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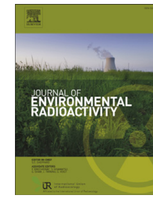

Atmospheric fallout radionuclides in peatland from Southern Poland

Tomasz Mróz [a,*], Edyta Łokas [b], Justyna Kocurek [c], Michał Gąsiorek [d]

[a] Pedagogical University of Cracow, Podchorążych 2 30-084 Kraków, Poland
[b] Institute of Nuclear Physics Polish Academy of Sciences, Radzikowskiego 152, 31-342 Kraków, Poland
[c] AGH University of Science and Technology, Al. Mickiewicza 30, 31-120 Kraków, Poland
[d] Department of Soil Science and Soil Protection, University of Agriculture in Krakow, Al. Mickiewicza 21, 31-120 Kraków, Poland

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Two peat profiles were collected in a peat bog located in Southern Poland and their geochronology were determined using ^{210}Pb, $^{238,239+240}$Pu and ^{137}Cs radiometric techniques. The ^{210}Pb chronologies were established using the constant rate of supply model (CRS) and are in good agreement with the Pu isotopes and ^{137}Cs time markers. Maximum activities of Pu isotopes were found at a depth corresponding to the early 1960s, which is the period characterized by the maximum nuclear weapon tests. The results showed that the ^{210}Pb method is the most accurate technique for the determination age and accumulation rate of a peat. The next part of this study calculated linear accumulation rates by analyzing $^{238,239+240}$Pu and ^{137}Cs vertical distributions in the profiles. Activities of fallout isotopes were also measured in plants covering the peatland. The highest activities of ^{137}Cs and ^{210}Pb were found in *Calluna vulgaris* samples, and $^{239+240}$Pu were found only in two samples (*C. vulgaris* and leaves of *Oxycoccus quadripelatus*).

1. Introduction

Peatlands are a great archive of atmospheric pollutants, such as metals and organic contaminants. In the same way, peatlands can accumulate radioactive elements and isotopes. Natural and artificial fallout radionuclides have been widely applied as a material for studying recent sediments and peat chronology (Turetsky et al., 2004; Appleby, 2008; Le Roux and Marshall, 2011; Łokas et al., 2013; Fijałkiewicz-Kozieł et al., 2014). Accurate dating of this type of sample may be very useful to understand the dynamics of environmental conditions in the past, because peats and sediments are an effective record of many environmental factors on the scale of decades and centuries. In the 1970s, a few methods of using natural and artificial radionuclides for geochronological studies were developed (Appleby, 2008). One such method was the application of the ^{210}Pb isotope developed by Krishnaswamy et al (1971), which can be used for dating to approximately 150 years in the past. This method is often used in parallel with methods based on artificial fallout radionuclides produced in nuclear bomb tests or released from nuclear facilities. Both groups of radionuclides are deposited directly from the atmosphere and are mainly absorbed by fine grained materials, including organic matter. Assuming that, once deposited, elements do not migrate in a peatland environment, we can obtain a continuous chronological sequence. This method is very useful in the determination of peat chronology.

Peatlands are an important element of the natural environment. Specifically, peatlands cover approximately 3% of the Earth and are a significant storage of carbon, accumulated over a very long period. The Northern Hemisphere peatlands store approximately 450 billion tons of carbon, which is approximately 30% of the global soil carbon stock and 75% of pre-industrial carbon mass in the atmosphere. Peatlands are also very sensitive to the impact of climate change. For example, loss of water from a peat profile connected with the warming of peat increases organic matter oxidation rates and emission of greenhouse gases, such as CO_2 and N_2O. In 2005, emission of CO_2 from peatlands was calculated to be $6 \cdot 10^9$ kg year^{-1} (Strack, 2008). Accurate determinations of peat chronology and accumulation rate are important for understanding the impact of climate change on peatland dynamics, not only at the local level, but also on a global scale.

Since peatlands accumulate atmospheric fallout pollution, a multitude of dating and monitoring methods can be used, including stable isotopes analysis and radiometric methods using natural and

* Corresponding author.
E-mail addresses: tmrozek@o2.pl (T. Mróz), Edyta.Lokas@ifj.edu.pl (E. Łokas), Lipiarzj@gmail.com (J. Kocurek), rrgasior@cyf-kr.edu.pl (M. Gąsiorek).

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artificial radionuclides. ^{137}Cs has a relatively short half-life of 30.2 years; more than 60% of the original inventory of global fallout has subsequently decayed, and at some point in the future it will be impractical to routinely measure fallout ^{137}Cs in environmental samples. An alternative fallout radionuclide to ^{137}Cs for determining chronostratigraphic dates is $^{239+240}Pu$ (Jaakkola et al., 1983). In the Podhale region global fallout is the main source of $^{238,239,240}Pu$ isotopes but ^{137}Cs comes from two sources: global fallout and the Chernobyl accident.

Raised peat bog ecosystems deserve more research in radioecology. In early studies, some attention has been given to the transfer of the radioactive caesium isotope ^{137}Cs within ombrotrophic peat bogs (Bunzl and Kracke, 1989). However, the processes that determine caesium uptake and binding in raised bogs are not well understood. Among the most nutrient-poor ecosystems, the raised bogs are characterised by a relatively high ^{137}Cs bioavailability to bog vegetation and mosses in particular (Bunzl and Kracke, 1989; Orlov et al., 1999). Uptake of ions by the moss species occurs by ion exchange and complexation processes. Recent studies (Schleich et al., 2000) show that in Sphagnum layers, Cs is translocated continuously towards to the growing apex of the Sphagnum shoots, where it is accumulated. But the data about the Pu bioavailability and transfer to the fruits and leaves are rather scarce, when compared to radioceasium data. Heather (*Calluna vulgaris*) is a common species in Poland growing in the acidic soils. Heather takes up high amounts of caesium compared with other plant species in the same ecosystem (Harrison et al., 1990). The low pH of the soil favours the uptake of caesium. Isotope transfers seem to be very important because leaves, flowers and, in some cases, fruits provide an important part of the diet of bees and some animals (eg. sheep) so a heather honey or meat of animals are eventually is a part of human diet (Beljaars et al., 1997; Salt et al., 1994). Previous studies (Fijałkiewicz-Kozieł et al., 2014; Mietelski et al., 2007) observed higher activity concnetrations of ^{137}Cs in the upper segments of the peat bog profiles and interpreted this fact as a upward transport of ^{137}Cs through the plants growing in the peat bog. The transfer of ^{137}Cs from peat to plants varies depending on the species, physical and chemical interactions, and localization of the study site. Therefore, the radionuclide avaibility for plants and fruits were compared in this study.

The main aim of the present study was to determine the activity concentrations and deposition of natural (^{210}Pb) and artificial fallout radionuclides (^{137}Cs, ^{238}Pu and $^{239+240}Pu$) in two parts of a small raised peat bog, Puścizna Mała peatland (the Orawsko-Nowotarska Basin, Southern Poland). The results were used for the determination of peat chronology and linear accumulation rates and were compared with other authors. We were particularly interested in identifying a transfer factor of ^{137}Cs and potential role of bog vegetation, including fruits, in the biological retention of ^{137}Cs and $^{239+240}Pu$. Because different plant species have a different transfer factors, we wanted to look for differences in ^{137}Cs age determination between two profiles covered by different plants.

2. Materials and methods

2.1. Study area and sample collection

The Puścizna Mała bog is located in the Orawsko-Nowotarska Basin in the northern part of the Orawsko-Podhalański depression consisting of 26 bogs and a total area of 23.47 km^2. It is located between the Pieniny and Gorce mountains (Fig. 1). Its genesis dates back to the Neogene period. Within the depression located between the hills is a lake, which has begun to fill with sediment gravels and clays. These deposits are now important factors in the development of peat bogs in this area by reducing water outflow and ensuring proper water level for the peat bogs (Łajczak, 2001; Miechówka and Zaleski, 2005). Puścizna Mała comprises an area of approximately 100 ha. It is a raised-type peatland, and the average peat depth in this area is 1.74 m, with a maximum of 4.25 m. The estimated volume of peat is approximately 1,810,000 m^3 (Lipka et al., 2004; Śmieja-Król and Fiałkiewicz-Kozieł, 2014). Analysis of the peat composition shows dominance of two plant species: cotton-grass (*Eriophorum* sp.) and mosses from *Sphagnum* genus. The average decomposition degree (ratio of dark, amorphous matter to the non-decomposed matter in peat (Drzymulska, 2016)) is approximately 15%. Plants growing on this peatland are mainly *Ericaceae,* such as *Vaccinum uliginosum, Calluna vulgaris* and *Oxycoccus quadripelatus*. Trees growing on this area are *Pinus silvestris* and *Pinus mugo* (Lipka et al., 2004).

Two cores were collected in July 2012 using PVC pipes with an internal diameter of approximately 10 cm and a height of 50 cm. The pipes were transported to the laboratory where vegetation was separated from the top layer of the core sample. Vegetation samples were dried and collected in polyethylene bags. Plants collected from the top of the cores and around the cores were acidophilic representatives of the *Ericaceae* family: heathers (*Calluna vulgaris*), cranberries (*Oxycoccus quadripelatus*) and bog blueberries (*Vaccinum uliginosum*). Unripe fruits and leaves were separated from the bog blueberries and cranberries into two different samples. Cores removed from the pipes were subsampled at 2–5 cm layers and dried in a temperature of 105 °C for 24 h. After drying, samples were weighed and milled for homogenization. Due to the high volatility of Po, samples were not combusted (Mabuchi, 1963).

Activities of fallout radionuclides were determined in two peat profiles and in six samples of vegetation collected from the top of the peatland. The analytical procedure was the same for peat and vegetation samples.

2.2. Radioisotope dating

The ^{210}Pb isotope with half life of 22.26 years is produced in the atmosphere by radioactive decay of ^{222}Rn, which is a member of the ^{238}U series. ^{222}Rn is a radioactive noble gas that diffuses into the atmosphere from soil. ^{210}Pb produced by radon decay is deposited on the ground surface. The deposition rate for a selected area is relatively constant; therefore, activities in a core collected from a sampling site have an exponential distribution (Sheets and Lawrence, 1999; Shukla, 2002). There are two models of ^{210}Pb dating: the CIC (*Constant Initial Concentration*) and CRS (*Constant Rate of Supply*) (de Souza et al., 2012). In this study, the CRS model was used, which implies that the atmospheric flux of ^{210}Pb does not change depending on the rate of accumulation of the dry mass of peat and the cross-section area is considered as a reservoir of ^{210}Pb activity (Lubis, 2006). This activity has two components: supported activity that does not change with profile depth and is taken as equal to the activity of ^{226}Ra in the profile, and unsupported activity associated with the delivery of atmospheric ^{210}Pb (Appleby, 2008). We believe this model is suitable for determining age of peat cores because it allows for the accurate determination of core age, despite temporal variations in sedimentation rate, and because peatlands depend solely on atmospheric delivery of ^{210}Pb (Turetsky et al., 2004). The activity of ^{210}Pb isotope in the sample can be directly determined by gamma spectrometry (measurements of 46.5 keV gamma line), but generally it is determined by measuring the activity of alpha-emitting ^{210}Po isotope which is in radioactive equilibrium with ^{210}Pb (Begy et al., 2011; Beilet-Kovalenko et al., 2013). The radioactive equilibrium is met because of the short half-life of the ^{210}Bi isotope ($T_{1/2} = 5.013$ d).

The next method of peat dating is the application of measuring Pu isotope activities in the samples. The main source of Pu isotopes