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Studies of Radioactive Contaminations and Heavy Metal Contents in Vegetables and Fruit from Lublin, Poland

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Abstract

This paper presents studies of the level of some gamma radioactive elements and heavy metals in fruits, vegetables and plants from Lublin.

Potassium ^{40}K isotope was the most prevalent element in the examined samples. It concentrated mainly in aboveground parts of some vegetables, for example in parsley and carrots haulm and in leaves of red beet and leek (from 1135 to 1940 Bq/kg). Considerably lower concentrations of this element were noticed in the roots of the vegetables, ranging from 210 to 448 Bq/kg of dry matter. In examined fruit, the ^{40}K contents ranged from 490 to 510 Bq/kg.

Transfer factors of ^{40}K, from the soil to the vegetables and fruit, ranged from 0.3 to 2.9.

The natural isotopes of uranium series account for 17% of total activity, whereas thorium series was 19-20% of this activity. In fact, in examined fruit (raspberry, red and black currants) and roots of the vegetables caesium ^{137}Cs was not detected, whereas some amounts of it were noticed in green parts of vegetables, from 4.0 to 8.4 Bq/kg of dry matter. The transfer factor of ^{137}Cs from the soil to examined samples ranged from 0.03 to 0.4.

In all studied samples examined on heavy metal contents no valid safely standards for these elements were exceeded.

Keywords: radioactive isotopes, soil, plant, vegetables

Introduction

Global pollution by radioisotopes come to Earth in the latter part of '50s and at the beginning of '60s. Then, many nuclear explosions were executed in upper layers of the atmosphere [1]. Radioactive nuclides, produced due to these explosions, formed clouds that moved across the whole globe and settled down as radioactive fallout.

Immediately after the atom bomb explosions the high concentration of the following isotopes was detected: ^{141}Ce, "Mo, ^{133}Xe, ^{140}Ba, ^{140}La, ^{131}I, ^{132}I, ^{133}I, ^{,43}Pm, ^{132}Te, ^{90}Sr, ^{137}Cs, ^{106}Rh, ^{144}Ce, ^{95}Zr. In time, long lived isotopes stayed, among them the most dangerous for human beings ^{90}Sr and ^{137}Cs [2]. Radioactive fallout contaminated the entire environment. These radioactive elements are concentrated mostly in the surface layers of soil as their migration downwards is limited and depends on the many chemical and physical conditions of the soil system [3,4].

The low mobility of radioactive elements in soil holds them in the root zone. Plants assimilate the radioactive substances with others necessary for their growth, then dangerous isotopes may get into animal tissues and finally as food into organisms of human beings.

The breakdown of the Chernobyl power plant in 1986

Corespondence to: S. Chibowski; radcheml@hermes.umcs.lublin.pl liberated huge amounts of radioactive substances that caused global pollution. It was the greatest release of radionuclides in the history of nuclear energetics. Fortunately, the territory of Poland was not contaminated too much.

It is well known that the most dangerous isotopes are absorbed by organisms by nutrition and much less by inhalation. In this group the most dangerous are radioactive isotopes of iodide, caesium and strontium.

Considering that Poland (especially its eastern territory, including Lublin), is in a dangerous zone. Our studies deal with determination of the gamma radioactive isotopes in the samples from Lublin. The influence of Chernobyl and test explosions on the radioactivity of soils and cultivated plants was learned. Also, the influence of industrial dusts on the concentration of natural radioactive isotopes was learned. From gamma spectrometry of the samples we calculated soil-plant transfer factor for ^{137}Cs. The measurements of heavy metal contents were done to achieve a detailed picture of the Lublin regional environment.

Experimental

Collection and Processing of the Samples

All plants and soil samples were collected in allotment gardens located in Lublin, according to the procedure recommended by International Atomic Energy Agency (IAEA) [5]. Plant samples consist of fruit and vegetables (leguminous plants, deciduous plants and roots). In the group of leguminous plants their stalks, beans and pods were analyzed, whereas for carrots and garden beets their roots and leaves were. For the first stage, all plant samples were washed carefully to remove soil. The leaves were air dried and ground in a mortar. Roots were cut and dried at 105°C. Currants and raspberries were dried at 105°C and then combusted in the furnace.

Soil samples were dried at room temperature, then in a drier at 105°C. After that the soil sample was grounded in mortar and sieved by lmm sieve, to remove stones and organic parts.

0.5 dm^3 of the sample was weighed and placed into the standard Marinelli vessel, enabling constant geometry of the measurement system. A qualitative and quantitative analysis of the samples was made with a gamma spectrometer.

The concentration of heavy metals in soil and plant samples was measured by XRF method. In the samples the following elements were detected: K, Ca, Mn, Fe, Cu, Zn, Br, Sr, Y, Mo and Pb.

Methods and Apparatus

Gamma radioactivity of the samples was measured with a Silena spectrometer (Italy), equipped with IGC-13 germanium cylindric detector by Princeton Gamma Technology, cooled with liquid nitrogen. Detector resolution was 1.75 keV at 1.33 MeV at 15% of relative capacity. The spectrometer worked with a 4096 channel analyzer with SIMCAS 4.11 software allowing qualitative and quantitative analysis of the obtained data. Measured energy ranged from 50 keV to 2.1 MeV, enabling us to identify and measure most of the natural and artificial gamma isotopes. The standard deviation of the radioactivity measurements ranged from 0.4 to 30 Bq/kg of the sample, depending on the total radioactivity of the isotope in the sample. The measurement error was smaller than 5-7%. Samples were measured over 1200 min. Results are calculated in Bq per kg of the sample for each isotope.

To analyze heavy metal contents the XRF method was applied. The XRF spectrometer by Canberra was equipped with a Sj(Li) detector and a/d conversion Canberra 1510 detector. X-ray radiation was induced by ^{109}Cd, ^{55}Fe and ^{241}Am. Quantitative analysis was done with Canberra S-100 and AXIL software.

Results and Discussion

In examinations of the environment attention focus on the issue of soil contamination. The existence of radionuclides in soil and their possible transfer to plants is a significant problem. The transfer factor depends not only on the type of plant but also on the binding mechanism of the isotope by the soil system [6, 7]. These studies also have practical aspects, as they allow us to find places of isotope accumulation and evaluate the possibility of the isotope transfer into the food chain.

Obtained results are presented in Tables 1-3.

Tables 1 and 2 show that ^{40}K, from the group of natural isotopes is the main source of soil contamination. It gives 60-65% of total radioactivity irrespective of the sample site. Such amounts of ^{40}K result from the natural concentration of potassium in the earth's core. Radioactive isotope state 0.0119% of total potassium contents, making 370 Bq/kg of the soil [8]. The mean radioactivity of ^{40}K in Lublin region soils ranged from 450 to 500 Bq/kg [9]; meanwhile in examined ones radioactivity was much higher and ranged from 595 to 670 Bq/kg. Increased levels of radioactivity may be caused by mineral fertilizing and contamination of soil by dusts from coal incineration of the industrialized Lublin region. The concentration of radioactive potassium in fruit and vegetable samples was higher. For samples consisting of leaves or haulm it achieved the following values: leek 1,940 Bq/kg of dry weight, parsley - 1,863 Bq/kg, garden beet - 1,263 Bq/kg, carrot - 1,135. Much lower concentrations were observed in the roots of these vegetables. They ranged from 210 Bq/kg for leek to 448 Bq/kg for garden beet. Carrots were an exception with 943 Bq/kg. Potassium ^{40}K activity, for string beans measured in stalks, pods and beans was 418, 892 and 531 Bq/kg respectively. A similar level of radioactivity was detected in raspberries - 490 Bq/kg, black-currants 510 Bq/kg and red-currants 465 Bq/kg. All values are calculated for kg of dry weight.

Analysis of the above data and calculated from soil to plant transfer factors of ^{40}K isotope show that relocation of potassium depends much on the kind of

Table 1. Radioactivity content of ^{137}Cs and ^{40}K and their transfer factor from soil to the plant (TF) for examined samples.

Sample	^{137}Cs radioactivity [Bq/kg]	TF – ^{137}Cs	^{40}K radioactivity [Bq/kg]	TF – ^{40}K
Raspberry soil	11.3		613.5	
Raspberry	–	–	490.1	0.8
Currant soil	13.6		595.1	
Black currant	–	–	510.7	0.86
Red currant	–	–	465	0.78
Vegetable soil	22.9		670	
Leek	8.4	0.4	1940.7	2.9
Carrot (root)	–	–	943.6	1.4
Carrot (haulm)	1.9	0.083	1135.2	1.7
Garden beet (root)	0.7	0.03	448.6	0.7
Garden beet (haulm)	3.95	0.17	1263.6	1.9
Parsley (root)	–	–	310	0.5
Parsley (haulm)	7.5	0.33	1863.6	2.8
Celery (root)	–	–	210	0.3
Celery (haulm)	–	–	353	0.5
Bean (stalks)	3.9	0.17	418.8	0.6
Bean (pods)	4.4	0.19	892	1.3
Bean (grains)	–	–	531.1	0.8

Table 2. Concentration of radioisotopes in soil samples.

Soil sample	Radioactivity [Bq/kg]												
	^{134}Cs	^{137}Cs	^{40}K	^{228}Ac	^{212}Bi	^{214}Bi	^{212}Pb	^{214}Pb	^{226}Ra	^{228}Th	^{234}Th	^{208}Tl	Σ
Raspberry soil	< 0.6	11.3	613.5	35.1	38.5	28.2	35.3	29.7	72.7	78.4	41.9	12.5	997.7
Currant soil	< 0.6	13.6	595.1	34.6	37.9	27.2	34.0	28.3	68.3	86.8	42.7	12.6	981.7
Vegetable soil	< 0.9	22.9	670.0	36.9	44.9	29.1	38.1	29.4	80.3	70.1	44.9	21.8	1089.3

Table 3. Heavy metal concentration in ppm or % of dry sample.

Sample	K (%)	Ca	Ti	Mn	Fe (%)	Cu	Zn	Rb	Sr	Y	Zr	Nb	Pb	Mo
Raspberry soil	2.6	6974.0	4911.9	499.5	1.6	42.3	47.9	75.2	86.2	28.9	625.6	9.2	33.4	–
Raspberry	1.7	1791.1	–	38.6	–	10.5	39.9	3.5	9.7	2.2	–	–	–	4.1
Currant soil	1.3	5097.8	3782.8	406.2	1.4	33.2	47.7	72.8	82.2	25.9	587.8	9.3	30.3	–
Red currant	1.8	2849.7	–	–	–	26.5	22.6	8.3	11.8	1.4	–	–	–	2.8
Vegetable soil	2.6	9529.5	4214.9	411.9	1.7	45.1	75.1	76.1	85.6	30.1	561.3	9.0	37.5	–
Bean (rains)	1.8	1283.2	–	–	36.8	17.3	39.5	5.9	5.9	1.4	–	–	–	4.6
Carrot (root)	3.0	3268.2	–	–	37.2	27.6	41.4	9.4	20.4	2.5	–	–	–	1.8
Parsley (root)	3.1	3312.1	–	30.2	27.4	24.5	35.1	10.8	26.8	1.5	–	–	–	1.4
Garden beet (root)	3.0	1341.7	–	61.2	74.9	17.6	59.5	11.6	16.7	1.7	–	–	–	1.5
Garden beet (leaves)	4.9	1.7	–	90.7	297.1	15.6	45.6	32.3	62.7	1.4	11.5	–	–	4.3

plant. Transfer factors, calculated for plants cultivated on the same soil ranged from 0.3 for celery roots to 2.9 for leek.

The other natural isotopes which form the radioactivity of soil are ^{214}Bi, ^{214}Pb, ^{226}Ra and ^{234}Th from the uranium series and ^{228}Ac, ^{212}Bi, ^{212}Pb, ^{228}Th and ^{208}T1 from the thorium series. The isotopes from the first series form 17% of total radioactivity whereas the second ones give 19-20% of it. The concentrations of uranium and thorium series isotopes in three examined soils were almost the same irrespective of their collection site. (Tabs. 1 and 2). Because the natural isotope activity, ^{226}Ra 70-80Bq/kg, ^{234}Th 43 Bq/kg and ^{228}Th 70-80 Bq/kg - is relatively higher than in uncultivated areas of Lublin region, [9] one can assume some connections with fertilization, such as for ^{40}K. Mineral fertilizers contain some isotopes that from ^{238}U and ^{232}Th series [10]. The industrial activity of the town also may have an influence on the increased amount of these radionuclides in the soil. Although relatively high amounts of natural isotopes are in the soil, their concentrations in examined fruits and vegetables is minute except for ^{40}K. This isotope, as mentioned earlier, is responsible for radioactivity levels of the plant samples. Only in black-currants were 5.0 Bq/kg of ^{214}Bi and 4.9 Bq/kg of ^{214}Pb detected. In other examined samples the concentration of natural isotopes (excluding ^{40}K) was at the detection level.

Considering radioactive contamination of the environment the most dangerous isotopes are ^{134}Cs and ^{137}Cs and they are used as an environment contamination index. ^{137}Cs is particularly dangerous, even if one considers its half-life period ($T_{1/2}$ = 30 years). The second isotope of caesium, ^{134}Cs, is less dangerous now ($T_{1/2}$ = 2.06 years) and its concentration in the environment is meaningless. This isotope could be detected only in a few samples.

Examined samples of the surface soil (down to 10 cm) contained the following amounts of ^{137}Cs: 11.3, 13.6, and 22.9 Bq/kg of the dry matter (Tabs. 1 and 2). These data confirm our previous studies [9, 11], discovering that caesium is firmly adsorbed by soil particles. Considering soil-plant transfer factors for ^{137}Cs we can say that there is no infiltration of this isotope into the roots and fruit of the plants. Only in garden beet roots was ^{137}Cs detected (0.7 Bq/kg). Some amounts of the radioactive caesium were observed in green parts of vegetables, for example parsley haulm 7.5 Bq/kg, leek 8.4 Bq/kg and beet leaves 4.0 Bq/kg. However, these concentrations are meaningless and do not cause an introduction of the extra amounts of the radioactive caesium to a human being.

The consumption of caesium from the soil by some plants is probably connected with its high chemical similarity to potassium so that they may form similar sorption complexes in the soil. This analogy suggests that elements are complementary to another and potassium shortage may be supplemented by more dangerous caesium. From the analysis of the obtained data one can see that caesium transfer to the vegetable and fruit samples is complicated. It is absent in roots and well detected in leaves. As there is no radioactive caesium fallout now, its presence in green parts of vegetables must originate from the soil. Considering the radioactivity in roots and leaves, one can say that roots do not adsorb radioactive caesium but only forward it from the soil up to the leaves.

^{137}Cs transfer coefficient was calculated from the relation:

$$\mathrm{TF} = \frac{\text{caesium activity in sample [Bq/kg]}}{\text{caesium activity in soil [Bq/kg]}}$$

In our studies this ranged from 0.03 for garden beet roots to 0.4 for leek (Tab. 2). These values are much higher than for grass [9,12] or corn [13]. Obtained results confirm that roots of vegetables and some fruit like currants or raspberries do not accumulate caesium from the soil. Observed accumulation in green parts of vegetables is meaningless and these segments may be applied as safe green fodder.

Heavy metals, omnipresent in our environment, are very toxic when in excessive concentrations. Therefore, all collected samples were examined for these elements. Obtained results are presented in Table 3. In fruit (raspberries, currants) the mostly met were Cu, Zn, Rb, Sr, Y and Mo and in vegetables Fe, Cu, Zn, Rb, Sr Y, Mn and Mo. The detected concentrations of these elements were below safety standards for fruits and vegetables [14], and they were connected with metabolism of the plants.

Particularly dangerous elements, which reflect the contamination of the environment are Cd, Pb and Cr. Among them, only the lead was present and only in the soil samples in the concentrations below Polish safety standards [14]. From Table 3 one can judge that heavy metal transfer from the soil to plant does not depend on its concentration in the soil but only on the type of plant. The most striking is iron, whose concentration in parsley root was 27.4 ppm and garden beet root 74.9 ppm, whereas in garden beet leaves as far as 297.1 ppm. However, in all examined samples the heavy metal concentrations do not differ much from literature data [14].

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