Concentration of Caesium Isotopes in Foodstuffs in Poland

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INTRODUCTION

adiological contamination of the environment is caused by nuclear activities on the globe: nuclear weapon tests and recently, the Chernobyl accident. The transfer of radionuclides to the organism via ingestion is one of the sources of doses obtained by people. To assess the doses received by humans the intake of isotopes with daily diet was defined. The concentration of radionuclides in particular foodstuffs was determined.

he network of Service for Measurement of Radioactive Contamination systematically controls all kinds of important food products such as milk, meat, vegetables, fruit, cereals and forest products: mushrooms, blueberries etc. Measurement stations involved in food monitoring act within Sanitary Epidemiology Stations, Veterinary Hygiene Units and Chemical - Agricultural Stations. All activities are co-ordinated by the Centre for Radioactive Contamination Measurement in the Central Laboratory for Radiological Protection (1).

he concentration of ^{131}I, ^{137}Cs, ^{134}Cs and ^{90}Sr in foodstuffs has been controlled in detail after the Chernobyl accident. The contamination with ^{131}I was decreasing quickly after June 1986. The contamination related to ^{90}Sr increased slightly in comparison to the pre-accident period. The only considerable concentrations were observed for the caesium isotopes: ^{134}Cs and ^{137}Cs. The level of activity of caesium isotopes has regularly been monitored in collected samples originating from different administrative districts of Poland. Since 1994 the ^{134}Cs concentration has been below the detection limit. The activity of ^{137}Cs has been measured to determine long-term effect of the accident on the contamination of milk, meat and other foodstuffs.

MATERIALS AND METHODS

he sampling rate has been dependent on the radiological situation. Before the Chernobyl accident food samples were collected on monthly, quarterly and annual basis of all administrative districts in sampling points. Shortly after the Chernobyl accident milk was collected daily, meat, fish, poultry and eggs weekly. The most common kinds of vegetables and fruit were sampled daily, particularly these ones, which were ready for consumption at that time. This schedule of sampling was obligatory from May to July 1986. Afterwards milk was collected weekly other products monthly. Since 1987 a special sampling program has been valid. Milk has been collected weekly and the average monthly sample has been tested. Meat (of different kind), fish and poultry have been collected once per 3 months, eggs once a year, and the most popular vegetables and fruit while harvesting.

o analyse the distribution of radioactive contaminants over the country, since the sixties the territory of Poland has been divided into 8 control regions shown in Figure 1.

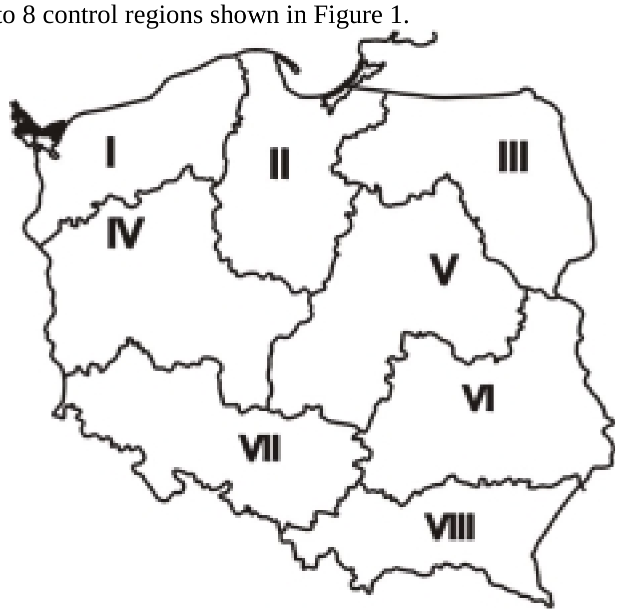

Figure 1. Control regions in Poland.

he activity of caesium isotopes was determined by radiochemical method and gamma spectrometry. For radiochemical determination samples were ashed at 450°C and dissolved in nitric acid (1:1) and the solution was filtered through a radiochemical funnel with ammonium molybdophosphate (AMP) bed selective for caesium. The activity of caesium in the bed was determined using a counter with thin plastic scintilator. The bed absorbed all caesium isotopes; therefore the ^{137}Cs was inferred from the activity ratio: ^{137}Cs to ^{134}Cs. The ratio has been changing in time following the accident from 2:1 in May 1986 to 120:1 in 1999.

or the spectrometric measurement a gamma spectrometer with HPGe or NaI(Tl) detector was used. Measurement of caesium isotopes in Marinelli geometry was performed without preparing of milk and after homogeneity of other samples or in ash samples in geometry of a flat cylinder placed on the detector. Efficiency calibration of the detectors was performed by using mixed gamma radionuclides standard solution and IAEA reference materials such as milk and soil.

RESULTS AND DISCUSSION

efore the Chernobyl accident activity of ^{137}Cs in foodstuffs was below 1 Bq kg^{-1}. The regional differences were not observed over the whole territory of Poland. ^{137}Cs in foodstuffs was the consequence of ^{137}Cs accumulated in soil as a follow up of nuclear weapon tests. The situation changed drastically immediately after the Chernobyl accident when the ^{137}Cs and ^{134}Cs nuclides appeared in large quantities, both in air and total fallout, resulting in the soil, pasture and plants contamination. The pasture season for cows begins early in May. The distribution of contamination was non-uniform with significant local differences as is shown in Figure 2. The north-east area (region III and a part of region V), Opole district (region VII) and Cracow district (region VIII) were the most contaminated area. The low level of contamination was registered in region I and IV (2).

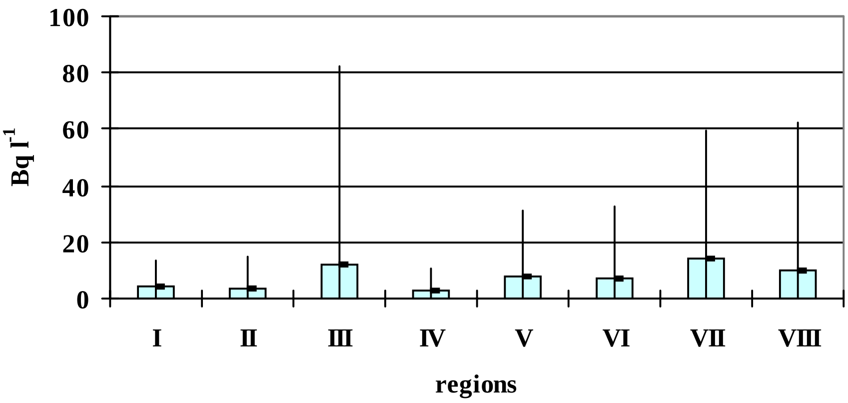

Figure 2. Average annual activity of ^{134}Cs +^{137}Cs (and range) in milk in 8 control regions in Poland, 1986 [Bq l^{-1}]

ilk is important for the monitoring of radioactive contamination as radionuclides appear in it immediately after a cow has eaten contaminated fodder, as well as the fact that it has great share in our diet. Mean activity of ^{137}Cs and ^{134}Cs in milk was 25 Bq l^{-1} and 13 Bq l^{-1} respectively in May 1986. In samples of milk from some cows the activity of several hundreds Bq l^{-1} was found. In June the contamination level decreased approximately by half, during the next months was dropping gradually. From the beginning of 1987 an increase was observed again. This was caused by feeding cows with hay collected in June 1986 (3).

n the following years the activity of caesium isotopes in milk was decreasing and it reached a level of approx. 1 Bq l^{-1} at the end of 1992. Activity of ^{134}Cs has been below detection limit in most samples of milk since 1992. Figure 3 shows the changes of ^{137}Cs in mean monthly samples of milk in Poland during the period between 1986-1999.

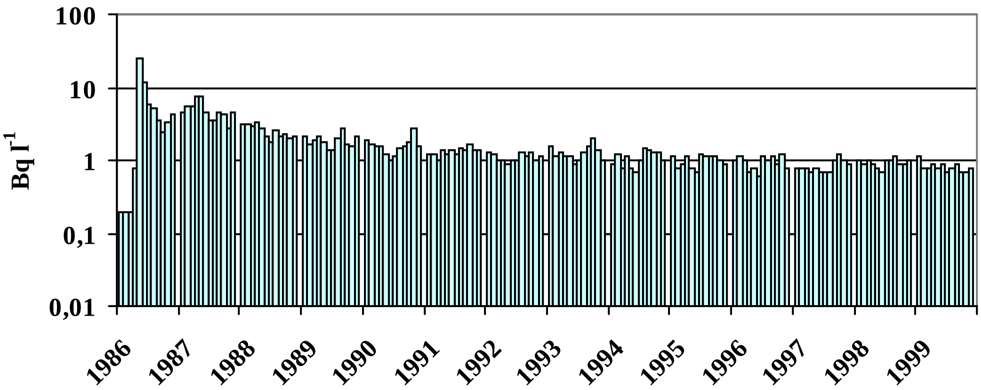

Figure 3. Mean monthly activity of ^{137}Cs in milk in Poland, [Bq l^{-1}].

our kinds of meat were tested: pork, beef, mutton and horse meat. Until the middle of May 1986 the concentration of caesium isotopes did not exceed 26 Bq kg^{-1} of ^{137}Cs and 14 Bq kg^{-1} of ^{134}Cs but it increased later because of the accumulation of these isotopes in animal organism. In some cases between June and August 1986 the concentration of both caesium isotopes exceeded 500 Bq kg^{-1} – 800 Bq kg^{-1} in single samples. Table 1 shows the activity of caesium isotopes in different kinds of meat in 1986 -1988.

Table 1. Activity of ^{134}Cs + ^{137}Cs in meat in Poland in 1986-1988, [Bq kg^{-1}]

	^{134}Cs + ^{137}Cs Bq kg^{-1}	Beef	Mutton	Horse Meat	Pork
1986	mean	50	57	65	35
	range	<5 – 527	<5 – 860	7 – 594	6 -316
1987	mean	45	53	61	21
	range	<5 - 622	<5 - 533	<5 - 501	<5 - 106
1988	mean	19	21	32	10
	range	<5 - 226	<5 - 209	<5 - 728	<5 - 44

uring the few years the level of meat contamination has been decreasing systematically. Since 1994 the activity of ^{134}Cs was below the detection limit in most samples. The average activity of ^{137}Cs has been on the level of 2 - 3 Bq kg^{-1} reaching 50 Bq kg^{-1} in few samples. The differences in activity between kinds of meat were less visible. In 1986 the contamination of poultry and fish was below some tens of Bq kg^{-1} and decreasing gradually. At present the activity of ^{137}Cs in poultry is below 1 Bq kg^{-1}. Mean concentration in fish is on level of the 2 Bq kg^{-1} reaching 20 Bq kg^{-1} in single samples.

igure 4 summarises the result for game such as boar, roe deer and stag. It was immediately noted that the contamination of game has been much more significant than that reported above for other meat. After a detailed analysis of the results presented on the graph, one perceives that the level has not significantly changed in the years 1986 - 1999. This high content of caesium and its fairly constant level in time can be attributed to the specificity of caesium recycling in forest ecosystems. Caesium slowly penetrates into the forest soil and accumulates in litter, mosses and lichen being fodder for wild animals. Wide scatter of results may be explained by the nonuniform distribution of radioactive contaminants in ingested food and, by the differences in nutrition habits (and opportunities) of single animals, e.g. availability of certain kind of food in the area, etc.

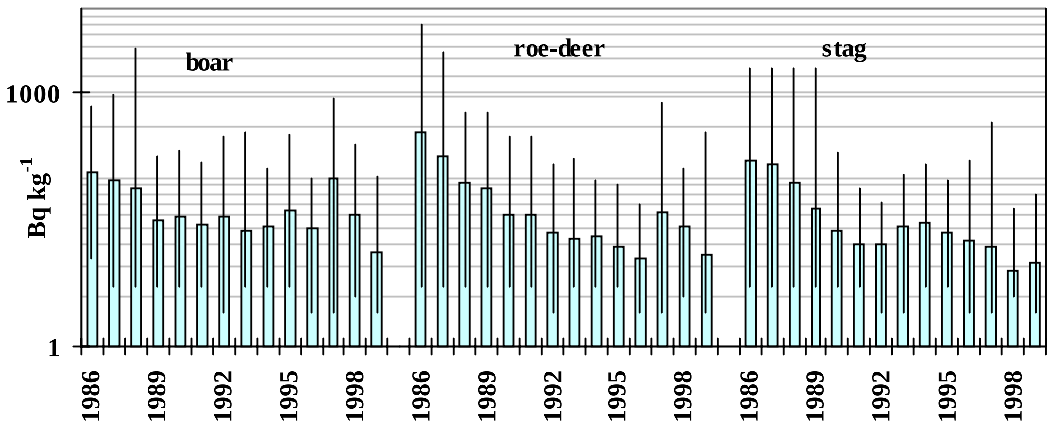

Figure 4. Activity of $^{134}Cs+^{137}Cs$ in game in Poland, [Bq kg^{-1}]

he season of vegetation starts in May and during summer month most of fruit and vegetables is harvested. Contamination of the environment after the Chernobyl accident caused high contamination of these plants. The highest average activity of $^{134}Cs+^{137}Cs$ in fruit was registered in currant 200-400 Bq kg^{-1}, in single samples even 2000 Bq kg^{-1}. The mean activity of other fruit ranged between 28 -120 Bq kg^{-1}. Fruit picked in September and later was less contaminated. Activity of both caesium isotopes in vegetables and cereals was less significant except leafy vegetables picked in May and June, in which the activity exceeded some hundreds of Bq kg^{-1}. Already in 1987 the content of accumulated radionuclides significantly decreased in comparison to 1986 and since 1989 the average activity has been below 1 Bq kg^{-1} as is shown in table 2.

Table 2. Activity of $^{134}Cs+^{137}Cs$ in fruit and vegetables in Poland, [Bq kg^{-1}]

	Fruits Bq kg^{-1}			Vegetables Bq kg^{-1}		
		mean	range		mean	range
1986	black currant	385	31 –2201	cabbage	13	<5 – 93
	other currant	291	26 – 1769	carrot	17	<5 – 93
	gooseberry	120	13 – 1219	potatoes	17	<5 – 49
	raspberry	79	<5 – 546	other vegetables	28	<5 - 215
	strawberry	28	<5 – 205			
	apple	31	<5 – 323			
	plum	29	<5 - 141			
1987	currant	10	<5 – 44	all kinds of vegetables	1,2	0,3 – 15,4
	other fruit	3,8	0,3 – 35,7			
1988	currant		<5 – 20	all kinds of vegetables	1,0	0,3 – 17,9
	other fruit	2,0	0,3 – 9,8			
1989 -1999	all kinds of fruit	0,8 – 0,5	<0,1 – 19,2	all kinds of vegetables	0,9 – 0,5	<0,1 – 11,4

mong forest fruit the greatest contamination has been observed in blueberries. Detailed results from 1986-1999 are shown in Figure 5. The high level of contamination in blueberry is connected to the specific behaviour of caesium in forest ecosystem.

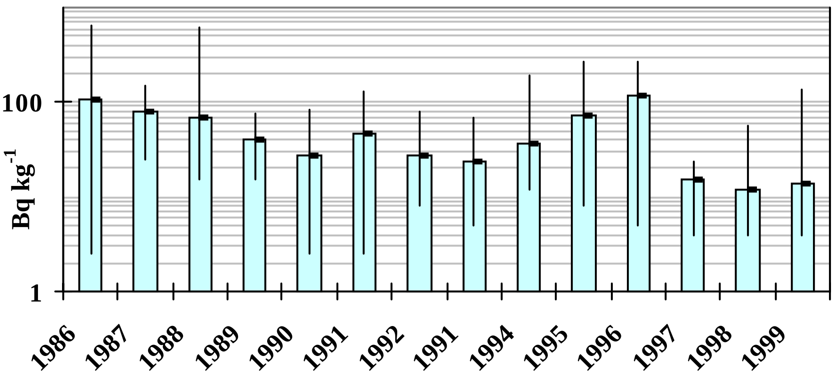

Figure 5. Activity of ^{134}Cs + ^{137}Cs in blueberries [Bq kg^{-1}]

igh level of contamination of caesium isotopes has also remained in mushrooms. Xerocomus Badius, Cantarella Cibarius,and Boletus Edulis are the most common mushrooms in Poland. This drastic contamination of mushrooms in comparison to other foodstuffs derives from the specific condition of growth (mushrooms grow in lichen and mosses which accumulate radionuclides). Xerocomus badius is the most contaminated one because of his tendency to accumulate caesium. Table 3 shows activity of both caesium isotopes in mushrooms in Poland during the period between 1986 – 1999 and activity of ^{137}Cs in single samples in 1985.

Table 3. Activity of ^{134}Cs and^{137}Cs in mushrooms in Poland, [Bq kg^{-1}]

	Xerocomus Badius		Boletus Edulis		Cantharella Cibarius	
	Bq kg^{-1}					
	mean	range	mean	range	mean	range
1985	169		60		77	
1986	528	80 - 2036	50	15 – 165	91	11 – 710
1987	484	108 – 2854	49	17 – 111	82	12 – 784
1998	729	68 – 2060	103	42 – 259	53	15 – 148
1989	752	100 – 2360	70	8 – 178	42	18 – 132
1990	599	40 – 2098	82	12 – 231	84	20 – 381
1991	547	144 – 1381	74	15 – 236	145	14 – 806
1992	558	96 – 2084	109	24 – 649	52	12 – 208
1993	386	20 – 1240	93	8 – 445	52	5 – 313
1994	374	44 – 864	38	5 – 68	28	5 – 136
1995	381	160 – 1040	49	10 – 105	47	9 – 176
1996	200	48 – 460	94	14 – 440	31	5 – 78
1997	476	60 - 1070	61	18 - 140	20	<5 - 62
1998	376	64 - 1880	81	7 - 256	48	<5 - 269
1999	270	17 - 1201	69	5-1011	63	<5 -609

ne can be surprised by the fact that in a few samples of mushrooms from 1985, there were more ^{137}Cs than in other food products (e.g. Xerocomus badius 169 Bq kg^{-1}) (4). This suggests that the overall radioactive contamination of forest mushrooms might equally be attributed both to the nuclear weapon tests in the atmosphere and to the Chernobyl accident

The calculation of an average intake of ^{134}Cs and ^{137}Cs has been based on statistical data for consumption of certain products important in a daily diet. Milk and meat, mainly pork and beef, have been the main contribution of caesium isotopes with a share of 30-50% and 20-30% respectively. High consumption of potatoes, vegetables and cereals, in spite of fairly low activity in such products, has contributed with 10-15% to the annual isotope intake. In 1985 the average intake of ^{137}Cs due to contaminated food consumption was on level 325 Bq per capita. The mean annual intake of ^{134}Cs and ^{137}Cs in 1986 per capita was 2054 Bq and 4324 Bq respectively. In 1987 the annual intake dropped to 805 Bq of ^{134}Cs and 2246 Bq of ^{137}Cs per capita. During the next years, the annual intake of radionuclides was decreasing according to the activity drop in foodstuffs and half life of ^{134}Cs. The level of ^{137}Cs has not changed since 1993; annual intake of this isotope is on a level of about

500 Bq per capita per year. Some uncertainties in that estimation could be related to the wide scatter of the ^{137}Cs content in various products and the change of habits observed over the years in the period under investigation (1986-1999).

There are no references to the consumption of game and mushrooms. The radioactive contamination of these products has been high, but their share in the daily diet could be significant for a very limited number of consumers. For most of the population it has only been a minor pathway of nuclides intake and therefore, it has a small effect on the estimation of average diet.

SUMMARY AND CONCLUSIONS

he Service for Measurement of Radioactive Contamination has assured a continuous monitoring of the foodstuffs on whole territory of Poland. During the period between 1986 –1999 some hundreds of each kind of tested samples were analysed. The great amount of samples has allowed a good statistic dose assessment (5).

Before the Chernobyl accident there were no territorial differences in the radioactive contamination foodstuffs. Fallout originating from nuclear explosion in the world covered Poland uniformly. After the Chernobyl accident the situation changed. There have been regional and even spot differences in contamination. Radioactive contamination of ^{134}Cs according to half-life lost importance in 1994. As the long-term effect on environment the activity of ^{137}Cs has been determined.

Radioactive contamination of fruit, vegetables, cereals, eggs, poultry is on the same level as it was before the accident. Higher radioactivity remains in milk, meat, fish and forest products.

Based on the data concerning activity of caesium isotopes in different kinds of foodstuffs, an annual effective dose related to the consumption of contaminated food could be assessed for the period between 1985 and 1999. Table 4 presents the doses obtained by the population of Poland as an effect of consumption contaminated food.

Table 4. Mean annual effective dose due to caesium isotopes intake via ingestion in Poland [μSv]

	^{134}Cs	^{137}Cs
	μSv	
1985	-	4
1986	34	54
1987	13	28
1988	4	13
1989	3	12
1990	2	12
1991	2	11
1992	1	8
1993 - 1999		6 -7

aving considered the diversity of dietary habits of consumers and their place of living, the effective dose received via ingestion may even be five times higher. In Poland the accepted effective dose limit for people exposure to ionising radiation caused by contamination of environment with artificial radionuclides is 1 mSv.

his limit is in accordance with recommendation of the IAEA and the International Commission of Radiological Protection (6). As compered to this, Polish population obtained doses being on the level of some percent of the internationally accepted dose limit.

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