sussa et al., 2009).

The incorporation of radionuclides in the body occurs mainly through the food chain. In the case of vegetable consumption, the amount of radioactive elements present depends on the soil-to-plant transfer factor. The transfer factor (fv) for our sample was calculated according to IAEA-TECDOC-1616 (2009) for the determined radionuclides in the aerial parts and the root of *P. pellucida* and the results presented in Table 2.

Observed transfer factor values reported in literature vary enormously and various factors determine this variability (IAEA-TECDOC-1616, 2009). The Th transfer factor has a lower value, both in the aerial parts and roots, as this element is considered to be rather immobile. The higher values were observed for Ra-isotopes and ²¹⁰Pb. Comparison of the values obtained in this study with those reported in IAEA-TECDOC-1616 (2009) shows that fv value observed for ²³⁸U is in the same order of magnitude for herbs and is slightly higher than leaf vegetables (1.09E-01 and 3.57E-02, respectively). The Th values for leaf vegetables ranges from 9.38E-05 to 2.11E-01 and our value was in accordance with this and in good agreement with the arithmetic mean of 1.18E-01. The reported fv for Ra in herbs varies from 5.30E-03 to 3.31E+00 with mean value of 2.61E-01. The transfer factor measured for *P. pellucida* is in good agreement with those values. Finally, ²¹⁰Pb has a reported mean value varying from 3.21E-03 to 2.46E+01 for leaf vegetables. The value observed in our results was slightly lower than the mean value in the same crop group (2.11E+00). The higher values of fv for ²¹⁰Pb and the small difference between the value observed for the root and aerial part confirm the importance of the atmospheric uptake of this nuclide.

Fig. 2 shows the ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb extraction percentage results for maceration and infusion carried out on the *P. pellucida* samples and the comparison between maceration and infusion

Table 2
Transfer factors (dry weight basis) of the radionuclides ²³⁸U, ²³²Th, ²³⁰Th, ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb.

	²³⁸ U	²³² Th	²³⁰ Th	²²⁶ Ra	²²⁸ Ra	²¹⁰ Pb
Aerial parts	1.13E-01	1.37E-02	5.53E-02	2.30E-01	6.96E-02	5.59E-01
Roots	1.97E-01	5.65E-02	1.87E-01	6.27E-01	2.17E-01	6.24E-01

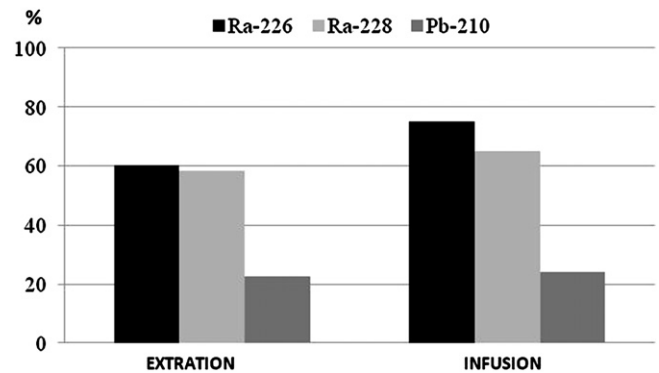


Fig. 2. Percentage of ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb extraction in maceration and infusion.

extraction. The activity concentration for those nuclides in the extract is also presented in Table 1. The percentage trend is the following: infusion > maceration.

This difference is related to the interaction of metals with substances in plant structures, such as macromolecules and small molecules and the solubility of metals in different solvents (Veiga et al., 2007). From the results it can be concluded that Ra-isotopes are mostly extracted by both methods while ²¹⁰Pb only migrate from plant to extract in an extent lesser than 25%. Nevertheless, the activity concentration for ²¹⁰Pb was higher than that of Ra so that the total amount of ingested radionuclide (in Bq kg⁻¹) is almost the same as depicted in Table 1.

4. Conclusions

In this paper, a study of the natural radioactivity in sample of *P. pellucida* was presented. ²³⁸U, ²³²Th and ²³⁰Th were determined by alpha spectrometry and ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb were determined by gross alpha and beta counting after radiochemical separation. The percentage of ²²⁶Ra, ²²⁸Ra and ²¹⁰Pb extraction in maceration and infusion was also determined.

As no reference values for radionuclides in medicinal herbs are available, the results were compared with vegetable foodstuff, tea and herbal medicines concentrations reported in published literature. The level of radionuclides determined in this study presented the same order of magnitude or slightly higher concentrations than that used for comparison except those from contaminated areas.

Considering the maceration process, one of the methods used for extracting active compounds from medicinal plants for the production of phytotherapies and one of the ways by which the herbs are consumed by the population (tea), the study of radionuclides concentration in medicinal plants has great significance as this is the main via of radionuclides ingestion, and as showed, approximately 50% of the Ra isotopes and almost 25% of ^{210}Pb present in the plant is transferred to the extract but in the same concentration.

Transfer factor analysis indicated that the *tf* values for *P. pellucida* was in good agreement with other plants used as tea and medicine and confirmed the importance of ^{210}Pb atmospheric deposition in the absorption of this nuclide.

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