DETERMINATION OF FOOD RADIOACTIVITY BY MEANS OF LOW-LEVEL GAMMA SPECTROSCOPY

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ABSTRACT

The recognition of radionuclides from characteristic gamma rays is the most widely used method for activity concentration determination. It is shown that by means of high resolution low background gamma spectroscopy most of radionuclides in the food can be determined. By means of passive and active shielding of the detectors detection limits far below 1 Bq/kg can be achieved. Some recent results on radionuclide content of food on the market will be present. The role of TENORM sources in food contamination will be discussed.

INTRODUCTION

Nowadays, both natural and produced radionuclides are present in the environment. In the soil and water the content of natural radionuclides is enhanced by non-nuclear technologies (coal combustion, mining, fertilizing, etc.) while the artificial radionuclides have entered the environment due to atomic bomb explosions and nuclear arm testing. The most significant present-day produced radionuclide is ^{137}Cs of Chernobyl origin. Radionuclides from the environment are transferred to foods and human body through complex nutrition chains (ICRP, 1975; UNSCEAR, 1982; Linsley, 1997).

To assess the internal dose of radionuclides taken in by food, the activity of each radionuclide in food and water has to be measured. Since all radionuclides except ^{90}Sr and ^{3}H, emit gamma rays, gamma-spectroscopy is accepted as the most suitable method for simultaneous

multi-isotopic analysis. The ingestion dose can be estimated from the activity concentration of each radionuclide in food, taking into account the amount of the ingested food and using appropriate models (ICRP, 1991).

The Laboratory of Nuclear Physics at the Faculty of Sciences in Novi Sad has been monitoring radioactivity in food for years. The program covered both natural and artificial radionuclides in the environment and the ambient gamma-dose rate level. To the monitoring program, the Laboratory introduced new methods of sample preparation and new systems to detect and determine low levels of natural and artificial radionuclides in environmental samples.

MATERIAL AND METHOD

Samples of foodstuffs were collected on Novi Sad markets in 2002. The average mass of the samples was about 0.5 kg. After the homogenization and drying at 105^{o}C to a constant mass the samples were put in special cylindrical vessels. The radionuclide content of the samples was measured by means of a reversed electrode "GMX" type HPGe spectrometer (ORTEC, nominal efficiency 36%, resolution 1.9 keV). The detector had a thin dead layer on its outer surface and a beryllium entrance window enabling it to detect gamma radiation below 100 keV with efficiency of about 1% for bulk sources in contact geometry. The detector is inside the 25 cm thick iron shield made of pre-Second World War cast. The detector was calibrated in cylindrical geometry by the reference standard material NBS 4350B. The average counting interval was about 50 ks. A modified version of the SAMPO program was used to process the spectra presenting spectral intensities or detection limits for 20 selected nuclides. The measuring uncertainties were quoted at 95% confidence level.

Animals and indirectly human population can be exposed to radioactivity by ingestion of plant origin forage. Today Lucerne is considered to be the most important fodder crop in the world because it is an excellent source of iron, calcium and beta-carotene. In this paper radiation levels of radium and cesium in Lucerne samples determined by gamma-spectrometry measurements in actively shielded Ge detector will be presented. The samples of Lucerne were taken from twelve different locations in Vojvodina during the summer period July – September 2004.

Raw Lucerne trees were cut on the height of 4 – 5 cm under the ground in the amount of 2 – 3 kg. The samples were then dried on the air and after that ground, powdered and mineralized by method of dry burning on the temperature of 450ºC ± 10ºC (IAEA, 1989). In the air dried samples the wet content was determined on the temperature of 105º. Gamma spectrometry measurements of the Lucerne ash were performed by means of ultra low-level background germanium detector using both active and passive shielding. Time of measurements was 80 ks. Extended range (10 keV-3 MeV) GMX type HPGe detector, nominal efficiency of 32% in 12cm thick cylindrical lead shield, lined with 3.5 mm Sn + 0.5 mm Cu is surrounded by five 5cmx50cmx50cm plastic veto shields. Veto plastic scintillators and Ge detector operate in anticoincidence mode and on that way all events that are simultaneously detected in any veto and Ge detector will be rejected. The active shield reduces the integral background by factor 3 in the energy range from 50 to 2800 keV (Bikit, 2006).

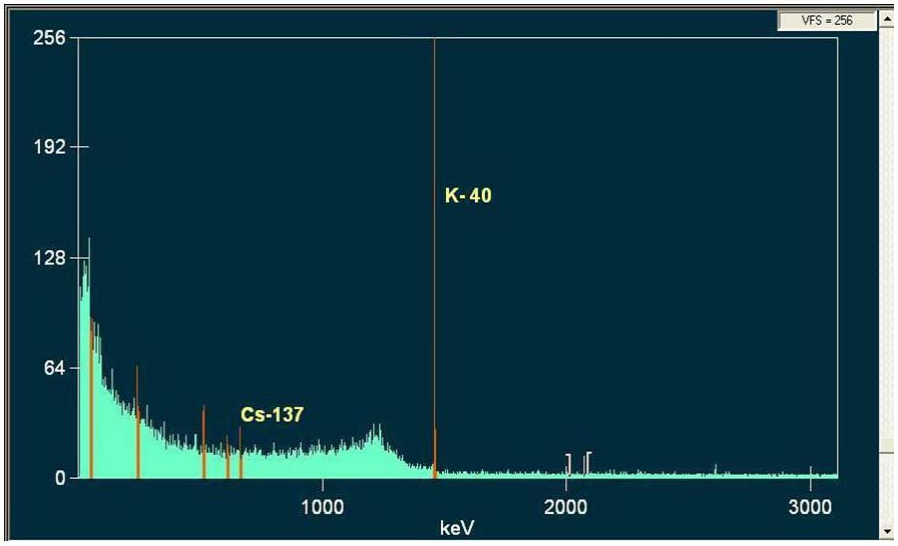

Figure.1. Gamma spectrum of Lucerne sample
measured by active shielded HPGe detector

The example of gamma spectrum of lucerne sample is shown in Fig 1. The background is significantly suppressed by the developed active

shielding method devices. For cesium ^{137}Cs 10 mBq/kg order of magnitude detection limits were achieved.

RESULTS

Samples of vegetables (13), cereals, wheat and cereal products (10), baby food (12), dairy products (9), meat products (9), tea and herbs (6), meat (10), fruit (9), and cacao powder sugar and salt (4) were measured. The average activity concentrations of the radionuclides in the investigated food samples are presented in Table 1. ^{40}K was the only radionuclide detected in all the samples. Therefore, the ingestion dose was estimated only for this isotope (Table 2).

Results of gamma spectrometry analysis of Lucerne samples are shown in Table 3. Activity concentrations of radionuclides: ^{134}Cs, ^{137}Cs, ^{226}Ra, ^{232}Th and ^{40}K were expressed in units of mBq/kg. In some of samples cesium ^{137}Cs were detected in traces. These concentrations are 200 times lower as compared with the results in 1988, two years after Chernobyl accident (Bikit, 1990) when the average ^{137}Cs activity concentration was (9.0±1.0) Bq/kg.

DISCUSSION

The estimated annual ingestion dose (Butezatu et al, 2002) for the adult population is 194 μSv/yr, while that for infants is 61 μSv/yr. The estimated dose values based on the ^{40}K activity concentration did not exceed the lower limit. Generally, the estimated values are in two orders of magnitude less than the authorized annual dose limit of 1 mSv/yr absorbed from all radiation sources. The consummation of dairy products seemed mostly contribute to the estimated dose.

In conclusion, the results of gamma-spectroscopic measurements of 86 foodstuffs confirm that the radioactivity of food from the Novi Sad markets is very low. While after the Chernobyl accident in 1986, ^{137}Cs was the dominant radionuclide in the environment (Dragović and Stanković, 2001), it is now detected only in some samples of activities far below the limits prescribed by the Yugoslav Regulation (YR, 1999.).

Table 1/a. The average activity concentrations in food on Novi Sad markets: A_S [Bq/kg]

	Vegetables	Cereals, wheat and cereal products	Baby food	Dairy products
Radionuclide	A_S [Bq/kg]			
^{75}Se	<0.04	<0.1	<0.1	<0.08
^{144}Ce	<0.3	<0.6	<1.0	<0.5
^{141}Ce	<0.1	<0.2	<0.3	<0.15
^{125}Sb	<0.11	<0.2	<0.3	<0.24
^{7}Be	1.3±0.5	<0.8	<1.0	<0.7
^{103}Ru	<0.04	<0.1	<0.1	<0.08
^{134}Cs	<0.05	<0.1	<0.1	<0.09
^{124}Sb	<0.04	<0.1	<0.1	<0.08
^{106}Ru	<0.5	<1.0	<1.0	<0.9
^{110m}Ag	<0.03	<0.05	<0.1	<0.06
^{137}Cs	0.14±0.07 (1)	<0.1	0.6±0.3 (2)	<0.14
^{95}Zr	<0.05	<0.1	<0.2	<0.11
^{95}Nb	<0.04	<0.08	<0.1	<0.10
^{58}Co	<0.05	<0.1	<0.1	<0.09
^{160}Tb	<0.11	<0.2	<0.4	<0.27
^{60}Co	<0.04	<0.08	<0.2	<0.07
^{238}U	<1.4	<2	<4	<2
^{226}Ra	2.4±0.4 (2)	<0.4	<0.5	<0.5
^{232}Th	0.14±0.11 (1)	0.75±0.4 (2)	1.0±0.5 (4)	<0.24
40K	138±3	68±6	178±6	96±4

However, marked amounts of ^{137}Cs were still present in some tea and herbs samples. Natural ^{226}Ra and ^{232}Th were found only in a few samples and even in these close to the detection limits. Supraliminal amounts of the natural cosmogenic radionuclide ^{7}Be were present in some samples of tea and herbs, while those of ^{40}K were as expected, present in all foodstuffs.

Nevertheless, the vicinity of several nuclear reactors and the possibility to import foodstuffs of considerable radioactivity justify the necessity of regular and systematic radiological monitoring of foodstuff markets.

Table 1/b. The average activity concentrations in food on Novi Sad markets: A_S [Bq/kg]

	Meat products	Tea and herbs	Meat	Fruit	Cacao powder, sugar, salt
Radio-nuclide	A_S [Bq/kg]				
^{75}Se	<0.07	<0.31	<0.06	<0.04	<0.11
^{144}Ce	<0.5	<2	<0.4	<0.4	<2
^{141}Ce	<0.16	<0.6	<0.11	<0.7	<0.16
^{125}Sb	<0.23	<0.8	<0.17	<0.12	<0.3
^{7}Be	<0.8	16±4 (3)	<0.6	1.3±0.4 (1)	<1.1
^{103}Ru	<0.08	<0.3	<0.07	<0.04	<0.14
^{134}Cs	<0.09	0.8±0.4 (1)	<0.07	<0.05	<0.1
^{124}Sb	<0.16	<0.4	<0.08	<0.05	<0.09
^{106}Ru	<0.7	<3	<0.8	<0.6	<1.0
^{110m}Ag	<0.06	<0.26	<0.06	<0.04	<0.08
^{137}Cs	<0.14	26±3 (2)	0.41±0.27 (1)	0.1±0.06 (1)	1.0±0.6 (1)
^{95}Zr	<0.12	<0.6	<0.11	<0.08	<0.19
^{95}Nb	<0.10	<0.4	<0.08	<0.06	<0.11
^{58}Co	<0.11	<0.3	<0.08	<0.06	<0.14
^{160}Tb	<0.25	<0.11	<0.22	<0.15	<0.4
^{60}Co	<0.06	<0.3	<0.08	<0.07	<0.12
^{238}U	<2	<13	<2	<1.7	<3
^{226}Ra	<0.5	1.6±1.2 (1)	<0.25	1.9±0.4 (2)	2.7±0.5 (2)
^{232}Th	<0.24	2.0±0.7 (2)	<14	<0.2	1.5±0.5 (3)
40K	79±3	591±12	98±3	55±2	432±13

In parenthesis: number of samples in which the radionuclides were detected. < Less than the value of the detection limit given after the symbol.

The sophisticated sample preparation and counting techniques enabled the detection of ^{226}Ra and ^{232}Th in almost all samples. The dominant ^{40}K activity concentration is expected for plants.

Table 2. Average activity concentrations of ^{40}K and estimated ingestion dose of ^{40}K

	Average activity concentration A_{av}[Bq/kg]	Estimated ingestion dose D [μSv/yr]
Vegetables	138	43
Cereal, wheat and cereal products	68	38
Dairy products	96	53
Meat products	79	6
Herbs and tea	591	2
Meat	98	9
Fruits	55	8
Cacao powder, sugar, salt	432	35
Total		194
Baby food	178	61

ACKNOWLEDGMENTS

The authors acknowledge to the Ministry of Science of Republic of Serbia (project No 141002B: Nuclear spectroscopy and rare processes) at the financial support of this research, as well as to financial support of the European Agency for Reconstruction through the Ministry of International Economic Relations within the Neighbouring Programme Hungary-Serbia (Action entitled "Regional Water Resources Investigations in the Scope or Sustainable Development", Grant No. 04SER02/01/009).

Table 3. Activity concentrations of ^{134}Cs, ^{137}Cs, ^{226}Ra, ^{232}Th and ^{40}K in Lucerne samples taken from different locations in Vojvodina during 2004

Location	Vršac	S.Crnja	B.Topola	Šid
Isotope	A [mBq/kg fresh mass]			
^{134}Cs	<75	<100	<80	<75
^{137}Cs	122±22	<100	70±15	<100
^{226}Ra	175±100	302±70	<300	<150
^{232}Th	90±62	218±25	80±15	<38
^{40}K	(121±4) x10^3	(140±4)x 10^3	(92±3)x1 0^3	(103±3)x 10^3

Location	Sr.Mitrovica	Temerin	Kula	Sombor
Isotope	A [mBq/kg fresh mass]			
^{134}Cs	<100	<100	<75	<75
^{137}Cs	<100	<125	<75	<100
^{226}Ra	<1500	275±225	450±150	175±150
^{232}Th	112±28	132±30	335±22	65±20
^{40}K	(141±4)x1 0^3	(191±6)x 10^3	(88±3)x 10^3	(144±5)x 10^3

Table 3 continued

Location	Sanad	Klek	S.Pazova	S.Moravica
Isotope	A [mBq/kg fresh mass]			
^{134}Cs	<125	<68	<125	<1125
^{137}Cs	<150	<75	<150	125±32
^{226}Ra	198±53	200±150	<375	<350
^{232}Th	125±75	75±58	188±55	105±38
^{40}K	(272±8)x10^3	(133±4)x10^3	(158±7)x10^3	(115±6)x10^3

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