Polish Journal of Environmental Studies Vol. 8, No. 1 (1999), 19-23

Examination of Radioactive Contamination in the Soil-Plant System and Their Transfer to Selected Animal Tissues

S. Chibowski, A. Gładysz

Department of Radiochemistry and Colloid Chemistry, Faculty of Chemistry, Maria Curie Skłodowska University, 20-031 Lublin, PI. M. Curie Skłodowskiej 3, Poland

Received 16 July, 1998
Accepted 8 October, 1998

Abstract

This paper investigates gamma emitter radioactivity in a system consisting of soil and plants. Some selected samples of tissues of animals fed with the plants from these sites were also measured.

In soil and plant samples artificial (^{137}Cs, ^{134}Cs) and natural (thorium and uranium series) isotopes were detected. Despite the relatively high content of the natural isotopes in plants and their seeds, their accumulation in animal tissues was not detected. The ^{40}K isotope was transferred in the chain soil-plant-animal in the highest degree. From the group of the natural isotopes, only ^{212}Pb was detected in examined animal tissue samples. Other natural isotopes were below detection level. In the samples heavy metal content was also examined. In any sample no clement concentration was noticed above trade acceptable limit.

Keywords: radioactivity, soil, plants, food, contamination

Introduction

The equilibrium in the environment has become disturbed because of the expansion of technical civilization accompanied by intense application of chemicals. In the second part of our century violent growth of the population has increased food demand, causing intensive use of fertilizers in agriculture. The continuous increase of applied chemicals has produced many problems due to its harmful influence on animals and human beings.

In recent years, examinations of radioactive isotopes in the environment and their role for living things of our planet has attracted growing attention.

Beside its natural presence radioactivity is connected with nuclear detonations most of which happened in the '60s and nuclear energetic development [1, 2].

Apart from some advantages that man can get from the existence of radioactivity, there are many drawbacks and dangers connected with contamination of the environment by these substances. Radiation hazards caused by radioactive elements result from willful, intentional or unintentional, and sometimes thoughtless activity of man. Most often it is connected with unsuitable procedures for radioactive waste, nuclear power plant failures, exploitation and processing of radioactive ores, nuclear detonations, and fossil fuel utilization. As a result, the concentration of radionuclides in the environment has increased. This may threaten living organisms by entering the food chain from plants to animals and, finally, to man.

As radioisotopes spread in the whole ecosystem of the Earth they do not produce an essential problem for the human beings. Meanwhile, some isotopes that enter the food chain may achieve concentrations that may be toxic for plant or animal organisms or their consumers.

Because a higher concentration of radioactive substances in the environment is undesirable, this paper presents some investigations of gamma emitting elements in environmental samples. Samples consist of soils and plants from the same sites (growing on them). To obtain comprehensive information of radionuclide transportation in the environment, some animal tissue samples coming from animals fed with examined plants were measured.

Methods

Soil samples, plants growing on this soil, and animal tissue samples were collected for measurement. The soil and plant samples were collected at three places, at Kanie, Fajslawice, and Elizowka villages in the vicinity of Lublin in middle-eastern Poland, according to the procedure recommended by IAEA (International Atomic Energy Agency) [3].

Soil samples were initially dried at room temperature and

Table. I. The radioactivity of selected soil and plant samples in [Bq/kg].

Site	Isotope	Soil of potatoes	Potato tubers	Potato straws	Soil of wheat	Wheat grains	Wheat straws	Soil of rye	Rye grains	Soil of oat	Oat grains
Elizówka	^{228}Ac	31.2	–	9.1	33.4	–	–				
	^{7}Be	–	–	90.3	–	–	14.9				
	^{134}Cs	–	–	–	0.8	–	–				
	^{137}Cs	21.4	0.72	3.7	22.9	–	2.7				
	^{40}K	653.6	801.9	870.9	667.2	165.9	2143.3				
	^{210}Pb	36.7	10.9	61.6	37.1	–	56.2				
	^{212}Pb	31.5	1.7	2.4	34.2	0.3	7.0				
	^{214}Pb	18.6	1.2	4.5	21.8	–	2.0				
	^{226}Ra	67.9	16.8	37.6	66.3	26.9	79.3				
	^{228}Th	48.6	22.3	49.5	46.1	–	112.0				
	^{234}Th	41.5	19.8	43.4	39.8	17.7	79.2				
	^{208}Tl	11.4	–	2.5	12.2	–	–				
	^{210}Tl	–	–	1.2	–	0.5	1.9				
	^{212}Bi	33.5	–	–	37.6	–	–				
	^{214}Bi	20.2	–	–	23.8	–	–				
Fajsławice	^{228}Ac	11.4	–	7.6	25.08	–	9.2				
	^{7}Be	–	–	135.1	–	–	51.3				
	^{134}Cs	0.2	–	–	0.8	–	–				
	^{137}Cs	16.1	1.9	8.3	35.8	0.54	7.2				
	^{40}K	251.8	521.1	504.8	406.8	165.2	405.3				
	^{210}Pb	13.8	9.5	85.1	31.4	11.2	50.4				
	^{212}Pb	11.6	0.4	7.6	26.1	1.8	3.0				
	^{214}Pb	6.2	1.1	3.8	17.7	0.4	4.0				
	^{226}Ra	25.3	6.1	26.2	58.6	6.8	35.6				
	^{228}Th	14.6	–	14.3	30.6	–	29.6				
	^{234}Th	13.9	7.2	9.2	38.1	12.6	27.6				
	^{208}Tl	3.9	–	2.8	9.2	–	2.1				
	^{210}Tl	–	–	–	–	0.4	1.0				
	^{212}Bi	9.8	–	–	28.7	–	–				
	^{214}Bi	6.6	–	4.8	17.6	–	–				
Kanie	^{228}Ac	–	–	–	14.76	–	–	8.5	–	9.1	–
	^{7}Be	–	–	–	–	–	–	–	1.9	–	4.23
	^{134}Cs	–	–	–	–	–	–	0.1	–	0.2	–
	^{137}Cs	–	0.5	–	9.8	0.23	–	11.9	0.4	9.8	1.0
	^{40}K	–	788.6	–	287.2	155.1	–	210.6	152.2	219.9	162.7
	^{210}Pb	–	5.5	–	25.3	10.7	–	14.2	6.6	14.1	13.2
	^{212}Pb	–	0.9	–	15.0	0.3	–	8.9	1.1	9.3	1.72
	^{214}Pb	–	1.6	–	8.7	–	–	5.6	0.6	5.7	–
	^{226}Ra	–	12.1	–	36.7	9.8	–	25.6	6.0	28.9	17.0
	^{228}Th	–	13.9	–	18.3	12.4	–	16.5	18.8	11.5	27.3
	^{234}Th	–	11.0	–	5.4	12.6	–	17.5	12.7	17.7	20.5
	^{208}Tl	–	–	–	5.2	0.2	–	3.1	–	3.4	–
	^{210}Tl	–	–	–	–	0.3	–	–	0.2	–	0.4
	^{212}Bi	–	–	–	16.1	–	–	9.2	–	9.8	–
	^{214}Bi	–	1.6	–	9.7	–	–	6.4	–	6.0	–

then at 105°C. The dry soil sample was crushed in mill and sieved by 1 mm sieve to remove mechanical waste.

Plant samples consisted of wheat, oat and rye grains, their green parts as well as potato tubers and stalks. Before measurement the collected grains were air dried only. Green parts of plants were dried at 105°C, to constant weight, and were then crushed in a Fritsch mill. The potatoes were washed and then dried at 105°C and finally crushed in the mill.

Animal tissues consist of pork, poultry and their bones. Additionally, eggs from the place of interest were examined. Meat, liver and bone samples were dried at 105°C to constant weight. Then they were incinerated to ash in the oven by slowly increasing the temperature to 450°C for 20 hours. Eggs were divided into three parts (whites, yolks and shells) and dried to 105°C.

To ensure standardized geometry of the radioactivity measurements, 500 cm^3 of the soil and plant samples were weighed and inserted in a Marinelli vessel. For animal tissues and egg samples special vessel was constructed to keep the same geometry of the measurements.

The results are presented in Table 1.

The animal tissue samples were the subject of heavy metal content measurements with an XRF spectrometer. The examined elements and their concentration are presented in Tab. 4.

Apparatus

The gamma radioactivity measurements of all samples were done with the Silena spectrometer (Italy) equipped with germanium cylindric detector JGC-13 by Princeton Gamma-Tech cooled with liquid nitrogen. The spectrometer is equipped with multichannel analyzer (4096 channels), which enables measurements of gamma radiation energy in the range 50-2000 keV. The resolution of the detector was 1.75 keV at the 1.33 MeV peak, at 15% of relative capacity. That enables identification and measurements of the radioactivity for most of natural isotopes, as well as artificial ones, occurring in the environment because of human activity. Qualitative and quantitative analysis of the results was performed with SIMCAS 4.11 software. The standard deviation of radioactivity measurements ranged from 0.1 to 15 Bq/kg of the sample (depending on total radioactivity of the isotope in the sample and type of vessel). Measurement error was smaller than 5-7%. Samples were measured over 1200 min. Using Curie equation [4| for the blank sample, a lower limit of detection (LLD) and the minimum detectable amount (MDA) values for selected radionuclides were calculated. Obtained LLD values for the examined radionuclides ranged from 0.06 to 0.24 cpm and MDA from 0.13 to 0.40 Bq/kg. For calibration of the spectrometer a radioactive source with eleven isotopes supplied by Amersham was used. Their energy ranged from 59.54 keV to 1836 keV. As a standard, "Whey powder IAEA 154" source was applied. Among others, it contained isotopes ^{134}Cs, ^{137}Cs, ^{40}K, and ^{90}Sr. The spectrometer was tested and calibrated with these samples, enabling us to obtain reliable results of the activity of the examined samples.

XRP spectroscopy was applied to heavy metal content in pork, poultry and bone samples. Our Canberra spectrometer was equipped with Si(Li) detector and a/d converter Canberra 1510. X ray radiation was induced by ^{55}Fe, ^{109}Cd and ^{241}Am. Quantitative analysis was performed with Canberra S-100 and AXIL software.

Results and Discussion

The main task of the investigation was to establish the level of the radioactivity of the gamma emitters in the selected samples of the environment from three sites: Elizowka, Kanie and Fajslawice.

From the data presented in Table I, it can be seen that main radioisotopes responsible for the radioactivity level in soil and plant samples are of the natural origin, mainly $^{4"}K$. Its concentration produces from 62 to 95% of total radioactivity. This is connected with the natural concentration of this isotope in the lithosphere that is 0.0119% with stable potassium [5], producing 370 Bq/kg for the average soil sample.

A higher radioactivity was observed for all samples from Elizowka, because of the high contents of ^{40}K (65-91%) in the green parts of corns (wheat) 2143 Bq/kg of dry weight, in green parts of potatoes - 870 Bq/kg and their tubers - 801,9 Bq/kg. A lower-to-the-mean value (^{40}40 Bq/kg) was observed in soil samples from Kanie. Also, samples of corn grains from this place revealed rather low activity of the ^{40}K isotope.

Obtained results proved that the amount of derived potassium depends on its content in the soil, though final results, as is suggested, depend on many other factors [6, 7]. We think, however, that this parameter is more important than others. Furthermore, potassium being the fundamental element for the growth of the plant is present in all their parts, including grains.

The other isotopes (which compose the radioactivity of the examined soils) were as follows: ^{214}Bi, ^{214}Pb, ^{210}Pb, ^{226}Ra, ^{234}Th, ^{210}Tl from uranium series, and ^{228}Ac, ^{212}Bi, ^{212}Pb, ^{208}Tl, and ^{228}Th from thorium series. The radioisotopes from uranium series make from 4 to 21% of the total radioactivity of the sample, whereas thorium series give from 1 to 15%. The number and the amount of the isotopes depend on the geochemical and soil formation processes but also on atmospheric fallout, dust and ash that come from coal combustion. One kg of coal contains about 20-40 Bq of ^{40}K, ^{238}U and ^{232}Th isotopes and their derivatives. After combustion their concentration increases in volatile ashes and dust, reaching even 100 times higher activity for ^{210}Pb [8]. The additional factor, which increases the activity of these isotopes in soil and plant samples, is made by mineral fertilizer application as some fertilizers contain natural radioisotopes.

From the obtained results concerning isotope activity in surface layer of the soil one can see the influence of the big agglomeration on the radioactive contamination of the soil. This very distinct example is Elizowka; a village near large, industrial Lublin. The natural isotope content was noticeably higher than that taken from other examined soils far from cities and factories. It should be mentioned that all soil samples were the same type and grade.

Beside ^{40}K, other isotopes also sometimes penetrate from soil into plants in an appreciable degree (Tab. 1). This is visible especially for such isotopes as ^{226}Ra, ^{228}Th or ^{234}Th. The greatest detected amounts of these isotopes were seen in wheat straws and potato stalks, though potato tubers and corn grains also contained some of them.

Similarly to soils, greater radioisotope content was observed for Elizowka. As previously mentioned, most probable is the influence of the Lublin vicinity, as these isotopes appear as a result of industrial activity. More activity of

Table 2. Soil-plant transport coefficients of ^{137}Cs.

Site	Type of sample	TF
Fajsławice	Potato tubers	0.12
	Potato stalks	0.51
	Wheat grains	0.015
	Wheat stalks	0.20
Kanie	Wheat grains	0.023
	Oat grains	0.10
	Rye grains	0.032
	Potato tubers	0.04
Elizówka	Potato tubers	0.03
	Potato stalks	0.17
	Wheat grains	–
	Wheat straw	0.12

radionuclides originate from atmospheric fallout than from their penetration from the soil.

The cosmogonic radioisotope that was present in examined plant samples was ^{7}Be. This isotope falls from the atmosphere and is adsorbed on leaves and the aboveground part of plants. This isotope was found only in green parts of all plants. It consists of from 7 to 16% of the total radioactivity of these samples. Because of its properties and short half-life, this does not cause any serious problem in radioactive contaminations of the plants.

The main indicator of the contamination of the environment is the presence of ^{137}Cs and ^{134}Cs isotopes. ^{137}Cs, which has appeared because of power plant failures and nuclear detonations is a very important and dangerous isotope, even if one considers its half-life equal to 30 years. For comparison, the half-life of ^{134}Cs equals 2.06 years [9]. The contamination of samples by these isotopes is relatively low, with higher values observed for soils than for plants for all examined sites. This proves the durability of caesium adsorption in the soil particles [10, 11]. This isotope content in soil samples ranges from 10 to 35 Bq/kg, accounting for 2 to 4% of total activity of the soil. Analyzing obtained data concerning the transfer of ^{137}Cs from the soil to green parts of plants, it should be noticed that all samples reveal the transportation of this isotope. To calculate the transfer coefficient of this isotope the following equation was applied.

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

The greatest values of the transfer factor (TF) were observed for potato stalks, and reached 0.51 for Fajstawice (see Tab. 2). It is worth mentioning that caesium contents in the soil is not the main factor responsible for its transfer from soil to plants (see Tab. 1).

The potassium content in soil, as the biologically active element of the same group as caesium, does not have clear influence on the caesium transfer from soil to plants. Obtained results confirm literature data of some factors that influence the transfer. Among them, the most important are pH of the soil solution, soil humidity, type and grade of the soil, degree of soil dispersion, form of caesium, the concentration of the ions that can compete with caesium at adsorption or complex formation, and many others.

Obtained data confirm that the amount of transferred caesium from the soil to the plant is small and ranges from 0.2 to 3 Bq/kg. The only higher values, 8 Bq/kg were obtained for Fajslawice in straws and potato samples. The above results suggest uniquivocally that ^{137}Cs present in the soil does not have a noticeable influence on the total amount of radionuclides transferred to human organisms. The transfer of radionuclides from soil to plants is of the basic meaning for the next stages, the transfer of radionuclides to animals, and finally to humans. Thus, the transportation of large amounts of ^{40}K from soil to plant has a straight influence on its contents in the animal tissues. The isotope ^{40}K accounts for 80 to 100% of the total gamma radioactivity in liver samples of poultry, where total activity reached 1180.8 Bq/kg of the ash sample, 1293.9 Bq/kg for poultry meat and 3026 for pork meat. Such high radioactivity results from ^{40}K, which reaches 0.01% of total potassium content. This amount in the organism is still filled, as potassium is present in any food.

The animal tissue samples revealed the low level of radioactivity coming from other isotopes. Only ^{212}Pb and ^{137}Cs were detected. The latter was present in poultry bones 1.2 Bq/kg and egg shells 1.8 Bq/kg. Radioactive ^{212}Pb was present in the same samples with activity ranging from 3 to 18 Bq/kg.

A comparison of radioactivity of soil, plant and animal tissue samples fed with these plants allows us to say that despite some transfer effects of some radioisotopes to the animal organisms, isotope accumulation excluding ^{40}K is small. A high amount of accumulated potassium results from its high contents in green parts of the plants, grains and potato tubers. Besides, this isotope is easily available via miscellaneous animal tissues. Other natural isotopes were not accumulated in selected animal tissue samples. The artificial isotope ^{137}Cs was detected in poultry bones and egg shells. From the data of Table 3, one can say that a diet which consists of meat and eggs does not introduce a meaningful amounts of radioactive isotopes.

Table 3. Radioactivity of selected samples of the animal tissues in [Bq/kg].

Type of sample	^{40}K	^{137}Cs	^{210}Pb	^{212}Pb	^{214}Pb
Egg whites	282.84	–	< 1.326	< 1.856	–
Yolks	51.26	–	–	2.965	< 1.373
Egg shells	28.284	1.759	–	2.423	–
Poultry meat	1293.3	–	–	7.887	< 3.815
Poultry liver	180.9	–	–	18.383	< 13.74
Poultry bones	55.838	1.23	–	3.04	< 1.163
Pork	3026.6	< 1.397	–	–	–
Pork liver	720.4	–	–	–	–
Pork bones	477.0	–	–	–	–

Because heavy metals toxic to living organisms are omnipresent in our environment, their content also was examined for animal tissue samples. The results are presented in Table 4.

Table 4. Heavy metal contents in selected samples of animal tissues in [ppm].

Type of sample	Fe	Cu	Mn	Sr	Cd	Pb	Cr	Zn
Poultry bones	220	11.4	9.77	406	< 9.5	10.5	< 7.2	320
Poultry meat	126.5	37	< 13	–	< 8.4	5.27	< 2.2	192
Poultry liver	1367	53.5	34	–	< 8.4	< 2.7	< 1.7	357
Pork bones	89	20.2	3.30	318	< 9.5	13.5	< 7.5	249
Pork meat	1100	–	–	–	–	23.2	–	–
Pork liver	4051	70.9	< 16	–	< 9.5	5.1	< 25	713

The highest concentration in all samples was noticed for iron. This results from its significance for living organisms (as is true of Zn and Cu). Both these elements are beneficial for the proper work of the organisms. The trade acceptable **limit** of the concentration of these elements in meat is 80 mg/kg for Zn and 8 mg/kg for Cu [12].

The presence of such metals as Cd, Pb and Cr proves that contamination in the environment is harmful for organisms due to their carcinogenic properties. Acceptable concentrations in meat and their products are: Cd - 0.05mg/kg and Pb - 0.70 mg/kg [12]. From the data presented in Table 4, it can be seen that for most of the samples the obtained data were very low or below lower detection limits of the method applied. The other element, detected only in bones of poultry and pork, was strontium. This is because it is built in bones as a calcium equivalent.

In summary, it should be mentioned that heavy metal concentrations in all examined samples were lower than trade acceptable limits.

References

1. JAGIELAK J., BIERNACKA M, GRABOWSKI D., HENSCHKE J., Zmiany sytuacji radiologicznej srodowiska Polski w okresie 10 lat po awarii w Czarnobylu, Panstwowa Inspekcja Ochrony Srodowiska, Biblioteka Monitoringu Srodowiska, Warszawa p. 7, **1996.**
2. HRYNKIEWICZ A.Z., Dwa oblicza energii jadrowej, Sympozjum Promieniowanie - Historia i Wspolczesnosc, Lodz 24-25.04. **1997** p. 21-36.
3. IAED, Measurement of Radionuclides in Food and the Environment - Technical Reports, Ser. No. 295., IAEA, Vienna, p. 5-27, **1989.**
4. CURRIE L.A.. Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry, Anal. Chem. **40,** 586, **1968.**
5. United Nations Scientific Committee on the Effects of Atomic Radiation: Ionizing Radiation Sources and Biological Effects, New York, **1982.**
6. **SAAR** A. **R.,** WEBER J.H., Fulvic Acid: Modifier of Metal-Ion Chemistry, Environ. Sci. Technol., **16,** 510A, **1982.**
7. LEMBRECHTS J. A Review of Literature on the Effectiveness of Chemical Amendments in Reducing the Soil to Plant Transfer Radiostronlium and Radiocaesium, Sci. Total Env. **137,** 81, **1993.**
8. CELINSK1 Z., Energetyka jadrowa, a spoleczeristwo, Wydawnictwo Naukowe PWN, Warszawa, p. 54, **1992.**
9. BROWNE E., FIRESTONE R.B., (Ed. Shirley V.S.), Table of Radioactive Isotopes, John Wiley and Sons, New York, p. 131-3, 137-43, **1986.**
10. ZYGMUNT J., CHIBOWSKI S., KLIMOWICZ Z. The Effect of Sorption Properties of Soil Minerals on the Vertical Migration of Caesium in Soil, J. Radioanal. Nuclear Chem. **231,** 57, **1998.**
11. ZYGMUNT J., CHIBOWSKI S., KLIMOWICZ Z. Studies of Radiocaseium Migration in Soils with High Organic Matter Content, Polish J. Environ. Studies, **6,** 57, **1997.**
12. Projekt Rozporzadzenia Ministra Zdrowia i Opieki Spolecznej, Zalacznik Nr 5, **1998.**