

Potassium-40 and Cesium-137 in the Surface Layers of Arable Soils and Food Supplies

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Abstract

The concentrations of ¹³⁷Cs and potassium in the surface layers of arable soils were analyzed in the selected region of Eastern Poland approximately 20 years after the Chernobyl power plant accident. Additionally, the occurrence of the elements in soil in the forms available for plants was determined. In relation to the total concentrations of ¹³⁷Cs and ⁴⁰K in the soil, cesium is found in larger quantities in the forms available for plants than potassium. Radiation doses through ingestion were estimated on the basis of ¹³⁷Cs and ⁴⁰K measurements in food samples. It was proved that during the period 1998-2007 the annual average intake of radiocesium per person exceeded twofold the national average, whereas the intake of potassium satisfied the body's daily demand for this element.

Keywords: arable soils, food supplies, radiocesium, potassium, Eastern Poland

Introduction

The measurement of ¹³⁷Cs concentrations in food supplies [1-3 and others] is part of the monitoring project that has been carried out in Poland since the Chernobyl accident in 1986. At present the concentration of ¹³⁷Cs in food samples is mainly determined by its concentration in the soil. One of the factors influencing cesium mobility in different soil types is their granulometric composition. The isotope shows a strong tendency to stably bond with the inorganic fractions of soil [4-6]. Cesium is particularly strongly fixed by colloidal clay fractions. Soils that contain more sand are characterized by the increased availability of radiocesium. The other factors influencing cesium fixation in the soil are pH and the monovalent cation content, e.g. NH₄⁺ and K⁺ [6-8]. The solubility and mobility of cesium increase considerably as the pH values diminish [6, 9, 10]. The presence of K⁺ ions can visibly lower the level of Cs⁺ ion absorption [11].

For instance, fertilizing the soil with K⁺ ions can reduce cesium transfer into plants by as much as 60% [12]. Radiocesium concentration in plants, as opposed to ⁴⁰K concentration, is reported [13] to increase with a rise in the carbon content of organic compounds in the soil, although this correlation is not strictly proportional. An increase in the Corg. content in the soil enhances the concentration of the mobile forms of cesium [6].

⁴⁰K and ¹³⁷Cs are isotopes representing elements that are distinguished by only slight differences in their chemical properties but that have different origins in the environment. ⁴⁰K is a natural isotope whose percentage in the total potassium content is estimated at 0.0119 (this value is determined by the constant isotopic ratio) [14]. ¹³⁷Cs was released into the environment in the 20th century as a result of nuclear weapons testing and the Chernobyl power plant accident in 1986. The latter distributed large quantities of ¹³⁷Cs to many parts of Poland, in which the radioactive fallout considerably exceeded the national average. One such place lies in the vicinity of Siedlce [5, 15].

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The main source of ^{40}K and ^{137}Cs for people is food [16]. Regarding food quality, it is essential that potassium supplies from food satisfy the body's daily demand, whereas the ^{137}Cs content is as low as possible. The levels of both radioisotopes in food depend, among other factors, upon their availability in soil.

As the soils in the Siedlce area are rather poor in potassium [17, 18] but relatively rich in ^{137}Cs compared with other areas of Poland, the objectives of our research were firstly to determine the concentrations of potassium and ^{137}Cs in the arable soils in the forms available for plants approximately 20 years after the Chernobyl accident, secondly to find a correlation between the chosen properties of the arable soils and the concentrations of ^{40}K and ^{137}Cs , and thirdly to estimate the doses of ^{40}K and ^{137}Cs intake with contaminated food by the inhabitants of the former Siedlce province during the period from 1998 to 2007.

Research Area

The research was carried out in the former Siedlce province, Poland (within the administrative boundaries up to 1989). The area is situated in east-central Poland at a latitude of $51^{\circ}36'$ to $52^{\circ}42'$ north and a longitude of $21^{\circ}15'$ to $22^{\circ}44'$ east. Almost 70% of the area is taken up by agricultural land, half of which is arable [19]. Altogether, there were 59 measuring positions representing the physiographic characteristics of the area chosen for research. Arable soil samples from the surface layer were taken for laboratory analyses at 12 sites in the spring of 2005 and at 47 sites in the spring of 2006.

Material and Methods

Every sample consisted of 4 cores lying at the corners of a square with the length of a side 1 m. Every core was taken using a steel-tipped tube probe ca 15 cm in diameter from a depth of up to 12 cm. The samples from every site were then quartered, placed in a plastic bag, and submitted for laboratory analysis. First, the soil was dried at room temperature and then sieved through a 2-millimetre mesh to exclude large plant remains or cobble content. Finally, further analyses of the samples proceeded.

The samples were tested for: reaction in 1M KCl, the carbon content in organic compounds using the Tiurin method, and granulometric composition with the areometric method [20]. The samples were classified into granulometric groups and subgroups in compliance with the Polish Standard PN-R-04033 (1998).

In order to determine the concentration of potassium in the arable soils in its forms extracted from the soil with water, absorbed by the soil sorption complex and defined as potentially available, extracts were prepared with redistilled water, 1M $\text{CH}_3\text{COONH}_4$ and 1M HNO_3 , leaving the weight of 2.5 g of the soil in 25 cm^3 of the appropriate extrahent at room temperature for 24 hours. The analyses were repeated twice. Potassium concentration in the extrahents was deter-

mined with the AAS method, using the AAS-30 Carl Zeiss Jena spectrometer and an acetylene-air flame.

In order to estimate the concentration of ^{137}Cs in its water-soluble forms, absorbed by the soil sorption complex, 9 representative soil samples were selected, including 4 loamy sand samples, 2 sand samples and 3 sandy loam samples. The same extrahents as for potassium were used. The weights of 0.5 kg of the soil were extracted with redistilled water and a 1M $\text{CH}_3\text{COONH}_4$ solution at the ratio 1:10. The samples were left at room temperature for 24 h and then filtered through Whatman paper ($\text{Ø} = 0.45 \mu\text{m}$), separating the solution from the solid fraction. The latter was dried at room temperature and analyzed using the γ -spectrometric method. From the difference between ^{137}Cs concentrations in extracted and non-extracted soil samples the isotope was quantified in its forms soluble in water and absorbed in the soil sorption complex.

Food samples of pork, beef, chicken, fish, vegetables (including carrots, beetroots, cabbage, and potatoes), fruit (apples, cherries, sour cherries, plums, pears, and strawberries), and milk were collected at random from shops in the former Siedlce province during the period 1998-2007 by sanitary inspectors. Special care was taken that the samples came from the actual research area. Every sample weighed around 1 kg. Altogether, 416 food samples were tested for ^{137}Cs concentration and 543 samples for ^{40}K concentration.

The concentrations of the isotopes in the soil samples and in the food samples were determined using the γ -spectrometric method. Gamma-ray spectrometry measurements were performed using the Canberra detector integrated with an NaI scintillation detector $2'' \times 2''$. A gamma spectrum analysis was produced using the multichannel analyzer (with 2048 channels) and a Genie 2000 Software package enabling radionuclide identification and quantification. Every sample took 80,000s to measure.

The radiation doses from ^{137}Cs and ^{40}K through ingestion by a statistical inhabitant of the area were estimated on the basis of annual average consumption rates (the data came courtesy of the Statistical Office of Siedlce) of different food categories [in kg]: potatoes – 149, other vegetables – 119, fruit – 28.9, beef – 16.1, pork – 36.8, poultry – 8, fish – 5.4, and milk – 242 dm^3 .

Results

The investigated soils were represented mainly by sandy and loamy soils. The average sand fraction was found to constitute 75.4%, silt fraction – 19.5%, and clay fraction – 5.1% of soils. Loamy sand was represented by 22 samples, weak loamy sand by 12 samples, and sand by 7 samples. The granulometric composition of 15 samples was typical of sandy loam (2 samples represented loam and 1 – silt loam).

The reactions of the arable soils ranged from very acidic to neutral (pH in 1 M KCl: 3.91-7.14). Most soil samples were acidic or very acidic, with $\text{pH} < 6.5$. In this group 10 samples had extremely acidic reaction of less than 4.5. Neutral reaction was found in 7 cases.

Table 1. Spearman correlation factors for ^{40}K activity and different forms of potassium.

Extrahent			^{40}K
	Ammonium acetate	Nitric acid	
Water	0.93***	0.86***	ns
Ammonium acetate		0.87***	0.31*
Nitric acid			0.45***

*** $p < 0.001$; * $p < 0.05$; ns – non significant.

The carbon content in organic compounds in the surface layers of the arable soils ranged between 0.13 and 3.71%, the average being 1.2%.

The average concentration of potassium in the forms extracted from the soil with water was 92.8 mg kg^{-1} , in the forms absorbed by the soil sorption complex (extracted with a 1 M ammonium acetate solution) $164.69 \text{ mg kg}^{-1}$: and in the forms extracted with a 1 M nitric acid solution $272.03 \text{ mg kg}^{-1}$. The average ^{40}K concentration in the soils was estimated at $326.1 (144.0-608.8) \text{ Bq kg}^{-1}$, which equaled approximately 1% of the total potassium content in the soils.

There was a statistically significant correlation between potassium concentrations in the examined soil extracts (Table 1).

The reaction and granulometric composition of the soils were statistically significant factors determining potassium concentration in the surface layers of the arable soils in the forms leached out using three types of extrahents (Table 2). Moreover, a statistically significant positive correlation was found between the occurrence of potassium in the forms extracted with ammonium acetate or nitric acid and the organic carbon content of the soil. The total potassium concentration changed in direct proportion to the increasing soil reaction and organic carbon content. There was definitely more potassium in soils with a greater content of fine fractions (e.g. silt and clay).

^{137}Cs concentration in the arable soils ranged from 4.80 to 40.6 Bq kg^{-1} and averaged $15.47 \pm 7.21 \text{ Bq kg}^{-1}$. The concentration of the isotope in the forms extracted from the soil with water, quantified in representative soil samples, was $0.43 \pm 0.42 \text{ Bq kg}^{-1}$, and that in the forms absorbed in the soil

sorption complex – $0.81 \pm 0.38 \text{ Bq kg}^{-1}$. The radioactivity of the arable soil triggered by the presence of ^{137}Cs did not depend upon the analyzed properties of the soil in terms of statistical significance.

The concentrations of ^{40}K and ^{137}Cs in food supplies varied depending on a food category. Out of the three kinds of meat (beef, pork and poultry), it was poultry that contained the lowest ^{40}K concentration – $74.09 \pm 32.02 \text{ Bq kg}^{-1}$ on average. The estimates in beef and pork were $98.66 \pm 36.94 \text{ Bq kg}^{-1}$ and $97.12 \pm 31.56 \text{ Bq kg}^{-1}$ respectively. Similar concentrations to that in poultry were detected in fish – $75.83 \pm 32.6, \text{ Bq kg}^{-1}$ and in milk – $48.77 \pm 33.04 \text{ Bq dm}^{-3}$. The average ^{40}K concentration in vegetables, excluding potatoes, was estimated at $98.54 \pm 72.93 \text{ Bq kg}^{-1}$. Potatoes contained $120.13 \pm 43.05 \text{ Bq kg}^{-1}$ on average. A relatively low ^{40}K concentration of $58.62 \pm 33.43 \text{ Bq kg}^{-1}$ was detected in fruit, including apples and pears with a ^{40}K concentration of $38.00 \pm 22.68 \text{ Bq kg}^{-1}$.

An individual's annual ^{40}K dose in the former Siedlce province approximated 49.3 kBq. Potatoes were a major contributor to potassium intake (about 18 kBq year^{-1}), comprising almost 37% of the annual total. A comparable concentration, about 24%, was detected in milk and vegetables ($11.8 \text{ kBq year}^{-1}$ in each food category). A ^{40}K concentration of 3.5 kBq was ingested with pork. Fruit and beef contributed to a similar dose – about 3% (ca 1.6 kBq kg^{-1}), whereas poultry and fish comprised no more than 1 % of the annual intake (ca $0.5 \text{ kBq year}^{-1}$).

During 1998-2007 the average ^{137}Cs activity in the selected food categories [Bq kg^{-1}] was as follows: beef – 6.38 ± 6.98 , pork – 2.47 ± 2.25 , poultry – 2.22 ± 2.40 , fish – 2.48 ± 2.03 , vegetables (excluding potatoes) – 3.89 ± 3.35 , potatoes – 1.44 ± 2.45 , fruit – 1.82 ± 2.75 , and milk – $2.48 \pm 3.05 \text{ Bq dm}^{-3}$.

During 1998-2007 the most significant source of ^{137}Cs was milk, which comprised almost 38% of the dose (the annual average intake per person was 600 Bq year^{-1}). Also, potatoes and other vegetables had obvious significance for the ingestion dose – 14% (215 Bq year^{-1}) and 30% (463 Bq year^{-1}) respectively. The average ^{137}Cs intake from beef or pork was about 100 Bq, which made 6% of the total dose, while poultry and fish comprised no more than about 15 Bq. Fruit, with an average ^{137}Cs concentration of 50 Bq year^{-1} , also had little significance for dose. The concentrations of

Table 2. Spearman correlation factors for ^{40}K activity and chosen physical and chemical properties of the soils.

parameter		Extrahent			^{40}K
		water	ammonium acetate	nitric acid	
reaction	pH	0.42***	0.37**	0.47***	0.42***
C org	%	ns	0.27*	0.27*	0.36**
Fraction of soil	sandy	-0.53***	-0.61***	-0.62***	-0.53***
	silt	0.56***	0.65***	0.63***	0.52***
	clay	0.31*	0.33*	0.42***	0.41***

*** $p < 0.001$; ** $p < 0.01$, * $p < 0.05$; ns – non significant.

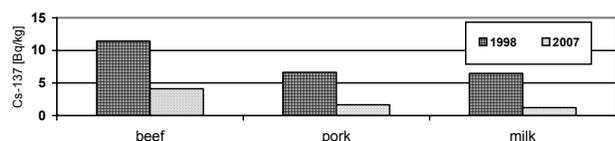


Fig. 1. The average cesium-137 concentration in chosen food categories in the area of investigation in 1998 and 2007.

radiocesium recorded in 2007 were not as high as those detected in the 1990s. The relevant data pertaining to ^{137}Cs activities in beef and pork [Bq kg^{-1}] and in milk [Bq dm^{-3}] registered in 1998 and 2007 is presented in Fig. 1. Twenty years after the Chernobyl accident the classes of food found to be predominant contributors to ^{137}Cs doses through ingestion were beef ($4.12 \pm 4.03 \text{ Bq kg}^{-1}$), pork ($1.65 \pm 0.20 \text{ Bq kg}^{-1}$), and milk ($1.23 \pm 0.22 \text{ Bq dm}^{-3}$). There was a clear downward trend during the ten years of research.

During 1998-2007 the annual average dose through ingestion per person in the Siedlce area approximated 1,555 Bq of ^{137}Cs and to 49,300 Bq of ^{40}K . In 1998 the ^{137}Cs intake was about 4,400 Bq. The predominant contributor to ^{137}Cs dose was milk, which contained 1,568 Bq. In 2007, the ^{137}Cs intake from the ingestion of contaminated milk decreased to a concentration of 300 Bq.

Discussion

Our research made it possible to quantify the concentrations of potassium and ^{137}Cs in the surface layers of the arable soils in the Siedlce area on the one hand and to estimate the intake of both elements through ingestion on the other. The records of total potassium concentration ($326 \text{ Bq of } ^{40}\text{K kg}^{-1}$) in the soils, based on the γ -spectrometric measurements, showed that potassium concentration in the Siedlce area was below the national average, estimated by Biernacka and Isajenko [21] at about 400 Bq kg^{-1} . Research conducted in the 1990s reported over 60% of the soils in the Siedlce area to be deficient in assimilable potassium [17, 18]. However, it is worth emphasizing that the total potassium concentration in the examined soils, estimated at 1% on the basis of ^{40}K measurements, stays within the range of 0.8-1.8% reported by Pondel et al. [22].

Potassium compounds are relatively well-soluble in a soil solution. Potassium availability in arable soils of the area is affected by the content of a fine fraction, soil reaction, and carbon content in organic compounds. A rise in these parameters enhances the concentration of potassium in the forms extracted into a soil solution, absorbed by the soil sorption complex and potentially available for plants, extracted from the soil with a 1 M nitric acid solution. This connection is confirmed by statistically significant positive correlation coefficients. Moreover, similar findings are reported in the literature [23, 24], according to which the concentration of potassium in the soils in the forms including those available for plants, depends upon the mineral composition of the soil, mainly its silt fraction content.

The author's own research also revealed statistically significant positive coefficients of a correlation between the concentration of potassium in the forms examined and the soil reaction. Similar observations on a correlation between the concentration of potassium in its available and replaceable forms in a soil solution and the soil reaction were reported by Kozak et al. [25].

The factors influencing soil abundance in the available forms of potassium include organic fertilization. Literature [26, 27, and others] emphasizes that the largest potassium supply is available for plants in soils systematically treated with manure. These measures additionally contribute to soil abundance in organic carbon. Statistically significant coefficients of a correlation between the concentrations of the different forms of potassium and the carbon content in organic compounds seems to confirm the reports.

The measurement results bear out the fact that post-Chernobyl radioactive contamination persists in the surface levels of the arable soils, although twenty years have passed since the accident and the isotope is being removed from the soil in crops.

The main source of cesium and potassium for plants is soil. The following properties of the soil are reported [6-8, 10, 11, and others] to affect cesium absorption by plants: reaction, the content of replaceable potassium cations and colloidal clay. However, research has revealed no statistically significant correlation between ^{137}Cs concentration and the above-mentioned properties of the arable soils.

The speciation analysis of the selected arable soil samples, carried out as part of the author's own research, showed that, predictably, the concentration of radiocesium in the forms leached out of the soil with distilled water was lower than in the forms extracted with an ammonium acetate solution; they were, respectively, 1.72% and 4.33% of the ^{137}Cs concentration in the soil samples without extraction.

The research into ^{137}Cs speciation conducted by Forsberg and Strandmark [28] for the soils in Sweden led to the conclusion that the concentration of ^{137}Cs in the forms soluble in a soil solution and absorbed by the soil sorption complex did not exceed, respectively, 1% and 2% of the forms extracted from the soil with nitric acid and of the so-called residual fraction. According to the authors, cesium concentration depends on the type of soil; in sandy soil it is slightly higher than in loamy soil. Niesiobędzka [6] estimated the average concentration of ^{137}Cs mobile forms in the soils of northeastern Poland at 3.56% of the total radiocesium concentration. It must be emphasized here that in the Siedlce area the percentage of cesium forms available for plants is higher than the percentage of potassium forms soluble in a soil solution and absorbed in the soil sorption complex; the estimates for potassium were, respectively, 0.9% and 1.6% of the total potassium concentration. Potassium, as a natural component of the soil, is strongly fixed in different mineral combinations. ^{137}Cs , which is an anthropogenic isotope, can be found in the soil in more mobile forms determined by a number of factors, including the granulometric composition of the soil. Sandy soils are predominant in the Siedlce area. According to the literature data [4-6, and others], the availability of cesium for plants is higher in sandy than in loamy soils.

The consequence of soil contamination with ^{137}Cs after the Chernobyl accident in 1986 and as a result of nuclear weapons testing that occurred in the 1950s and 1960s is that ^{137}Cs is still detected in food.

Out of the different classes of examined food, milk delivers the major portion, 30-50%, of ^{137}Cs . Due to the fact that milk represents a considerable fraction of the total diet, its radiotoxicity is the basis for estimating the ingestion dose from radionuclides. During 1996-2005 in Poland liquid milk contained less than 1 Bq dm^{-3} of ^{137}Cs on average [3]. During 1998-2007 in the Siedlce area the concentration was more than twice as high – 2.48 Bq dm^{-3} . For comparison, in 2005 in Lithuania, Traidaraite et al. [29] found ^{137}Cs concentrations in milk to range between 1.088 and 3.000 Bq kg^{-1} . The radionuclide was present at higher concentrations in the samples taken during the summer than in those taken during the winter.

During 1993-2005 in Poland the annual average ^{137}Cs concentration in beef and pork ranged from 1.5 to 2.6 Bq kg^{-1} , and in poultry – between 0.7 and 1.0 Bq kg^{-1} [3]. During 1998-2007 in the former Siedlce province the average concentrations detected in meat samples were as follows: pork – 2.47 Bq kg^{-1} , beef – 6.38 Bq kg^{-1} , and poultry – 2.22 Bq kg^{-1} . During 1992-2007 in Poland fruit and vegetables contained 0.4 - 0.7 Bq kg^{-1} of ^{137}Cs on average [3]. In 1998-2007 in the Siedlce area ^{137}Cs was present in vegetables at concentrations ranging from 1.44 Bq kg^{-1} (potatoes) to 3.89 Bq kg^{-1} (other vegetables).

The maximum annual radiation dose from ^{137}Cs through ingestion, equivalent to the annual effective dose of 1 mSv , is estimated at $80,000 \text{ Bq}$ [30]. The ^{137}Cs intake from food in the former Siedlce province during the period 1998-2007, roughly estimated at $1,555 \text{ Bq year}^{-1}$, was about $20 \mu\text{Sv year}^{-1}$; in 1998, its level was nearly two and a half times higher than the average established during the ten years of research. An individual's average ionizing radiation dose through ingestion in the investigated area was almost twice as high as the average dose in Poland during 1990-2003, estimated by Mazur at $11.7 \mu\text{Sv}$ [31]. Higher ionizing radiation levels in the Siedlce area during the post-Chernobyl period can be accounted for by more severe contamination of the area immediately after the nuclear reactor accident [5, 15].

^{40}K concentrations in vegetables from the Siedlce area did not differ from those detected in the selected medicinal plants from Karpacz (an area in the Karkonosze mountains), which contained 63.8 - 134.6 Bq kg^{-1} of ^{40}K [32]. The author's own measurements made as part of the research revealed that the average ^{40}K concentrations in potatoes and other vegetables were, respectively, 120.1 Bq kg^{-1} and 98.54 Bq kg^{-1} . Similarly, a ^{40}K concentration of 48.8 Bq dm^{-3} in milk did not differ much from the national average estimated at ca 43 Bq dm^{-3} [33].

The daily average doses of ^{137}Cs and ^{40}K through ingestion in the Siedlce area during 1998-2007, determined as part of the author's own research, were 4.26 Bq and 135.1 Bq , respectively. The 135 Bq dose of ^{40}K is equivalent to 4.26 g of potassium. Contrary to the previous statement about the soils in the Siedlce area being deficient in potas-

sium [17, 18], the daily supply of this element from food, estimated on the basis of ^{40}K concentration measurements, is most likely to satisfy the body's daily demand for potassium, ranging between 1.6 and 2.0 g day^{-1} [34].

Conclusions

1. The factors determining the occurrence of potassium in the surface layers of the arable soils in the Siedlce area are the following: soil reaction, carbon content in organic compounds, and the presence of a fine soil fraction.
2. No statistically significant correlations were found between the chosen properties of the arable soils and ^{137}Cs activity.
3. The radiation dose from ^{40}K through ingestion in the Siedlce area does not differ significantly from the national average.
4. The ionizing radiation dose from ^{137}Cs through ingestion during the period 1998-2007 in the Siedlce area was about three-fold larger than the national average.

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