

Chernobyl Fallout in the Environment of South-Eastern Poland - A Review

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Abstract: The influence of the extent of Chernobyl emissions on the environment of the territory of South-eastern Poland was reviewed on the basis of research on environmental samples originated from soil, plants and animals. The studies carried out showed that the radioisotope tested Chernobyl contamination in a matter of several years was reduced to an average value of observed environmental norms and coming mainly from global fallout residue.

Key words: Chernobyl disaster • radioactive pollutions • Poland

INTRODUCTION

Presently, 21 years have passed since the time of the greatest atomic emergency, the Chernobyl catastrophe, which took place on the 26th of April 1986. It is marked as the greatest emergency on a 7-measure scale worked out by the International Atomic Energy Agency [1]. Erroneous decisions of operators together with unfavorable parameters of this type of work for these reactors lead to damage of the reactor core. This resulted in the destruction of the reactor and releasing into the atmosphere large quantities of radioactive materials, as well as the radioactive contamination of large regions, particularly in Europe.

The total release of radioactive substances was about $1.4 \cdot 10^{19}$ Bq (as of April 26, 1986), which included $1.8 \cdot 10^{18}$ Bq of ^{131}I , $8.5 \cdot 10^{16}$ Bq of ^{137}Cs and other cesium isotopes, $1 \cdot 10^{16}$ Bq of ^{90}Sr and $3 \cdot 10^{15}$ Bq of plutonium radioisotopes. Also noble gases were released in large amounts [2]. Most of the strontium and plutonium isotopes were deposited within a 100-km zone around the reactor [2]. The most important isotopes are ^{137}Cs , ^{90}Sr , plutonium and ^{241}Am , which reveal a rather large half-life time [2].

The most air-born isotopes, such as ^{131}I or ^{137}Cs contaminated a large region of Europe, from which the deposition was highly heterogeneous. A characteristic trait of the Chernobyl contamination was the appearance of the so-called hot spots, meaning fragments of the reactor core of several micrometer size, containing

highly radioactive isotope, such as Eu, Pm and also Pu, Am and Cm [3].

In the course of 21 years, which have passed since this event, many diverse studies have been carried out and a summary of the effects of the catastrophe are presented in many reports by IAEA and NEA [2, 4]. In Poland, studies were also carried out on the different issues related with the appearance and migration of Chernobyl contamination.

The following work presents a summary of the influence of the extent of Chernobyl emissions on the environment of the territory of South-eastern Poland on the basis of research carried out in the Department of Radiochemistry and Colloid Chemistry, Maria Curie Skłodowska University, Lublin.

Experimental: Systematic studies of a contamination of the environment of Eastern Poland began several years after the Chernobyl catastrophe, after gathering measurement apparatus of appropriate quality (gamma spectrometer with germanium detector and alpha spectrometer with silicon detector). Using this apparatus, research was carried out on the contamination of diverse elements on the environment.

The analyzed samples were taken according to the IAEA guidelines [5]. The subject of the studies was non-cultivated soil samples (in some cases arable soil was also analyzed), sediments from rivers and lakes, specified plants (grass, cultivated plants), the bones and eggshells of birds.

Determination of the following anthropogenic isotopes was made: ^{137}Cs , ^{90}Sr and $^{238,239,240}\text{Pu}$. Determination each of these isotopes required appropriate preparation of samples, including radiochemical treatment necessary for identifying isotopes emitting alpha and beta rays.

Later apparatus and ways of preparing samples for determination of selected radionuclides were presented.

Determination of gamma emitting isotopes (including ^{137}Cs): The collected material for gamma spectrometric measurement was dried at 105°C and then mechanically crushed to the size of several millimeters wide (bones, plants) or 2 mm (sediments and soils). Small samples prepared in this way (about 20 g) were placed on a flat container of 50 mm diameter and about 5 mm height.

Samples of appropriately large volume (about 500cm^3) were placed in Marinelli beakers.

Samples were subjected to spectrometric measurements performed with the gamma spectrometer (Canberra/Silena) equipped with an HPGe detector of 87 cm^3 active volume, 17.5% relative efficiency and 1.8 keV FWHM resolution. A quantitative analysis was performed using Canberra Genie-2000 software with the goal of determining the activity of natural and artificial radioactive isotopes.

Determination of alpha emitting isotopes (^{238}Pu , $^{239,240}\text{Pu}$): Determination of plutonium in environmental samples requires separation of these isotopes in a pure form and fixing them on a steel plate, which enables alpha spectrometric measurements. This is realized by a radiochemical multi-stage procedure. In the first stage of this procedure a dried and ashed ($450\text{-}500^\circ\text{C}$) sample was leached with 6M HCl. From the obtained solution trace elements were co-precipitated with iron hydroxide (III) by 25% NH_4OH addition. The precipitate was dissolved in 6M HCl and then separated from Fe by co-precipitation with calcium oxalate at $\text{pH}=3$. Next, the precipitate was dried, burned in oven, dissolved in 12M HCl and then a second co-precipitation with iron hydroxide (III) was performed. In the next stage, precipitate was dissolved in 8M HNO_3 , boiled on sand bath with NaNO_2 until nitric oxides disappeared. This stage allows changing the oxidation state of plutonium to the Pu^{4+} form. The solution was introduced on ion-exchange column filled with Dowex 1x8 (50-100 mesh). Before sample introduction the column was washed with 8M HNO_3 in order to exchange chloride ions on nitrate ions. After the sample was passed through the column, the next solutions were introduced as follows: 8M HNO_3 ,

6M HCl and concentrated HCl in order to eliminate trace elements (Th, Am), which disturb Pu determination. In the final stage of anion exchange Pu was eluted from the column using concentrated HCl with 0.1M NH_4I , which changed the plutonium oxidation stage to +3. The obtained solution was evaporated to dryness with aqua regia to eliminate iodine and ammonium salts. Finally, Pu was electrodeposited onto stainless steel discs from 0.4M ammonium oxalate with 0.3M HCl and measured by alpha spectrometry [6].

Measurements were performed using the 7401 Canberra Alpha Spectrometer with a 1520 mixer/router and S-100 multichannel analyzer. The PIPS detector of 17keV FWHM resolution was used. For quantitative analysis the Canberra Genie-2000 software was used. Chemical yield was monitored by addition of a standard ^{242}Pu solution to the sample.

Determination of beta emitting ^{90}Sr : Determination beta radioactive ^{90}Sr in environmental samples demands isolating it from other, natural beta emitting isotopes appearing in the sample matrix. The procedure worked out by Solecki [7, 8] for separating this isotope with the goal of determination its concentration by the measurement method with liquid scintillator depends on sample ashing at a temperature of 600°C , leaching with concentrated nitric acid, extracting Y^{3+} with the help of TBP/ HNO_3 (after adding an yttrium carrier), precipitation of yttrium hydroxide using ammonia after dilution of the organic phase with ethanol, purification of Y by dissolving the ammonia precipitate with HNO_3 (Dowex 1x8 added to a bulk solution). Next, yttrium oxalate is precipitated and, after dissolving in nitric acid, ^{210}Pb is removed by scavenging with PbI_2 precipitation. After separating the precipitate from the remaining solution, yttrium oxalate is re-precipitated with the goal of yield determination by weight. After then, dry precipitate is suspended in water and is added to scintillation cocktail (Insta-gel Plus, Packard) in a low-potassium standard glass vial. Measurements were lead by the use of a Quantulus (Wallac-Perkin-Elmer) ultra low-level spectrometer for 300 min. The applied procedure allowed for obtaining a minimum detectable amount of 0.02 Bq/kg [7].

RESULTS AND DISCUSSION

In the period of several years after the Chernobyl catastrophe, it was possible to determine in the analyzed samples the activity of the gamma emitting isotope ^{134}Cs with a relatively short period of half-life (2.06 years). The entire quantity of this isotope was a result of Chernobyl

Table 1: Concentration of ⁹⁰Sr [Bq/kg] in soil and selected vegetable samples [7, 9, 10]

Samples	Min Max		Mean ± σ	Year
	value	value		
Soil of Bug river valley (near the river)	5.3	69.8	29 ± 21	1999
Soil of Bug river valley	6.8	85.3	24 ± 23	1999
Bug river sediment	3,7	20.5	10 ± 5	1999
Leczna-Wlodawa Lake District				
Arable soil	1.1	9.8	4.7 ± 3.5	2004
Non-cultivated soil	0.3	8.8	4.4 ± 2.9	2004
Potato <i>Solanum tuberosum</i>	1.0	2.4	1.0 ± 0.2	2004
Sugar beet <i>Beta vulgaris</i>	0.5	2.9	1.7 ± 0.3	2004
Carrot <i>Daucus carota</i>	0.2	0.6	0.4 ± 0.1	2004
Lake sediment	3.4	5.7	4.3 ± 4.5	2004

Table 2: Concentration of ¹³⁷Cs [Bq/m²] in soil samples [11, 13, 18]

Samples	Min Max		Mean ± σ	%Chem.	Year
	value	value			
Lake sediments					
(Piaseczno Lake) [Bq/kg]	0.7	238	77±68	40 - 73	1995
Lake sediments (Masluchowskie Lake)					
[Bq/kg]	3.5	290	101±94	39 - 100	1995
Artificial lake sediment					
[Bq/kg]	1.0	53	24±18	43 - 100	1995
Soil of Wieprz river valley					
(near the river)	237	4460	1620±910	42	1998
Soil of Wieprz river valley	588	3730	1750±840	52	1998
Wieprz river sediment			6.9	35	1998
Brown soil			4530		1998
Lessive soil			11920		1998
Podzol soil			5150		1998

fallout. Knowing the relation of cesium isotope from Chernobyl residue coming out to be ¹³⁴Cs/¹³⁷Cs = 0.528 [2], it was possible to designate the participation of ¹³⁷Cs coming from this fallout in the entire concentration in the sample. The remaining quantity of ¹³⁷Cs present in the environment was the result of atomic tests carried out to the greatest intensity in the '60s of the last century. The possibility of distinguishing the source of ¹³⁷Cs (global or Chernobyl fallout) was lost at the moment when ¹³⁴Cs activity lowered below the MDA value.

Analogously, testing the isotopic ratio ²³⁸Pu/^{239,240}Pu in samples, the participation of plutonium of Chernobyl origin can be identified. The participation of ²³⁸Pu in global fallout came out to about 4%, however in Chernobyl fallout – about 50%.

Results of the carried out studies are gathered in Tables 1-9, most often presented in the form of minimum

Table 3: Concentration of ¹³⁷Cs [Bq/kg] in selected lichen, animal, plant and peat samples [12, 18, 19, 24]

Samples	Min	Max	Mean ± σ	Year
Lichens, family <i>Parmeliaceae</i>	2.6	108	46±29	1998
Grey Heron <i>Ardea cinerea</i> - bone	0.08	3.2	1.4±1.0	2003
Grey Heron <i>Ardea cinerea</i> - eggshell	0.03	2.0	1.2±0.7	2003
Mute Swan <i>Cygnus olor</i> - eggshell	0.09	4.1	1.6±1.8	2003
Montagu's Harrier				
<i>Circus pygargus</i> - eggshell	0.85	2.0	1.2±0.7	2003
Marsh Harrier				
<i>Circus aeruginosus</i> - eggshell	0.68	0.7	0.7±0.02	2003
Saw Sedge <i>Cladium mariscus</i>	49	57		2005
Peat	34	53		2005

Table 4: Concentration of ²³⁹⁺²⁴⁰Pu [mBq/kg] in soil and sediment samples [6, 13, 15, 17]

Samples	Min Max		Mean ± σ	%chem	Year
	value	value			
Surface soil of					
Lublin region (0-2cm)	80	340	210 ± 80	15	1993
Lake sediment	10	650	2600 ± 100		1995
Forest soil profiles					
(0-45cm) [Bq/m ²]	60	335	40.4-61.2	0.7-2.9	1996
Lublin river sediments	70	315	50±36	17	1996
Soil of Wieprz river valley					
(near the river)	20	65	150±15	2-15	1998
Soil of Wieprz river valley	10	420	160±15	3-23	1998
Wieprz river sediment	10	200	40±10	2-27	1998
Soil of Bug river valley					
(near the river)			170±20	2-60	1999
Bug river sediment			60±5	35-70	1999
Soil of Wieprz river valley					
[Bq/m ²]			4-21	2-15	1998

and maximum values and arithmetic mean values, as well as the year in which the measurements were carried out. Table 1 concerns ⁹⁰Sr determination, Tables 2 and 3- determination of ¹³⁷Cs and Table 4 – determination of alpha emitting plutonium isotopes.

The average global fallout of ⁹⁰Sr came out to 3200 Bq/m², but up to the year 1986 (when additional amounts of this isotope were introduced into the atmosphere as a result of the Chernobyl disaster) it was lowered to the value of 1500 Bq/m² [25] as a result of the radioactive decay. Table 1 presents the average specific activities of ⁹⁰Sr in the Bug river valley, which are of this order and shows a not very large soil contamination by this isotope or/and of its quick vertical migration. This isotope relatively quickly enters deep into the soil, therefore the maximum concentration (⁹⁰Sr coming from global as well as Chernobyl fallout) can appear at a greater

depth than the depth of the taken samples (up to 10 cm deep). The concentration of ^{90}Sr in the soils of the Leczna-Wlodawa Lake District is relatively small (5-times less than in soils of the Bug river valley), since in this case the depth of the taken samples of soil was 5 cm. The concentration of ^{90}Sr in the studied vegetables came out to about 1 Bq/kg in relation to the dry mass, which shows that about 20% of ^{90}Sr contained in the soil comes from the tested plants. If one takes into account about 90% of the water present in vegetables, then the quantity of ^{90}Sr entering into them is reduced to the value of about 2%.

The results of Table 2 show that the radioactive fallout of ^{137}Cs on eastern territories of Poland was relatively not homogenous (large differences between the minimum and maximum values). Global fallout is considered rather as homogenous, therefore the observed non-homogeneity comes from a large participation of Chernobyl fallout in the entire fallout of ^{137}Cs , reaching even 100% (lake sediments).

Informations presented in Table 3 shows a rather large concentration of ^{137}Cs in only studied species of lichens from family *Parmeliaceae*. Similar quantities were observed also in samples of peat and saw sedge tested, taken from these same territories of calcareous peat bogs. Lichens comprise bio-monitors of impurities in the air, since most of their vital elements are taken from the atmosphere. Therefore, the observed contamination with ^{137}Cs points to their re-suspension character. The studied, on the other hand, concentration of ^{137}Cs in bones and eggshells of select species of birds show insignificant values, often on the limit of detection, which is also shown by the great uncertainty of measurement results.

Studies on ^{137}Cs content in the eggshells of Montagu's Harriers *Circus pygargus* showed specific activity 39-46 times lower in comparison to the concentrations discovered in Saw Sedge *Cladium mariscus*, the plant in which the mentioned raptors nest. The displayed concentrations in eggshells were also 27-43 times lower than those determined in the peat of marshes, the areas on which this plant grows [24]. The above observations indicate that the transfer of ^{137}Cs from the environment to the tissues of the raptors from food comprising of small mammals and insects related with the soil environment occurs in a very low degree. The determined mean specific activity of ^{137}Cs for all the studied birds was very low in comparison to the bones of other Polish raptors studied up to present times [23].

As can be noticed (Table 4), at the average value of $^{239,240}\text{Pu}$ in the soil comes out to about 200 mBq/kg;

concentration of plutonium in lake sediments 10 times exceeds this value, in the contrary, in river sediments they are about 4 times lower. Taking into account the same mechanism of the transfer of radionuclides from soil to water it can be supposed that the lack of water flow in the case of lakes does not allow for a removal of radio nuclides coming into the water. In this way plutonium entering into the water after a certain time is accumulated into the bottom sediment in quantities depending on the distribution coefficient of radionuclides between the water phase and residue, including the subsequent chemical reactions such as complexation, precipitation or creating of radio-colloids.

Similar behavior was also observed in the case of ^{137}Cs , an anthropogenic element coming from global and Chernobyl fallout [13, 17]. Both of these elements (plutonium and cesium), being the result of human activity, are closely bound to the upper layers of soil. Therefore, they do not easily enter into the water phase, even after their long-term contact with soil (as a result of occasional flooding of river valleys).

It is interesting to compare the quantity of Chernobyl fractions of $^{239,240}\text{Pu}$ in samples coming from the Wieprz and Bug river valleys, despite the relatively large error related with calculating this fraction. The average participation of Chernobyl $^{239,240}\text{Pu}$ in soils and bottom sediments of the Wieprz came out to about 8% and in the case of the Bug its part reaches about 35%. This difference can be tied to the fact that the Bug flows through territories that were to a greater degree exposed to Chernobyl fallout.

As results from the carried-out research in soils on the Lublin territory, a relatively large participation of the Chernobyl $^{239,240}\text{Pu}$ fraction is observed (contained in the range of several to several dozen percent of the total quantity of these plutonium isotopes). It is also observed that the territories placed along the eastern border of Poland (the Bug valley) are characterized by an average high concentration of Chernobyl plutonium.

CONCLUSION

Among the main classes of environmental contaminants, radionuclides tend to be generally less well known and less frequently studied than heavy metals or organic compounds [20, 21, 22]. The above comes from the fact that the problem of radioactive contaminants concerned a small number of regions to which, on account of military matters, there was limited access. However, it was this prediction of researchers which changed after the

disastrous explosion of the fourth unit of the Chernobyl nuclear power plant (1986). The global character of the then released radioisotopes and their accumulation in natural as well as transformed ecosystems and their transportation through biotic and abiotic pathways made them a subject of interest for research while taking advantage of various models.

The studies carried out showed that the radioisotope tested Chernobyl contamination in a matter of several years was reduced to an average value of observed environmental norms and coming mainly from global fallout. As literature data shows [9, 13, 14, 17] Chernobyl isotopes migrate more quickly than those coming from global fallout. Therefore, after the initial increase of their concentration in the environment, after several years a lowered concentration came about to the level prior to the Chernobyl catastrophe.

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