Radiation Emergency Medicine 2015 Vol.4, No.2 40–44

Note

^{210}Po, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs Concentration of Medicinal Herbs

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Received 24 February 2015; revised 9 June 2015; accepted 8 July 2015

The ^{210}Po, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentrations were measured related to dry material of ten types of medicinal herbs factory packaged for making tea. For ^{210}Po measurement alpha-spectrometry was applied, while the rest of the radionuclides were determined by gamma-spectrometry method. In case of ^{210}Po the highest activity concentration (10-19 Bq/kg) was measured in herbs consisting of only leaves, while the lowest activity concentration (≤ 2 Bq/kg) was measured in medicinal herbs consisting of solely flowers and for ^{210}Pb similar trend was found. For the rest of the radionuclides of natural origin the relation was not so obvious. In case of ^{137}Cs, the measured values were lower (0.4-1.6 Bq/kg) in cultivated medicinal herbs, than in case of wild grown samples (0.4-20 Bq/kg).

Key words: natural radioactivity; herbs; population dose; dose contribution

1. Introduction

The World Health Organization definition[1] for medicinal herbs (plants) is: herbs which have been proven or thought to have medicinal effects. Study of the elemental content in medicinal plants is very important because some of these elements are closely related to human health. As every plant, medicinal herbs (plants) contain naturally origin- (^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K) and artificial radioactive isotopes (^{137}Cs) as well[2-5]. The radionuclides presumably do not play any significant role in the health effect but due to the dosimetric aspect because mainly the tea of the herbs is used as infusion[6].

Primarily, radioisotopes origins from the soil and from the atmospheric fall-out are up-taken and assimilated in plants. Several researchers have performed experiments on clarifying the parameters determining the method, the degree of assimilation and the origin of radionuclides.

According to the studies so far, radioisotopes like ^{210}Po, ^{210}Pb primarily fall on the surface of the plants from the atmosphere, where they can stick for a shorter or a longer period depending on the surface quality of the leaves or may even assimilate into the given plant. High values were typically found in tobacco samples[7, 8] due to their longer growing season and special leaf-surface (large surface covered with "sticky hairs").

Radionuclides, like ^{226}Ra, ^{232}Th and ^{40}K primarily assimilate in the parts of the given plant by absorption through the roots from the soil. The degree of absorption is influenced by several parameters such as the texture, pH-, oxygen-supply of the soil, and its other cation and anion concentration etc.

Several parameters influence the transport processes within the plants, which determine in which part of the plant the given radionuclide accumulates. Based on the previous researches, it is apparent that great differences can be expected concerning radionuclide concentration in case of same species of plants but different origin/soils. As an example, in Germany in different plants values

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Table 1. Major data of medicinal herbs measured

Number	Name	Latin name	Part used	Cultivated/Collected
1	Malm	Melissa officinalis	Leaves	Cultivated plant
2	Calendula/Marigold	Calendula officinalis	Flowers	Cultivated plant
3	Lavender	Lavandula angustifolia	Flowers	Cultivated plant
4	Peppermint	Mentha x piperita	Leaves	Cultivated plant
5	Stinging nettle	Urtica dioica	Leaves	Feral plant
6	Camomile	Matricaria recutita	Flowers	Feral plant
7	Linden	Tilia platyphyllos	Flowers, Bracts	Feral plant
8	Birch	Betula pendula	Leaves	Feral plant
9	Hip	Rosa canina	Pseudo-crop	Feral plant
10	Milfoil	Achillea collina	Floral stem	Feral plant

related to wet mass were measured for ^{210}Po from 4 to 7400 mBq/kg, for ^{210}Pb from 4 to 4100 mBq/kg, while in case of ^{226}Ra from 6 to 1150 mBq/kg[2].

During food consumption the world average of the annual effective dose for elements of the uranium and thorium decay chains is 0.14 mSv/year, and for radionuclide ^{40}K it is 0.17 mSv/year[2].Taking the uranium and thorium decay chain into consideration, the activity concentration values of the assimilated radionuclides are the following: ^{210}Po (58 Bq), ^{210}Pb (30 Bq), and ^{226}Ra (20 Bq). Their contributions to the radiation exposure are: ^{210}Po (64%), ^{210}Pb (20%), and ^{226}Ra (4.5 %), that means almost 90% of the internal dose comes from these three isotopes. Among the artificial isotopes ^{137}Cs was measured. Nowadays, consumption of tea made of different medicinal herbs is rather popular and widespread. Growing medicinal herbs has a past in Hungary, it has been a considerable agricultural activity in this country. Medicinal herbs are grown partly in cultivated lands; however, in the majority of the cases self-grown/home grown plants are collected and sold in the shops. Unfortunately in most cases no data is available regarding to their origin, therefore the radionuclide concentration of the soils as well.

During our work the activity concentration of ^{210}Po, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were determined in some of the most popular and commercially sold medicinal herbs used as infusion of herb tea.

2. Measurements and Methods

2.1. Sampling

Herb samples were purchased from herb shops, supermarkets traded by the same manufacturer. Medicinal herbs were packed in bags in a form suitable for making tea. According to the descriptions the samples were only dried, homogenized, and packed by the manufacturer. Major data concerning the species, type of cultivation and the parts of the plant are summarized in Table 1.

From the measured samples four are from cultivated land, while six plants grown in nature and were collected. Four of the samples were leaves, six were flowers and pseudo-crops. During our work the concentration of ^{210}Po was measured applying alpha-spectrometry, while the activity concentration of ^{210}Pb, ^{40}K, ^{226}Ra, ^{232}Th, and ^{137}Cs were quantified applying gamma-spectrometry method.

2.2. ^{210}Po concentration measurement by alpha-spectrometry

Sample digestion

Known activity of ^{209}Po tracer (32 mBq) was added as an internal tracer to 4 g of dried herb sample. The samples were digested subsequently using three portions of 20–30 cm^3 concentrated HNO_3. Samples were carefully heated and evaporated nearly to dry, approximately 2-3 cm^3 solution left at the bottom of Erlenmeyer flask.

Afterwards, 20-30 cm^3 concentrated HCl was added to remove the nitrates from the solution. In order to digest organic materials completely, 1 cm^3 of concentrated H_2O_2 was added carefully dropwise, then, 20 cm^3 ultrapure water was added and evaporated to near dryness. Each digestion step was introduced three times altogether. Finally, 100 cm^3 stock solution corresponding to 0.5 M HCl was prepared from the residue with distilled water and concentrated HCl[9].

Alpha source preparation: Polonium was deposited spontaneously from 0.5 M HCl stock solution onto high nickel-content stainless steel disc (WNr. 1.4539, DIN 17740, 25% Ni) for 3 h at 80 °C.300 mg ascorbic acid/ 50 cm^3 solution was added to reduce Fe^{3+} into Fe^{2+}. Two sources were prepared from stock solutions. After depositing, the disc was removed from the deposition cell and rinsed with distilled water, ethanol and dried carefully (< 150 °C) on a hot plate. The activity of ^{210}Po was measured in ambient temperature using an alpha spectrometer equipped with a semiconductor silicon detector.

Measurement: The applied alpha spectrometer was a Eurisys Measures-made Canberra Model 7401 type alpha

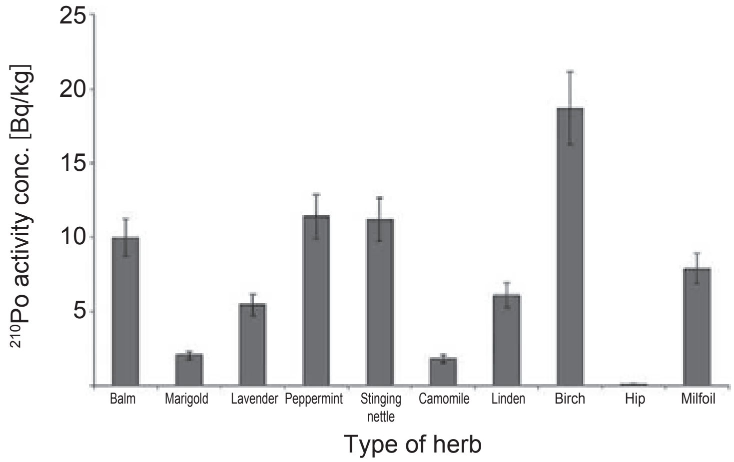

Fig. 1. Measured ^{210}Po concentration of medicinal herbs (error bars mean the standard deviation from the measurement of 3 parallel samples).

Table 2. ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentration values related to dry material-content of medicinal herbs

Number	Herb	Radionuclide concentration (Bq/kg dried sample)				
		^{210}Pb	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
1	Malm	43.8	1.15	3.01	639	1.57
2	Calendula/Marigold	32.6	1.14	3.41	733	0.58
3	Lavender	37.7	0.87	3.59	615	1.47
4	Peppermint	42.6	1.07	3.72	562	0.48
5	Stinging nettle	75.3	1.75	2.62	848	15.4
6	Camomile	34.1	0.84	4.79	848	20.0
7	Linden	33.2	<	<	456	4.07
8	Birch	120	1.93	4.52	437	11.2
9	Hip	<	1.17	2.92	625	1.26
10	Milfoil	37.2	1.26	3.65	660	0.42

("<" means below detection limit)

chamber equipped with 19 keV resolution PIPS detector. The minimum detectable activity (95% confidence level) was 0.36 mBq/g with a counting time of 200,000 sec.

2.3. Determination of ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentration by gamma-spectrometry

Herb samples were pulverized and closed in a polyethylene Marinelli vessel (volume 500 cm^3) and were stored for 30 days. The activity concentrations of natural radionuclides were determined by high-resolution gamma-ray spectrometry using Eurisys EGNC 20-190-R n-type HPGe detector with an efficiency of 20% and 1.8 keV energy resolution at the energy peak of 1,333 keV of ^{60}Co isotope. Gamma spectra were recorded by Tennelec PCA-MR 8192 multi-channel analyser. Data collection time was 200,000 sec.

The system was calibrated using a similar density and matrix moss reference material certified by the MgSzH Radio-analytical Reference Laboratory[10]. ^{226}Ra concentrations were determined by measuring the activities of its decay products, ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 and 1,120 keV), which were in secular equilibrium with ^{226}Ra following the 30-day storage. The activity of ^{210}Pb was measured by the 47.45 keV gamma ray, ^{40}K by the 1,461 keV gamma ray, ^{232}Th by the 911 keV gamma ray of ^{228}Ac, and ^{137}Cs by the 661.6 keV gamma ray[11].

3. Results and Discussion

^{210}Po concentration of the medicinal herbs is shown in Figure 1.

As is seen in Figure 1, the ^{210}Po concentration is not influenced either if it was cultivated or (1-4) or wild grown. However, it is shown that in case of the so-called pseudo-crop and the flower – calendula (2), camomile (6), and hip (9) – low ($\leq$ 2 Bq/kg), and in case of medicinal herbs containing flower and leaves or bracts – lavender

(3), linden (7), and milfoil (10) – middle level (5-8 Bq/kg), while in case of medicinal herbs consisting only of leaves – balm (1), peppermint (4), stinging-nettle (5), birch (8) – high ^{210}Po values were measured. This is probably due to the fact that plants during the atmospheric fall-out absorb polonium through their leaves. High values measured in tobacco leaves are also explained by this way elsewhere in the literature[11, 12)]. This is also confirmed by those observations that the mobilization of polonium is low in plants, so only a part of the polonium fallen and bound on the leaves is transferred to other parts of the plant[4, 13)].

The ^{210}Pb, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentration values related to dry material-content of medicinal herbs measured using gamma-spectrometry (max. ± 15%) are summarized in Table 2.

After analysing ^{210}Pb concentration it can be stated that it has similar/same distribution in the plants as ^{210}Po has, the highest values were found in leaves, while the lowest in flowers. The ratio of ^{210}Pb/^{210}Po in air is approximately 3-10[14)], so the high ^{210}Pb concentration is not surprising if the preliminary supposed fall-out is considered to be its origin. In case of hip the ^{210}Pb concentration was below the detection limit.

According to literature data the ^{226}Ra content of the plants origin from soil, and its concentration can be influenced by several factors, such as the Ca-, Mg- ion concentration of the soil, its phosphate-content, and also the pH of the soil[15, 16)]. Data concerning the origin of the medicinal herbs (growing location) therefore the properties of the given soils were not available. The relatively high measured activities for stinging-nettle with high iron-content can be explained by the absorption behaviour of radium and the iron content of the plant[17-19).]

In our work, the ^{232}Th activity concentration was found to be higher than the ^{226}Ra activity concentration and can be considered as a surprising result, as the mobility of thorium is lower than that of radium. Therefore, in general lower values are measured in plants as it is presented[4, 20)]. However, our measured values show an agreement with the measurements performed by Matiullah et al., where the concentration of ^{226}Ra varied from 1.35 to 1.71 Bq/kg, while the concentration of ^{232}Th ranged from 2.37 to 7.20 Bq/kg[21)].

The ^{40}K content is much higher than the concentration of any other radionuclides. This is not surprising, as the non-radioactive potassium-content of the plants is relatively high, so almost half of the internal radiation exposure entering the organism with nutrients, fertilizer comes from ^{40}K radionuclide[2)]. The potassium absorption of plants is also influenced by several factors (Ca/K ratio in the soil, pH-value, mobile K-content, oxygen-supply of the root zone, etc.), this can explain the significant difference found in the measured medicinal herbs. In case of the stinging-nettle, the high ^{40}K concentration can be explained by the high potassium-content of the plant.

The K-Cs ratio has been measured earlier by several researchers[22)]. In general, it can be stated that high the K-content is, the low the absorption of caesium is but we did not have any related data. Badran et al. have also observed significant differences between ^{40}K and ^{137}Cs-contents in plants[23)]. However, it is clear that in case of cultivated medicinal herbs the activity concentration of caesium is usually low, while in case of plants grown in the uncultivated soil, high values were measured in several cases.

4. Summary

It can be stated that the radionuclide-content of medicinal herbs significantly differs by species and by isotopes.

The highest ^{210}Po values (10-19 Bq/kg) were measured in medicinal herbs containing only leaves, therefore in case of polonium it is likely that the polonium fall out plays a great role in the ^{210}Po transport onto the plants. This is also confirmed by the fact that in case of flowers containing also stems/stalks (leaves) lower (5-8 Bq/kg) activity concentration was measured, while in case of medicinal herbs containing exclusively flowers even lower (≤ 2 Bq/kg) activity concentration was measured. The trend was similar for ^{210}Pb.

In case of ^{226}Ra, ^{232}Th, ^{40}K origin likely to be coming from the soil and no such trend was found.

For ^{137}Cs significantly different values were measured, but it is apparent that the radionuclide concentration of caesium in medicinal herbs grown in cultivated soil was lower (0.4-1.6 Bq/kg), while in case of samples grown widely it was higher (0.4-20 Bq/kg).

Since ^{210}Po and ^{226}Ra are alpha emitting radionuclides, they might result an excess of internal dose exposure in humans.

Acknowledgements

The present study was supported by the Hungarian State and the European Union projects Grant No. TÁMOP-4.2.2.A-11/1/KONV-2012-0071.

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