Concentration of ^{137}Cs, ^{40}K, ^{238}Pu and $^{239+240}Pu$ Radionuclides and Some Heavy Metals in Soil Samples from Two Main Valleys from Tatra National Park

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Abstract

This paper presents the results of determinations of radiocesium and plutonium activity concentrations and some heavy metals in soil samples from two main Tatra Valleys: Koscieliska and Rybi Potok Valley. The results were obtained during 2001. It has been found that the content of ^{137}Cs and $^{230+240}Pu$ is much higher at altitudes over 1300 m asl.

Keywords: caesium, plutonium, Tatra Mts, heavy metals, alpha-spectrometry, gamma-spectrometry, radiochemistry

Introduction

The southern part of Poland is known as a territory of exceptional beauty and high natural qualities. There are the highest Polish mountains: the Tatras. The Tatra Mts., located in the center of the Western Carpathians, constitute the highest mountain massif within the whole Carpatian Range. It is the only area in the Carpathians which preserves a typical postglacial relief, hanging valleys, glacial cirques and numerous high-mountain oligotrophic lakes. The area is additionally differentiated according to the geological structures into the Western Tatras (limestones) and the High Tatras (cristalline rocks). They are also the centre of occurrence of high-mountain flora and fauna in this part of Europe as well as in the whole Western Carpathians.

The Tatras are simultaneously the northernmost centre of endemism connected with the alpine system mountains. When comparing the area, the Tatra Mts. are equal only to a single medium-size valley in the Alps or to the area of Warsaw [1, 2].

The Tatra Mts form the border between Poland and Slovakia. The minor part (about one fourth of the area) located on the north slopes, is located in Poland. Since 1954 the Tatra region has had the status of a national park.

Environmental pollution is one of the main anthropogenic threats. Following the Chernobyl accident the natural ecosystem of Tatra National Park has been seriously contaminated by radioactivity. Radioactive ^{137}Cs and $^{239+240}Pu$ were introduced into the natural environment by nuclear tests conducted in the middle of the

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20th century, and in 1986 as a result of the failure of the Chernobyl nuclear reactor. Gamma-spectrometric measurements of radionuclides in the soil samples were performed by the Department of Nuclear Physical Chemistry of the Institute of Nuclear Physics. The AES-ICP (atomic emission spectrometry with inductively coupled plasma) measurements of these samples were done by the University of Mining and Metallurgy.

The aim of this work is to present the results of measurements of gamma-emitting radionuclides (^{137}Cs, $^{239+240}Pu$ and natural ^{40}K) and some heavy metals (Zn, Fe, Pb, Cu and Cd) concentrations in soil samples taken from selected places in the Tatra Mountains (Rybi Potok Valley region and Koscieliska Valley region).

Method of Measurements

Sample Collection

Soil samples were collected with 10 cm diameter plastic cylinders. Sampling depth was 10 cm. Monolith cores were taken out and preserved in the cylinders. The cores were sliced in laboratory into 3.5 or 2.5 cm thick slices. Each slice was considered a separate sample. Slices were dried at 105°C for one day and homogenised by means of grinding. The sample was sieved on vibrating screen (diameter of circular-hole screen was about 2 mm).

Sample Decomposition and Analytical Technique

Soil sample decomposition was the first step of the procedure after pulverisation. Metals content in the samples was determined by inductively coupled plasma absorption spectroscopy AS ICP. Microwave high-pressure and mineral acids and hydrogen peroxide were used to decompose the samples.

Gamma Spectrometric Measurements

All ground and homogenous samples were taken for gamma spectrometric measurements with a low-background gamma spectrometer equipped with Silena HPGe detector (10% efficiency, FWHM=1.8 keV at 1173 keV). The shield was made of 10 cm lead with additional lining made of Cd (2 mm) and Cu (20 mm), with inflow of liquid nitrogen vapours to reduce radon daughter background. Typical measurement time was four days. The spectrometer was calibrated according to the method described previously [3]. Obtained spectra were evaluated by using P.I.M.P code [4]

Plutonium Measurement

The measurement procedure followed the general concepts of the method applied by J.J LaRosa and coworkers for the analysis of Chernobyl Project samples in the International Atomic Energy Agency (IAEA) Laboratories at Seibersdorf [5] and slighty modified by Mietelski et al. [6, 7].

Each homogenised sample was taken for plutonium analysis. The procedure started with incineration at 600°C lasting for at least 8 hours. Then the samples of 10 g of ash were taken and the appropriate dry mass for each was calculated. Samples were transferred to PTFE beakers where the ^{236}Pu tracer (9.3 mBq, 1.03.2001) was added. Samples were dissolved by means of sequential use of hot concentrated acids: HF, HNO_3 HC1 with the addition of H_3BO_3 and again HNO_3. Then the solutions were converted to 1 M HNO_3 and filtered after cooling.

The oxidation adjustment procedure [5] was performed using hydrazine and $NaNO_2$. After that, the solutions were converted to 8 M HNO_3 and again cooled down. Thorium and plutonium were separated from all the other elements by anion-exchange chromatography using DOWEX 1x8, 200 mesh, Cl^- form (Sigma Ltd.). Thorium was stripped off the column by using 50 ml of 10 M HCl, whereas the plutonium was then striped off by passing through the column 25 ml of 0.1 M HCl - 0.1 M HF solution. Thin, alpha spectrometric sources were prepared directly from Pu fractions by using NdF_3 co-precipitation method [8].

Alpha spectrometric measurements were performed by using Silena Alpha Quattro alpha-spectrometer with silicon detectors (Canberra PIPS 450 mm^2 of area). Alpha-sources were measured while situated very near the detector. Obtained spectra were analysed with the ALF software developed by Mietelski.

The analysis of plutonium activity concentration ratios (^{238}Pu to $^{239,240}Pu$) allowed us to distinguish the Chernobyl and global fallout components of plutonium activities. Assumed ^{238}Pu to $^{239,240}Pu$ activity ratio in global fallout was equal to 0.04, and that for Chernobyl fallout was taken equal to 0.55, as discussed previously [7, 9].

The percentage *F* of Chernobyl plutonium can be calculated as follows:

where:

$$F = \left(\frac{A_{238}/A_{239} - 0.04}{0.55 - 0.04}\right) \cdot 100\% \qquad (1)$$

A_{238} - activity concentration of ^{238}Pu, A_{239} - activity concentration of $^{239,240}Pu$. Global fallout component is equal to 100% - F.

Calculation of Surface Deposition

Since the dry mass (m_s) of each complete slice of soil sample was known and the surface S of a cylinder was well defined, the calculation of surface deposition A^ of given radionuclide in one slice was equal to:

$$A_{ss} = m_s A/S \qquad (2)$$

where:
m_s - dry mass of slice [kg], A - activity concentration [Bq/kg].

Total deposition of a given radionuclide in first 10 cm of soil was calculated as the sum of surface deposition in all slices of the initial sample.

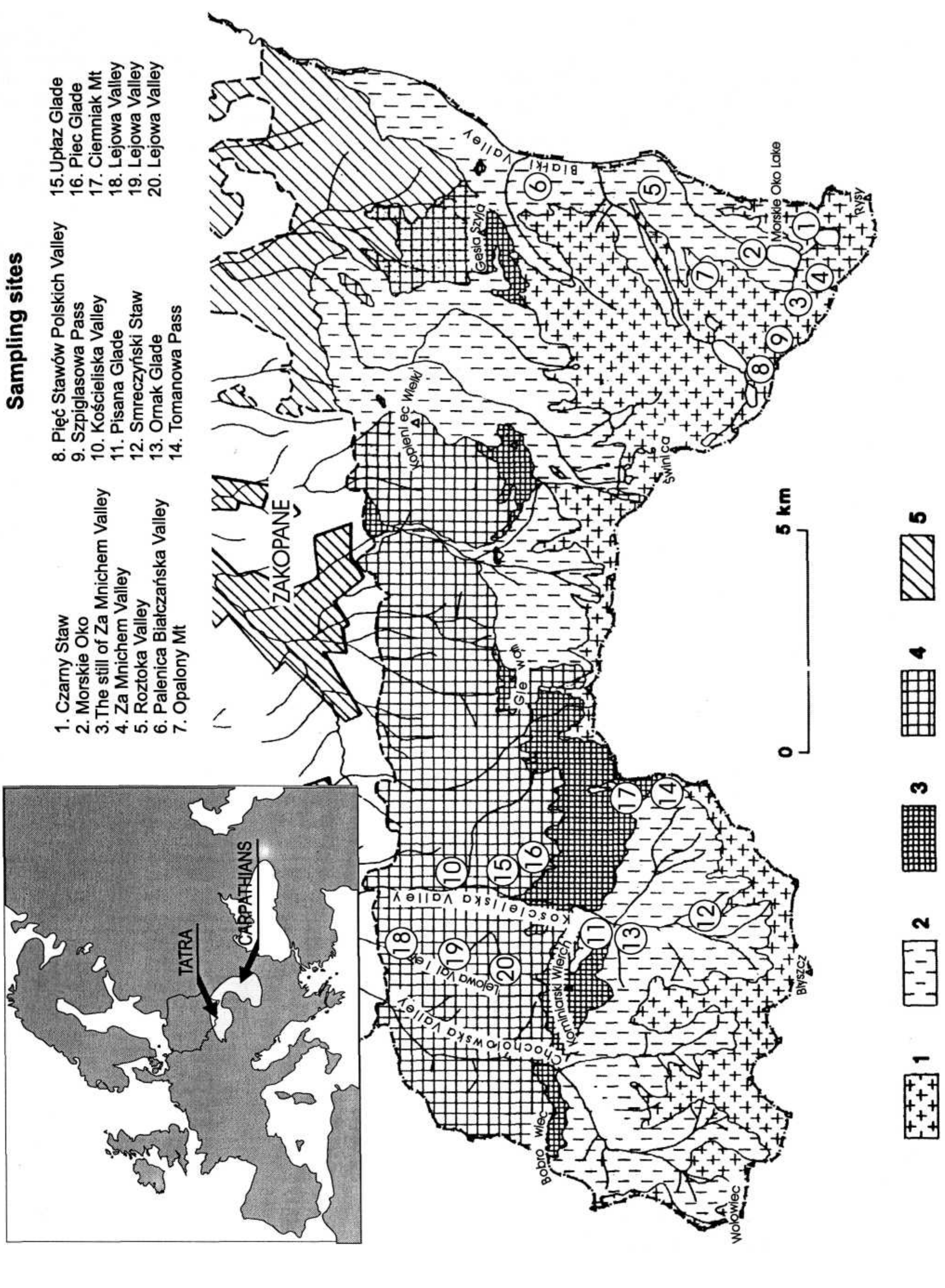


Fig. 1. Sampling site locations on the soil map of Tatra National Park 1 - Leptosols and Regosols, 2 - Orthic Podzols, 3 - Rendzic Leptosols, 4 - Rendzic Leptosols and Eutric Cambisols, 5 - Eutric Cambisols and Dystric Cambisols, 6 - sampling points.

Results and Discussion

Contamination of soil samples by artificial (^{137}Cs, ^{238}Pu and $^{239+240}Pu$) and natural (^{40}K) radionuclides from the two main Tatra valleys have been investigated. The spatial distribution and the altitude dependence were main problems of concern. The investigated samples of soil were collected in August and October of the years 2000 and 2001.

Rybi Potok Valley is situated in the eastern part of Tatra National Park, Koscieliska Valley to the west. These two valleys are geologically different. For example: Rybi Potok Valley is formed by crystalline rocks (granitoides) while Koscieliska Valley is built of metamorphic rocks, limestone, and sedimentary rocks (dolomites and sandstone) (Fig. 1).

The samples were collected from these two main regions regarding altitude. The results were obtained from three or four layers of soils (a - from depths 0-3.5 cm, b - 3.5-7 cm and c - 7-10 cm for the tree layers or a - from depts 0-2.5 cm, b - 2.5-5 cm, c - 5-7.5 cm and d - 7.5-10 cm) for the γ-emitters mainly ^{137}Cs and ^{40}K and α-emitters $^{239+240}Pu$ are presented in Table 1 and Figs. 2, 2a, 3, 3a. All activities refer to dry mass of the samples and they were calculated for the dates of 01.09.2000 and 01.09.2001 respectively, for each part of the samples.

In spite of the relatively small areas of Rybi Potok Valley and Koscieliska Valley (each valley is 10-12 km long and 2-4 km wide) the concentrations of caesium and plutonium were discriminated with altitude. In the samples from Rybi Potok Valley region ^{137}Cs activity varied from 1928 ± 3 $Bqkg^{-1}$ (Dolinka Za Mnichem Valley 1900 m asl)to 161 ± 4 $Bqkg^{-1}$ (Palenica Bialczanska Glade). In the samples of Koscieliska Valley region it was from 1294 ± 26 $Bqkg^{-1}$ (Mountain Pasture Piec 1470 m asl) to 149 ± 12 $Bqkg^{-1}$ (Mountain Pisana Pasture 1015 m asl).

The results for α-emitter $^{239+240}Pu$ were comparable in both regions; however, the maximum concentration of plutonium is observed in Dolinka Za Mnichem Valley (17.0 ± 0.9 $Bqkg^{-1}$) while in Palenica Bialczanska Glade this value was 0.38 ± 0.03 $Bqkg^{-1}$ (Fig. 3).

A few samples were taken from the western region of the Tatras to get meaningful results, thus it was difficult to evaluate any relationship between activity and altitude (Fig. 3a).

The concentration of caesium and plutonium in soil samples increases with altitude and depends on the soil surface. The activity concentrations of caesium and plutonium generally decrease with depth and the greatest values were determined in the uppermost soil layers (in a- the first and b- the second layer). In the case of the main valleys, the highest concentration of investigated

Table 1. The contents of activity of ^{137}Cs, ^{40}K, $^{239+240}p_u$ and ^{238}Pu in soil samples from two main valleys from Tatra National Park (Rybi Potok Valley and Koscieliska Valley). The sample number refers to the sampling site number on the map (Fig. 1).

No	Place	Layer	Cs-137 Ba/kg	K-40 Bq/kg	Pu-239+240 mBq/kg	Pu-238 mBq/kg
			Rybi Potok Valley region			
1	Czarny Staw lake near Rysy Mt 1580 m asl	a	750 ± 77	523 ± 37	7219 ± 366	328 ± 146
		b	94 ± 2	1020 ± 32	1264 ± 99	71 ± 36
2	Morskie Oko 1393 m asl	a	591 ± 13	60 ± 9	3332 ± 185	193 ± 15
		b	713 ± 15	428 ± 19	12142 ± 654	338 ± 23
		c	15 ± 9	557 ± 26	499 ± 33	21 ± 16
3	The still of Dolinka Za Mnichem Valley 1770 m asl	a	782 ± 14	42 ± 28	353 ± 21	53 ± 22
		b	1167 ± 19	96 ± 28	4182 ± 220	218 ± 27
		c	675 ± 10	452 ± 18	5483 ± 255	617 ± 56
4	Dolinka Za Mnichem Valley 1900 m asl	a	1928 ± 3	213 ± 15	17103 ± 915	503 ± 40
		b	1957 ± 35	227 ± 12	3978 ± 213	113 ± 15
		c	390 ± 8	229 ± 17	586 ± 40	34 ± 21
		d	83 ± 2	349 ± 14	262 ± 24	34 ± 23
5	Roztoka Valley 1031 m asl	a	84 ± 2	814 ± 76	2820 ± 137	4 ± 15
		b	7 ± .7	826 ± 77	1486 ± 104	23 ± 15
		c			356 ± 41	23 ± 10
6	Palenica Białczańska Glade 980 m asl	a	161 ± 4	138 ± 20	353 ± 27	16 ± 14
		b	205 ± 4	690 ± 26	1719 ± 100	61 ± 30
7	Opalony Mt 2124 asl	a	790 ± 13	96 ± 3		
		b	177 ± 8	321 ± 6		
		c	36 ± 2	290 ± 3		
8	Pięć Stawów Polskich Valley 1740 m asl	a	881 ± 7	42 ± 0.3		
		b	264 ± 6	422 ± 9		
		c	41 ± 3	415 ± 36		
9	Szpiglasowa Pass 2114 m asl	a	396 ± 25	181 ± 17		
		b	368 ± 13	373 ± 12		

Table 1. Continuation

No	Place	Layer	Cs-137 Ba/kg	K-40 Bq/kg	Pu-239+240 mBq/kg	Pu-238 mBq/kg
			Kościeliska Valley region			
10	Kościeliska Valley near chapel 970 m asl	a b	316.25 213.71	987 1725		
11	Pisana Glade 1015 m asl	a b c	143 ± 12 120 ± 13 95 ± 7	488 ± 28 772 ± 60 742 ± 16		
12	Smreczyński Staw 1226 m asl	a b c	577 ± 35 274 ± 48 67 ± 48	226 ± 6 314 ± 14 344 ± 23		
13	Ornak Glade 1108 m asl	a b c	447 ± 9 455 ± 9 52 ± 1	447 ± 40 598 ± 24 694 ± 23		
14	Tomanowa Pass 1683 m asl	a b c	729 ± 17 179 ± 4 78 ± 2	521 ± 50 694 ± 28 722 ± 69		
15	Upłaz Glade 1350 m asl	a b c	194 ± 5 163 ± 4 189 ± 5	365 ± 36 377 ± 17 455 ± 46		
16	Piec Glade 1470 m asl	a b c	1294 ± 26 507 ± 9 306 ± 6	226 ± 25 219 ± 12 207 ± 20		
17	Ciemniak Mt 2096 m asl	a b c d	631 ± 14 60 ± 2 16 ± 1 7 ± .9	873 ± 80 1158 ± 111 1181 ± 115 1196 ± 121		
18	Lejowa Valley 913 m asl	a b c	301 ± 7 233 ± 5 107 ± 3	761 ± 69 804 ± 84 30 ± 13	97.7 ± 11 111.1 ± 22 9 ± 0.3	60.3 ± 8 53.8 ± 11 18.2 ± 2
19	Lejowa Valley 927 m asl	a b c	538 ± 12 423 ± 7 123.0 ± 3.0	84 ± 21 85 ± 22 611 ± 22	1030 ± 55 7202 ± 353 2598 ± 132	64 ± 16 1361 ± 172 78 ± 41
20	Lejowa Valley 960 m asl	a b c	87 ± 2 121 ± 3 97 ± 2	348 ± 33 452 ± 18 514 ± 49	69.2 ± 8 27.4 ± 7 34.5 ± 9	50 ± 24 13.5 ± 6 20 ± 12

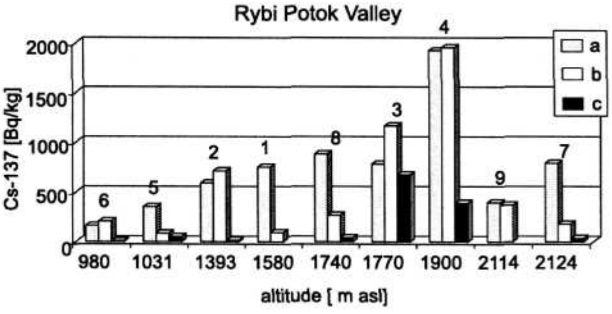


Fig. 2. Altitude dependence of ^{137}Cs activity (Bq/kg) in three soil samples (a, b, c) from Rybi Potok Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

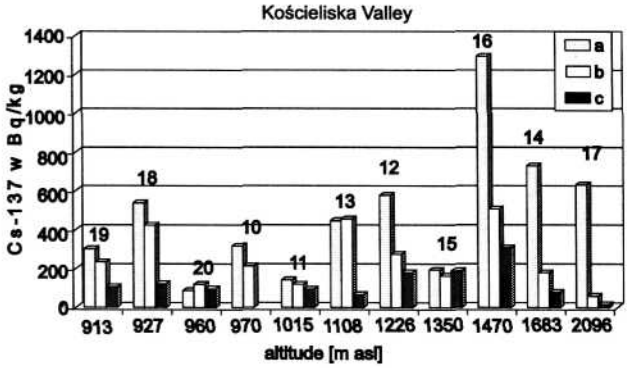


Fig. 2a. Altitude dependence of ^{137}Cs activity (Bq/kg) in three soil samples (in a, b, c layers) from Koscieliska Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

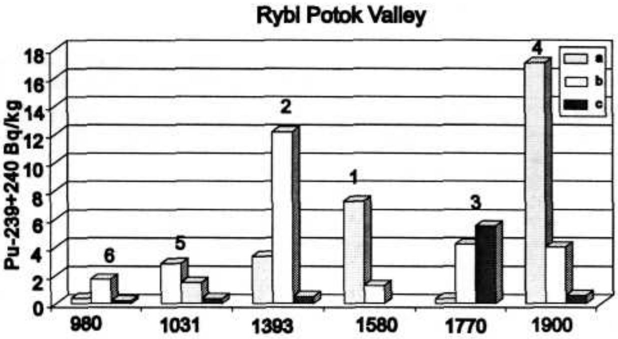


Fig. 3. Altitude dependence of $^{239+240}$Pu activity (Bq/kg) in soil samples (in a, b, c layers) from Rybi Potok Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

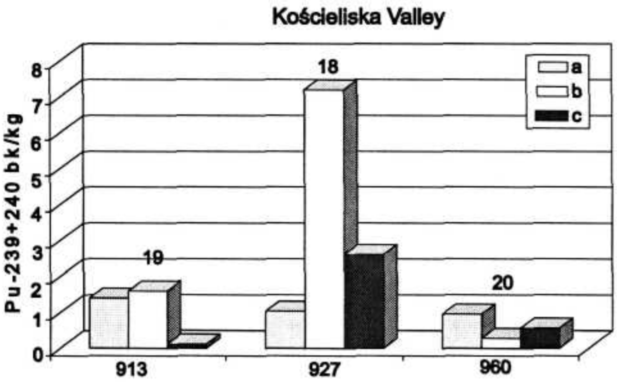


Fig. 3a. Altitude dependence of $^{239+240}$Pu activity (Bq/kg) insoil samples (in a, b, c layers) from Koscieliska Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

radionuclides was observed above 1200 m asl (for instance in Dolinka Za Mnichem Valley or in the vicinity of Czarny Staw near the Rysy in Rybi Potok Valley region and the Mountain Pasture Piec or the Smreczynski Staw in Koscieliska Valley region).

The radiocaesium and plutonium inventory gives the values for total deposition (Figs. 4, 4a and 5, 5a).

The surface activities Asurf in the layers were calculated according to equation 2.

In Dolinka Za Mnichem Valley (site 4 on Fig. 1) ^{137}Cs activity was 17.2 ± 2 kBqm^{-2}, in Palenica Bialczanska Glade (site 6 on Fig 1.) 5.8 ± 1.1 kBqm^{2}, the Mountain Pasture Piec (site 16 on Fig. 1) 14.1 ± 2 kBqm^{2} but in the Lejowa Valley (site 19 on Fig. 1) the concentration of ^{137}Cs was 5.5 ± 1.1 kBqm^{-2} (Figs. 4, 4a). The obtained results of gamma spectrometric measurements for caesium isotopes of the soil samples for the profiles of the investigated regions match those presented by Strzelecki et al. for Carpathian Foothills [10]. Chelmicki et al. [11] compared both concentrations of ^{137}Cs in soil samples (from Wielickie Foothills) and the properties of soils (pH, humus, exchangeable potassium) and no relationship was observed between these parameters and the concentration of caesium. In this work, those parameters have not been measured. The difference in caesium and plutonium concentrations in soil samples can be caused by climatic conditions.

The Tatra's climate is formed by the air masses of various origin and different physical properties which, interacting with air masses of various substrates, creates a variety of climatic types. Considerable climatic diversity in the vertical profile, a great abundance of local climates of cirques, valleys, slopes, passes and ridges, and a rich mosaic of microclimates connected with various kinds of vegetation occuring in the Tatra Mts. There are about 100 days with snow along the north border of the Tatras whereas in the highest region, the number of such days is 290 [2].

The increase of caesium and plutonium concentrations in the first layer of soil on the floor of the highest hung valley or cirque may be due to the swilling of the

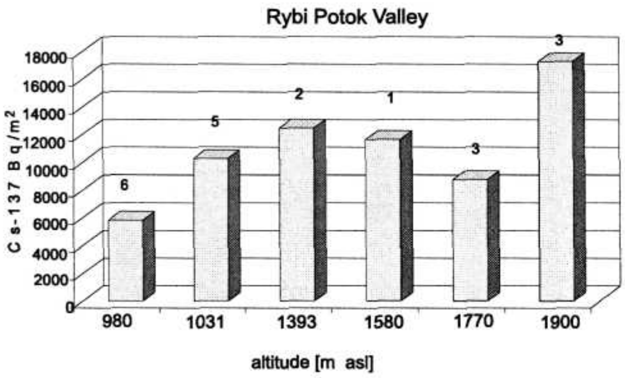


Fig. 4. Altitude dependence of ^{137}Cs activity (total deposition in Bq/m^{2}) in soil samples from Rybi Potok Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

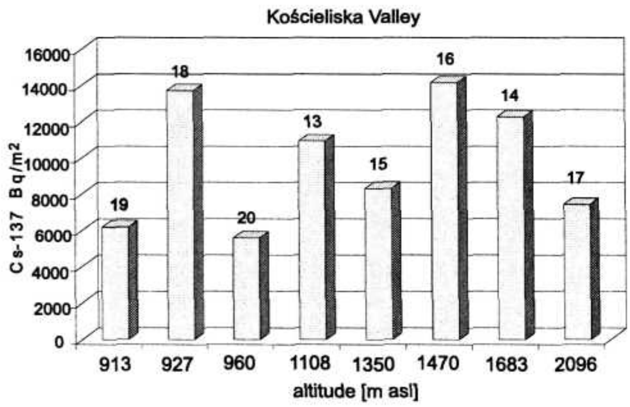


Fig. 4a. Altitude dependence of ^{137}Cs activity (total deposition in Bq/m^{2}) in soil samples from Koscieliska Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

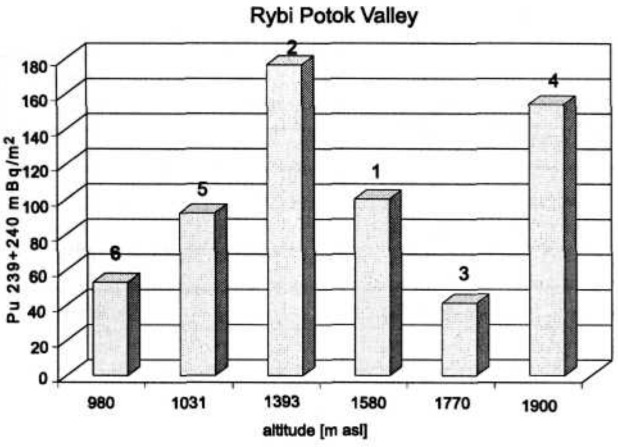


Fig. 5. Altitude dependence of $^{239+240}$Pu activity (total deposition in Bq/m^2) in soil samples from Rybi Potok Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

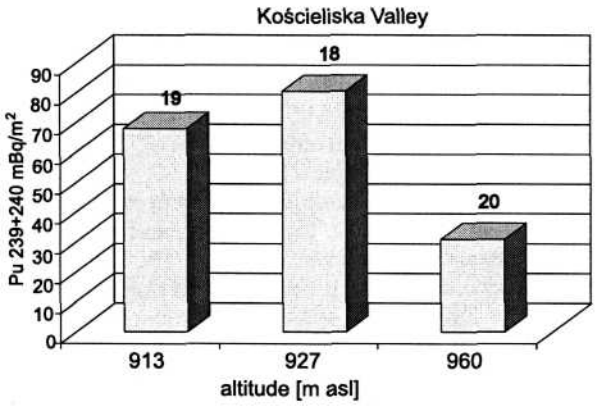


Fig. 5a. Altitude dependence of $^{239+240}$Pu activity (total deposition in Bq/m^2) in soil samples from Koscieliska Valley. Numbers above the bars refer to sampling sites (see Fig. 1).

When two apparent out layers are rejected the least square fit gave the slope which is exactly the global fallout value for ^{238}Pu to $^{239+240}$Pu activity ratio, each out layer contains most likely a hot particle from Chernobyl fallout. Only two points significally deviate from the linear dependence in Fig. 6. The highest activities for these two samples may suggest the presence of plutonium originated from "hot spot" from Chernobyl accident [9, 12].

We tried to find the correlation between the ^{137}Cs and $^{239+240}$Pu concentrations in all soil samples in both analysed valleys. Only for the first two soil layers can the correlation be found.

The fate of caesium in organic soils is different than in mineral ones [9,13]. No correlation was observed between the caesium concentration and organic matter content in soil samples.

The deposition of ^{137}Cs in mountain soil samples, which goes up to 17.2 ± 2kBq/m^2, is about 30% of maximum ^{137}Cs deposition observed in Poland till now [10]. The plutonium maximum deposition of 170 Bq/m^2 is about three times more than average predictions (58 Bq/m^2) for Polish latitudes given in the UNSCEAR report [16]. In Dolinka Za Mnichem Valley, for instance, the inventory in analysed layers seems to be a complete content, since the concentrations in the deepest analysed layer were very small. Although the activities of ^{134}Cs have been too small to make a precise analyses of the radioceasium origin, the ratio between ^{137}Cs and plutonium (which came practically solely from global fallout) suggests that in the vicinity of a half of ^{137}Cs activity comes from the global fallout, while the another half is from Chernobyl. This means that global fallout deposition in Tatra mountains was about three times higher than the average in the country. The increase of the level of radionuclides, activity concentration might be a result of higher deposition caused by atmospheric precipitates (rain or snow). In conclusion, Chernobyl-origin radioceasium deposition was thus rather moderate, on a level not exceeding 10 kBq/m^2. This is almost ten times less than the radionuclide out of the peak. This phenomenon was observed in the region of Morskie Oko lake. Taken together, in the steeply mountain-side region the caesium and plutonium concentrations increase five times to compare with the activities of these radionuclides in the mountain-top region (for example Dolinka Za Mnichem valley (1900 m asl, "4" in Fig. 1) 1927 ± 3 Bqkg^{-1} for the ^{137}Cs and 17.0 ± 0.9 Bqkg^{-1} for $^{239+240}$Pu while Szpiglasowa Pass (2124 m asl, "9" in Fig. 1) 396 ± 25 Bqkg^{-1} for the ^{137}Cs and 3.33 ± Bqkg^{-1} for $^{239+240}$Pu.

Some interesting observations may be made when considering the correlation between $^{239+240}$Pu and ^{238}Pu activities. The results are presented in Fig. 6. The determination of plutonium activity concentration (^{238}Pu . $^{239+240}$Pu) allowed the Chernobyl and global fallout components of the plutonium activities to be ascertained [6, 9]. The percentage of Chernobyl plutonium (F) can be calculated according to equation 1.

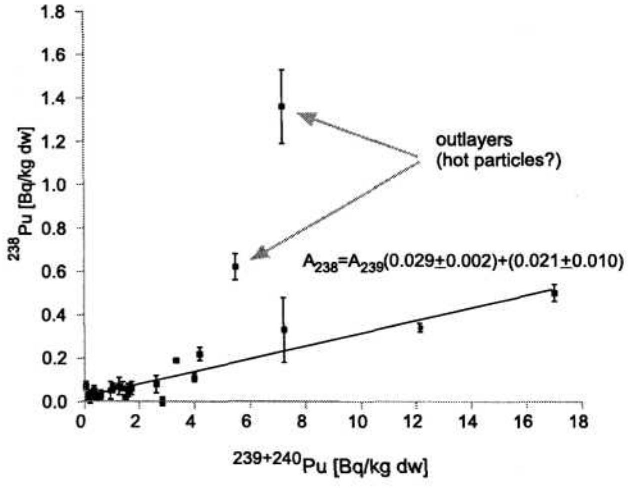


Fig. 6. Linear regressive plot (R^2 = 0.89, $p < 0.0001$) for ^{238}Pu and $^{239+240}$Pu activities in all examined soil samples from the Tatra Mountains.

Table 2. The contents of heavy metals in soil samples from the Koscieliska Valley region and Poroniec Glade (mg/kg).

Altitude [m asl]	Zn mg/kg	Fe mg/kg	Pb mg/kg	Cd mg/kg	Ni mg/kg
The Kościeliska Valley (950)	96.5	1837	18.7	0.3	4.9
The Lejowa Valley (970)	81.9	933	25.0	0.6	2.0
The Pisana Glade (1015)	124.7	20673	74.8	0.5	27.4
The Ornak Glade (1108)	66.8	424	25.6	0.3	5.9
The Smreczyński Staw (1226)	49.8	7989	113.8	6.2	16.6
The Iwaniacka Saddle (1459)	73.9	124	1.9	1.6	1.9
The Wierch Poroniec Glade (1105)	107 – 229		40 – 143.0	1.2 – 4.4	

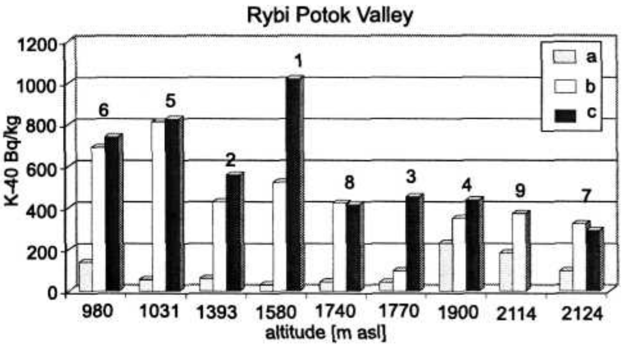
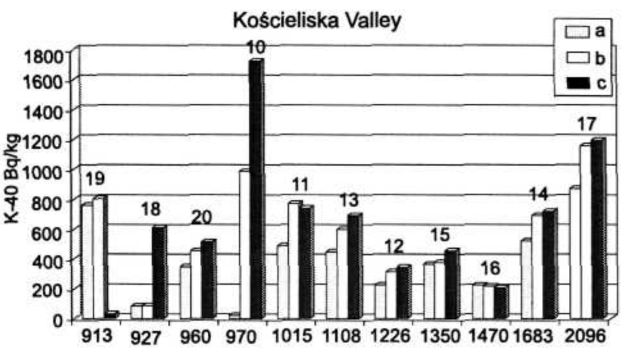


Fig. 7. Altitude dependence of ^{40}K in the soil samples (at a, b, c levels) from Rybi Potok Valley.

Fig. 7a. Altitude dependence of ^{40}K in the soil samples (at a, b, c levels) from Koscieliska Valley.



radioceasium concentration observed in Alps of Austria [17], Switzerland [18] or Germany [19, 20] or in high mountains of Norway [21].

Potassium (^{40}K natural) activity concentration for all soil samples was measured (Fig. 7 and 7a).

The presence of natural long-lived gamma emitters are due to the connected mineral bedrock. The activity of ^{40}K was directly proportional to total activity of potassium in soil; 1 g of potassium contains always 31.7 $Bqkg^{-1}$ of ^{40}K. The highest activities of potassium were found in layer "c" (from depth of 7-10cm). The difference in the potassium concentration is not related to altitude. ^{40}K activity was compared with the value of potassium concentration for Carpathian Foothills [11, 14].

Preliminary investigation of content of heavy metals (Zn, Pb, Cd, Ni and Fe) in soil samples taken from Koscieliska Valley was done. Generally, the concentration of these metals does not differ significally from the values of concentration presented by Miechowka and Mikotajczyk for samples taken from east part of Tatra, from Poroniec Glade. [15] (Table 2).

The majority of the examined soil samples were polluted by these metals to a high degree, and in some of that the permissible concentrations of Pb, Cd and Fe were even exceeded. The parent rocks of these soils contain: 52-129 Zn, 8-27 Pb and 0.4-1.0 Cd (mg/kg). An interesting observation may be made of soil samples from Smreczynski Staw and Pisana Glade. In the sample from first place there was a higher concentration of lead and cadmium whereas in the second one a higher concentration of zinc and iron was observed.

The concentration of these metals in soil samples might depend of the content of organic matter and communication pollution.

A comprehensive survey of world-wide emissions of heavy metals and radionuclides into the atmosphere show that the anthropogenic impact on the environment is becoming a decisive factor in the global cycle of many elements. The main sources of these pollutants in the atmosphere are industrial processes, thermal power stations, domestic heating systems and motor vehicles. In addition to the very high pollution levels in densely populated industrial areas, one has to consider the long range atmospheric transport of pollutants across national boundaries.

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