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# PRELIMINARY CHARACTERIZATION OF THE RADIOACTIVITY IN SEDIMENTS TAKEN FROM JAMSKE PLESO LAKE (SLOVAKIA)

## WSTĘPNA OCENA ZAWARTOŚCI RADIONUKLIDÓW W OSADZIE DENNYM POBRANYM Z JAMSKIEGO STAWU (SŁOWACJA)

**Abstract:** Jamske Pleso lake is one of almost 200 lakes located in the Tatra Mountains. This post glacial water reservoir is set on a strictly protected area. In this work, sediment samples taken from Jamske Pleso Lake were analysed for radioactivity of natural $^{226}$Ra, $^{210}$Po, $^{210}$Pb and artificial $^{137}$Cs. The sediment core was taken by use of Limnos corer. After preparation of the physical sample, gamma measurements were executed in hermetically closed vessels. A preliminary study showed that concentration of natural radionuclides is in typical values for lakes located in this area. An elevated concentration of $^{226}$Ra (mean 57 ± 29 Bq · kg$^{-1}$) is an effect of uranium rich bedrock of the lake. Radioactivity of $^{137}$Cs is close to that reported in other Tatras lakes. Using $^{137}$Cs as an indicator allowed a preliminary estimate of the depth of the sediments deposited in 1960s.

**Keywords:** sediments, Jamske Pleso, artificial and natural radionuclides, radioecology

## Introduction

Among many types of chemical and biological pollutants of the natural environment radioactivity is becoming more of a concern in recent years. Radioactive elements can be divided into two groups: natural and artificial (anthropogenic).

Natural radionuclides have always been present in the environment. Most of them exist in nature only in trace amounts. On the other hand, it is recommended to know the concentration of radionuclides like $^{40}$K, $^{222}$Rn, or $^{226}$Ra due to their contribution to the annual dose of radiation for living organisms. In environmental studies, some naturally occurring radionuclides (e.g., $^{14}$C, $^{210}$Pb) are used for establishing a time scale for different types of samples [1].

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Anthropogenic radioisotopes, like $^{137}Cs$ or $^{90}Sr$, were introduced into the atmosphere during nuclear weapon test in the 1950s and 1960s (with the maximum in 1963) [2]. In Europe, an important source of radioactive contamination was also the nuclear accident in the Chernobyl power plant (1986) [3]. In the case of sediments from European lakes and rivers, vertical distribution of $^{137}Cs$ usually allows one to use this nuclide as a time marker [4, 5]. Typically observed vertical distribution of $^{137}Cs$ in sediments reveals exponential decrease in the radioactivity, with two distinguish peaks. As the sediment is deposited from the top, the deeper peak can be ascribed to the global fallout after nuclear weapons testing (1963). The peak observed closer to the top of the profile, can indicate younger layer, deposited during Chernobyl fallout (1986).

The composition of lake sediments is strictly related to the condition of the environment at time of their formation. For this reason, sediments can be used as an indicator of the condition of the environment and as an inventory of historical information about the reservoir [6–8]. Despite the fact that information about water composition in Tatra Mountains lakes is well documented, only a limited number of studies encompassed lake sediments [9–14]. Most of them focused on chemical composition or paleolimnological studies. A small number of lakes were subjected to radiometric analysis, and only a few included sample dating. In order to fill this gap, Jamske Pleso Lake was selected for this research. The chemical composition of sediments and water from this lake was reported by Stuchlik et al. in 2006 [12]. The proposed research delivers more complex information about radioactivity of natural $^{210}Po$, $^{210}Pb$, $^{226}Ra$ and artificial $^{137}Cs$ in sediments. Selected reservoirs in the vicinity were subjected to radiometric analysis, thus, these analyses gathered information that will enrich our knowledge about the radiological situation in the Tatra Mountain area. In the study the cores of the sediments were sampled and sliced in a way that preserved natural layer stratigraphy. Each layer was analysed separately. This approach will allow to perform geochronological research, and thus determine the historical changes in sediment properties.

This work proposes the application of gamma spectrometry in the analysis of small amounts of sediment samples. Direct analysis of $^{137}Cs$ radioactivity and the indirect measurements of $^{226}Ra$ content via daughter isotopes. Based on the $^{137}Cs$ radioactivity, preliminary dating, using this radionuclide as time marker, was performed.

In order to determine $^{210}Pb$ radioactivity the radiochemical procedure of daughter $^{210}Po$ determination was established.

## Methods

### Study area

The Carpathians are the second largest European mountain range. They cover an area of about 190 000 $km^2$ on the territory of eight countries. The highest parts of the Carpathians are the Tatra Mountains, covering 785 $km^2$. They form a natural border between Poland and Slovakia, where the highest peaks exceed 2600 m. The Tatras were created in Alpine orogeny, which makes the landscape similar to the Alps, but with

significant lesser altitudes. The Tatra Mountains are known for their high number of mountain lakes (over 200), which made them attractive for scientists researching climate changes and anthropopression on the natural environment. One of the post glacial lakes located in the Slovak part of the High Tatras is Jamske Pleso. This small lake can be found in the Vazecka Dolina Valley, one of the shortest Tatra valleys located in the southern slope of Kryvan (Fig. 1).

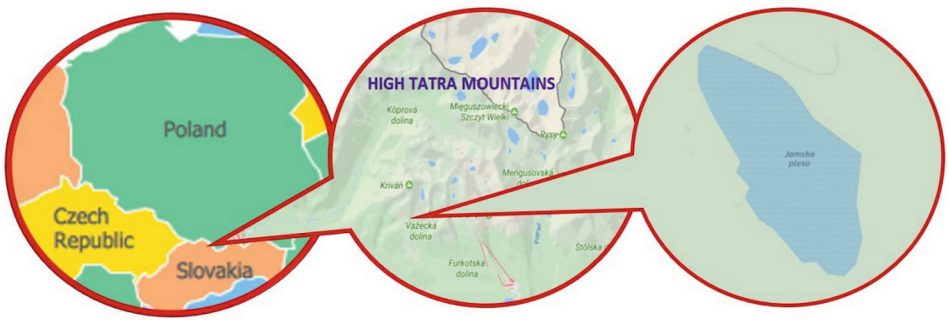


Fig. 1. Location of Jamske Pleso Lake

The Jamske Pleso Lake lies at 1447 m a.s.l. and covers an area of 0.069 km$^2$. Rich organic life in the lake makes the water transparency low, up to 2.5 m. The shore line is not diversified. In the beginning of the 20$^{th}$ century, a small hotel was located on the northern bank of the lake [15]. Currently only a hiking trail passes near the lake. The rest of the Vazecka Dolina Valley became strictly protected area and is not accessible to tourists.

## Sampling

Samples of sediments from Jamske Pleso Lake were collected in September of 2013 by the use of a Limnos gravity corer. During the sampling procedure, the construction of the corer allows the laminar flow of the water, thus the collected sediments layers remains untacked. In order to obtain longer cores, sampling set was equipped with led weights. The sampling point was located in the centre of the lake (49°7′50.0″ N; 20°0′45.1″ E). Cores were collected by use of inflatable boat. The sampling point coordinates were established by the Garmin GPS receiver. Cores of a 10 cm inner diameter were transported to the lake shore, and divided in-situ into 1 cm high layers. Each layer was packed separately into a polyethylene vessel.

## $^{137}$Cs and $^{226}$Ra determination

Radioactivity of artificial $^{137}$Cs and natural $^{226}$Ra was determined by means of gamma spectrometry. Sediment samples were dried in ambient temperature in order to prevent from the volatile $^{210}$Po loss. The samples were dried until constant mass was

reached. In order to obtain homogeneous, representative samples the sediments were pulverized by use of blade grinder, and sieved. The fraction of grain size below 0.2 mm was selected for the analysis. Samples were packed into 1.5 $cm^3$ hermetically sealed vessels and stored for at least 4 weeks. During the storage period the transient equilibrium between $^{226}Ra$ and its progeny $^{214}Pb$ and $^{214}Bi$ was reached. The hermetic vessels prevented from the gaseous $^{222}Rn$ (direct product of $^{226}Ra$ decay) loss. Radiometric measurements were performed on a broad energy high purity coaxial germanium detector (Canberra model BE3830, relative efficiency 34 % for 1333 keV). Counting time was 72 hours.

The $^{137}Cs$ (its half-life is $T_{1/2} = 30.07$ years) was measured directly at 661.6 keV, the radioactivity of $^{226}Ra$ ($T_{1/2}$ = 1599 years) was determined indirectly. Two gamma daughter isotopes of $^{226}Ra$: $^{214}Pb$ and $^{214}Bi$ were measured at energies of 295.2, 351.9 keV and 609.3, 1120.3 keV, respectively. The content of $^{226}Ra$ was calculated as the mean value of these four energies. The correctness of the method was checked by measuring the reference materials obtained from the international Atomic Energy Agency (Vienna): IAEA-447 and IAEA-RGU-1.

## $^{210}Po$ and $^{210}Pb$ determination

The radioactivity of natural $^{210}Pb$ was determined indirectly via daughter $^{210}Po$, by means of alpha spectrometry. As the equilibrium between the $^{210}Pb$ and $^{210}Po$ in sediment samples is disturbed due to differences in chemical properties. The method involves the double determination of the daughter isotope $^{210}Po$ with at least a six months interval. The content of $^{210}Pb$ can be than calculated from the growth of polonium radioactivity by an equation described by Szarlowicz et al. [16].

The determination of the radioactivity of $^{210}Po$ required an additional radiochemical procedure. Due to low amount of samples, alpha analysis were performed after the gamma spectrometry measurements. In order to determine the chemical recovery of the procedure for each sample separately, the sediment samples of about 0.1 g were spiked with known amount of a $^{208}Po$ tracer solution. Samples were decomposed in a MAGNUM-II (Ertec) microwave digestion system. The digestive agent was a mixture of concentrated $HNO_3$ and concentrated HCl (4 : 2 v/v), maximum digestion pressure in the digestion vessel reached 36 atm. Silicate residue was centrifuged at 3000 rpm for 10 minutes and discharged. The solution was transferred into small PTFE bakers and evaporated to near dryness. In order to control the evaporation process infrared heaters were used. After the evaporation a few portions of 2 mol · $dm^{-3}$ HCl were added in order to remove residual $HNO_3$. Each portion was also evaporated. Polonium was spontaneously plated on a silver disc from a 0.5 mol · $dm^{-3}$ HCl solution. In order to prevent competitive plating of $Fe^{3+}$ ions, tri-sodium citrate and hydroxylamine were also added. The plating was carried out at 85°C for 3 hours. After the reaction is completed, the plating set was dismantled and the obtained alpha source was rinsed several times with deionised water. The alpha sources were measured on an alpha spectrometer (Canberra model 7401, equipped with a Passivated Implanted Planar Silicon (PIPS) detector, with active area of 450 $mm^2$ efficiency 34 %). Counting time was 72 h.

## Results and discussion

The gamma spectrometric measurements allowed to determine the radioactivity of $^{137}$Cs in small samples of sediments. The maximum concentration of $^{137}$Cs was found in the uppermost layers (687 ± 10 Bq · kg$^{-1}$) and decreased exponentially with depth. Single discontinuity was found at a depth of 6–7 cm (Fig. 2), which can be attributed to the global fallout after intensive nuclear tests (1963). The content of this radioisotope is similar to that reported in other lakes located in the vicinity [17]. The elevated radioactivity of caesium in the uppermost layers precludes the identification of the second peak, which could be ascribed to the Chernobyl fallout (1986) [18]. This elevated radioactivity is caused by the delivery of caesium from eroded soils, where $^{137}$Cs was delivered during mentioned events.

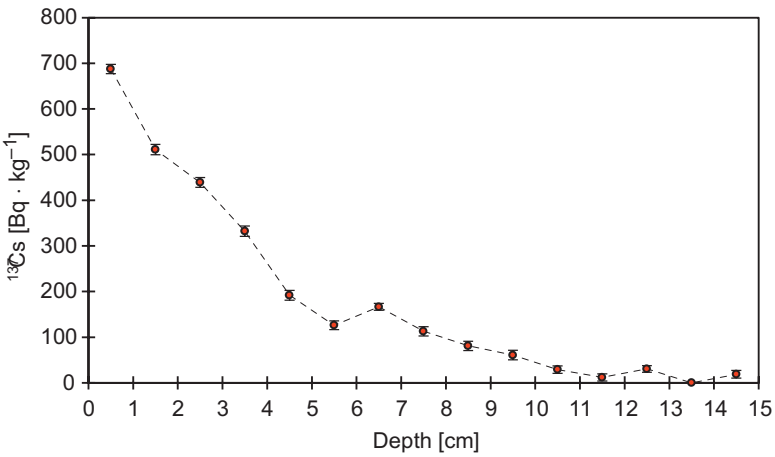


Fig. 2. Vertical distribution of $^{137}$Cs in sediment from Jamske Pleso Lake

Radioactivity of natural $^{226}$Ra ranged from 34 ± 17 Bq · kg$^{-1}$ to 105 ± 24 Bq · kg$^{-1}$. The vertical distribution of radium in the profile is poorly diversified, no clear pattern can be identified. The calculated arithmetic mean of 57 ± 29 Bq · kg$^{-1}$ is a typical value observed for sediments and soils in the High Tatras, which is almost two times higher than average for the lowland areas [19]. This elevated radioactivity of $^{226}$Ra is the effect of the uranium, present in the crystalline core of Tatra Mountains.

Radioactivity of $^{210}$Po ranged from 5.07 ± 0.67 Bq · kg$^{-1}$ to 639 ± 40 Bq · kg$^{-1}$ (Fig. 3).

Similar results were reported in Smreczynski Staw Lake in Poland [20]. In all samples, the radioactivity of $^{210}$Po in the first and second depositions was similar, and it can indicate close to the equilibrium state with the parent $^{210}$Pb. The maximum radioactivity of $^{210}$Pb in the uppermost layer was 717 ± 45 Bq · kg$^{-1}$ (Fig. 4.). This is almost two times higher than found in near Zelene Pleso. On the other hand, the value is much lower than in Smreczynski Staw Lake in Poland [17, 20]. The $^{210}$Pb concentration

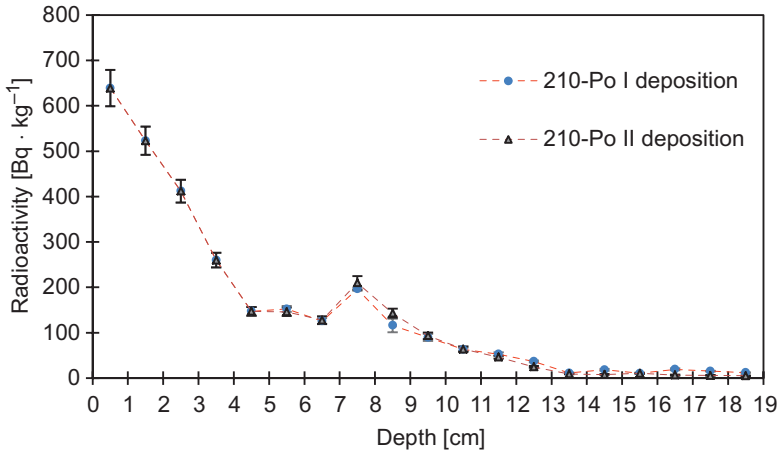


Fig. 3. Vertical distribution of $^{210}$Po in sediment from Jamske Pleso Lake

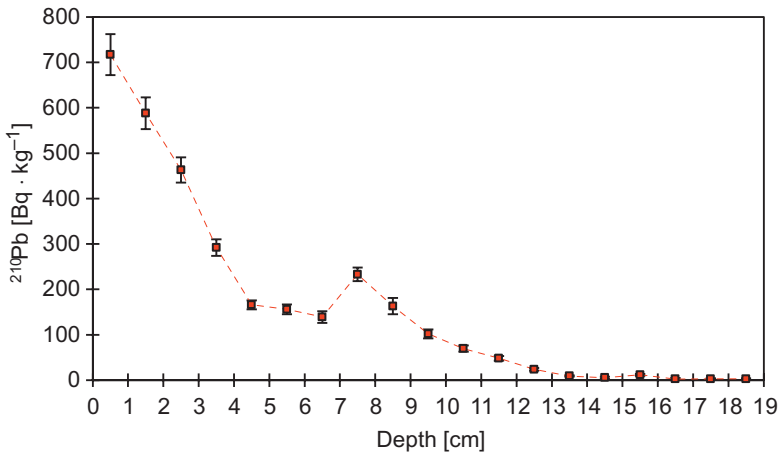


Fig. 4. Vertical distribution of $^{210}$Pb from two depositions of $^{210}$Po in sediment from Jamske Pleso Lake

decreased exponentially along with sample depth. Single discontinuity was found between 6 and 9 cm. The elevated radioactivity of $^{210}$Pb can be a result of excessive material delivery to the lake. Historic data provides information about aeolian dust fallout that occurred in the 1960s, which could be the source of excessive $^{210}$Pb [21]. Moreover, ascribing this date for layers between 6 and 9 cm is confirmed by preliminary $^{137}$Cs which is a time marker for 1963.

## Conclusions

This paper represents preliminary studies about the radioactivity of selected natural and anthropogenic radionuclides in sediments collected from Jamske Pleso Lake (High

Tatra, Slovakia). The Jamske Pleso lake is located on a uranium reach bedrock, thus the content of natural $^{226}$Ra, $^{210}$Pb and $^{210}$Po is elevated in comparison to the lowland areas. Based on the research, the radioactivity of artificial $^{137}$Cs is in typical value range for High Tatras lakes. Single disturbance in the radioactivity distribution was suggested to be created in a layer affected by the fallout after nuclear weapons testing (1963) and aeolian dust fallout in 1960's. Natural $^{226}$Ra is distributed evenly in the entire core. The average radioactivity of this element is typical for the studied area, however it is elevated in comparison to the lowland average. Applied radiochemical procedure allowed to determine the radioactivity of $^{210}$Po and to calculate parent $^{210}$Pb radioactivity. The observed values are typical for Tatra Mountains. The vertical distribution of $^{210}$Pb radioactivity can be used to perform geochronology research. Preliminary dating allowed estimating that layers between 6 and 9 cm were created probably in the 1960s.

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## WSTĘPNA OCENA ZAWARTOŚCI RADIONUKLIDÓW W OSADZIE DENNYM POBRANYM Z JAMSKIEGO STAWU (SŁOWACJA)

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**Abstrakt:** Jamski Staw (słow. Jamské Pleso) to jeden spośród niemal 200 tatrzańskich stawów. Ten zbiornik o charakterze polodowcowym położony jest na terenie objętym ochroną ścisłą. Celem niniejszej pracy było wyznaczenie poziomu radioaktywności naturalnych $^{226}$Ra, $^{210}$Po i $^{210}$Pb oraz sztucznego $^{137}$Cs w profilach osadów dennych pobranych z Jamskiego Stawu. Profile rdzeniowe osadów pobrano przy pomocy czerpaka firmy Limnos. Po wstępnej obróbce próbek i umieszczeniu ich w szczelnych naczyniach pomiarowych poddano je analizie z wykorzystaniem spektrometru promieniowania gamma. Wstępne wyniki pokazały, że radioaktywność naturalnych izotopów jest na zbliżonym poziomie, do notowanej w innych stawach tego regionu. Podniesiony poziom radioaktywności $^{226}$Ra (średnia 57 ± 29 Bq · kg$^{-1}$) jest związany z zawierającym uran, granitowym trzonem podłożem skalnym jeziora. Radioaktywność $^{137}$Cs jest na podobnym poziomie co w innych stawach tatrzańskich. Wykorzystanie $^{137}$Cs jako znacznika czasu pozwoliło na wstępną identyfikację warstw osadów nagromadzonych w latach 60. XX wieku.

**Słowa kluczowe:** osady denne, Jamski Staw, sztuczne i naturalne radionuklidy, radioekologia