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Use of lead-210 and carbon-14 in investigations of peat accumulation in Aukštumala raised bog, western Lithuania

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Abstract Two peat cores taken from the Aukštumala raised bog of Lithuanian maritime region were dated by lead-210 (^{210}Pb) and radiocarbon (^{14}C) methods. From combined chronological data the net and linear peat accumulation rates were calculated. Net peat accumulation expressed in terms of average dry mass accumulation rates for the last two hundred years was relatively constant for both cores, varying between 0.006–0.011 $g/cm^2/yr$ for core #1, and 0.008–0.014 $g/cm^2/yr$ for core #2, respectively. Mean linear peat accumulation rates during the last 200 years were the following: core #1 – 0.17 cm/yr, core #2 – 0.19 cm/yr. Recent mean peat accumulation rates were similar to the long-term linear peat accumulation rates (0.11–0.13 cm/yr) based on ^{14}C chronology that indicated a relatively undisturbed condition of Aukštumala bog in the studied areas. The values obtained in this study can be used to demonstrate timing of the environmental changes in the studied region during the last 6,000 years.

Keywords *Lead-210, carbon-14, dating, peat accumulation rate, raised bog, Aukštumala, Lithuania.*

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INTRODUCTION

The Quaternary paleoclimate variability, ecological changes, atmospheric metal pollution, and other environmental effects can be studied using dated peat cores from bogs (Cole *et al*. 1990; Froehlich and Walling 1992; Shotyk *et al*. 2000; Kim and Rejmánková 2002). The use of the fallout radionuclide lead-210 (^{210}Pb) and cosmogenic radiocarbon (^{14}C) as a dating tool appears to offer considerable potential for assembling information on physical, chemical and biotic properties of bogs within the context of a changing environment.

Aukštumala is a raised *Sphagnum* bog located in western Lithuania in the surroundings of the Nemunas Delta. The Nemunas Delta itself and its environs have long been studied with respect to geomorphology and hydrology, geology and paleogeography (Basalykas 1961; Červinskas and Kunskas 1982; Gudelis and Klimavičienė 1993; Bitinas *et al*. 2002). Different views on the geological history of the Nemunas Delta and its environs still exist. Some researchers relate the geological development of the area mainly to successive base level changes of the Baltic Sea (Gudelis 1998). Others indicate the leading role of neotectonic movements (Kunskas 1996) and suggest a concept of a buried Pre-Nemunas River valley (Bjerkéus *et al*. 1994; Savukynienė and Ruplėnaitė 1999).

New geological material was obtained and summarised in 1996–2000 based on geological maps at a scale of 1:50 000 for the northern part of the Nemunas Delta and the entire Lithuanian maritime region (Bitinas *et al*. 2002). The study included pollen, diatom and mollusc analyses, lithological characterisation, radiocarbon and optically stimulated luminescence dates to provide paleogeographical reconstruction of the southern part of the Lithuanian maritime region during the maximum extent of the Baltic Ice Lake (~13,800–13,000 cal. years BP), the Ancylus Lake (~9800–9500 cal. years BP), the Litorina Sea (~7000 cal. years BP) and the Postlitorina Sea (~4500 years cal. years BP) and to construct a geological and geomorphologic model of the Nemunas River Delta. This model suggests that the

Lithuanian part of the Nemunas River Delta (Rusnė island) could have formed in 900–1000 cal. years BP (Bitinas *et al.* 2002).

In terms of landform morphology, hydrology, water chemistry and vegetation that predetermine a distinctive ecosystem with peculiar plant, invertebrate and bird communities, the Aukštumala raised bog is considered to be one of the most important territories in the Nemunas Delta regional park (Management Plan 1999). Aukštumala is the first raised bog in the world, which was comprehensively studied at the end of the nineteenth century (Weber 2002).

The goal of this study was to define chronology and assess past and recent peat accumulation rates of Aukštumala bog.

MATERIAL AND METHODS

The study area comprises Aukštumala bog located very close to the recent Nemunas River Delta, which is represented by a low–lying alluvial plain (Figs. 1, 2). The top altitude of Aukštumala bog surface is 6.8 m above sea level (a.s.l.). Particularly Tenenis and Minija rivers westward and northward and Šyša River southward from the bog drain the area surrounding the bog. Lake Krokų Lankos is located at the south-western edge of the bog. The area of Aukštumala bog is 3018 ha and more than half of it (2627 ha) has been used by peat cutting industry. The reclamation of Aukštumala bog started in 1820, and peat production has taken place since 1882. A large area of the bog was drained in 1969–1988. Peat cutting has a negative impact on the rest of the natural bog ecosystem due to draining and hence lowering the groundwater table (Management Plan… 1999).

The geological settings of the area including the Aukštumala bog environs are shown in Fig. 2 (Bitinas *et al.* 2002). The dominant thickness of peat layer (b IV, bog deposits) in Aukštumala bog is about 6 m, yet the greatest thickness in the depressions of bog bottom may reach 9 m. A thin (up to 1 m) layer of gyttja (1 IV, limnic deposits) is usually underlying the peat layer. Both layers overly a few metres of thick marine Litorina Sea sediments of (sand, m IVL) composing the mineral basis of the bog and its environs. The lower part of the section is composed of Middle Pleistocene sediments including till, gravel, and variously grained sand (g II md).

Investigated sites

Two peat cores from different locations of Aukštumala bog (location of core #1 – N55°23’00”, E21°22’34”’, clastic fraction – up to 3%, biogenic fraction – up to 97% in the upper 0.5 m; location of core #2 – N55°24’18”, E21°19’43”, clastic fraction – up to 7%, biogenic fraction – up to 93% in the upper 0.5 m) were taken with a metallic open–end tube, 15 cm in diameter and 1 m long. Tubes were pushed into the peat to a depth of ~0.5 m. The cores were retrieved by digging small trenches adjacent to the tube, rotating it lengthwise, and lifting it out sideways in order to prevent any loss of the peat column. The distance between the two sites is four km (Fig. 1).

Dating

The ~0.5 m cores were carefully sectioned into 4 cm slices, packed into hermetic beakers, weighed, oven-dried, and reweighed to calculate mass/volume. The dry samples were combusted to 'white ashes' prior to gamma ray assay for ^{210}Pb dating. The homogenised ash samples were examined by direct gamma-ray spectrometry of ^{210}Pb (Kunzendorf *et. al.* 1998). Gamma ray assay was undertaken using 26.2% efficiency HPGe detector with 0.5 mm thick beryllium window. The gamma spectra were calculated using software *Gamma Trac Version 1.21* (Oxford Instruments Inc). A number of naturally occurring radioisotopes from the U and Th decay series, as well as man-made radioisotope ^{137}Cs, were also determined. As to natural radioactivity, special attention has been paid only to the ^{210}Pb activity concentration in samples because remaining gamma-radiation was often attributable to variation of the background. The gamma-ray spectrometric system has been calibrated against samples with known ^{210}Pb and ^{137}Cs activities. The measuring container filling height was taken into account during both efficiency calibration and actual sample measurements. Typical sample volume was 3 cm³, the detection limits for 100,000 s counting times were 1–7 Bq/kg ^{137}Cs and 15–30

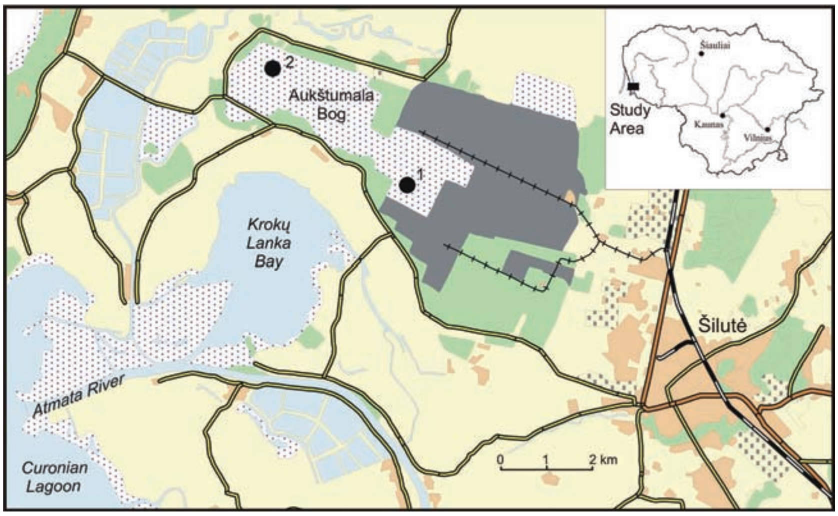

Fig. 1. Study area with coring locations.

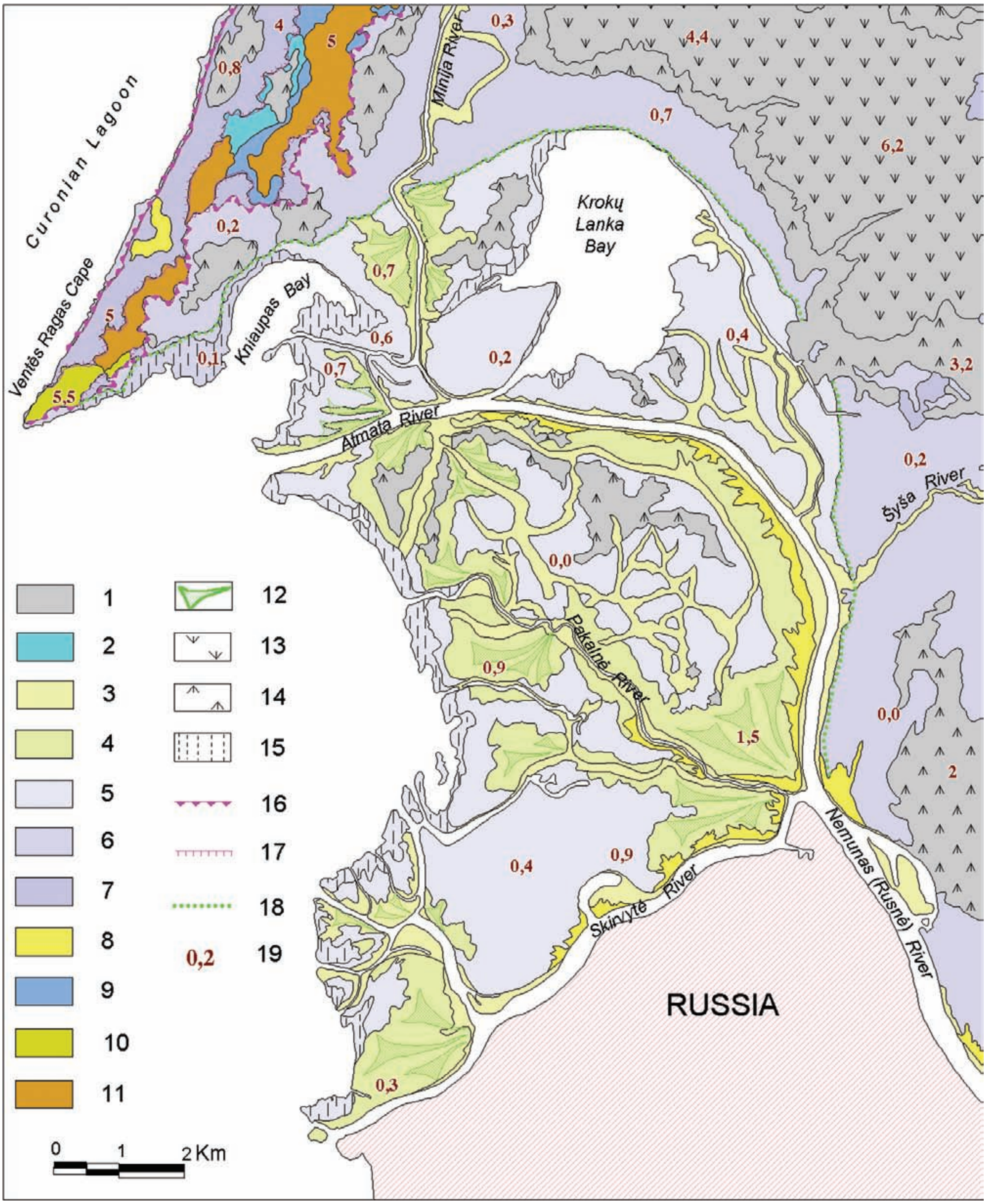

Fig. 2. Geological-geomorphologic structure of the Nemunas Delta (compiled by A. Damušytė and referenced from Bitinas et al. 2002): 1 – bog deposits; 2 – limnic sediments; 3 – alluvium of delta front; 4 – flood alluvium; 5 – lagoon sediments; 6 – marine sediments: Postlitorina Sea; 7 – marine sediments: Litorina Sea; 8 – aeolian deposits; 9 – glaciolacustrine sediments; 10 – glaciofluvial deposits; 11 – glacial deposits (till); 12 – alluvial fan; 13 – high moor; 14 – low moor; 15 – recent floating peat belt; 16 – shoreline of Postlitorina Sea; 17 – shoreline of Litorina Sea; 18 – limit of Nemunas Delta in Lithuania; 19 – altitude, m a.s.l.

Table 1. Results of gamma ray assay on sliced samples of the upper part of peat cores taken from the Aukštumala bog in 2003 (detection limits of 1-2 and 15-30 Bq/kg, while measurement errors ±10-15% and ±20-40%, for ^{137}Cs and ^{210}Pb, respectively; ^{226}Ra activities are very close to the detection limit and have only tentative meaning).

Borehole, No.	Latitude N; longitude E	Examined interval, cm	Activity concentrations of radionuclides [Bq/kg d.w.]		
			^{210}Pb	^{226}Ra	^{137}Cs
1	55°23’00” N 21°22’34” E	0-4	1970±390	34	280±30
		4-8	1540±300	28	150±20
		8-12	1420±350	10	97±15
		12-16	1350±400	10	60±9
		16-20	500±200	24	13±2
		20-24	200±80	14	8±2
		24-28	60±30	10	3±1
		28-32	20±15	10	2±1
		32-36	<dl	10	1±1
		36-40	<dl	10	1±1
		40-44	<dl	10	2±1
		44-48	<dl	10	5±1
2	55°24’18” N 21°19’43” E	0-4	980±190	24	120±10
		4-8	500±100	20	70±1
		8-12	230±60	14	13±2
		12-16	100±40	10	5±2
		16-20	90±40	16	4±1
		20-24	<dl	15	4±1
		24-28	<dl	15	6±1
		28-32	<dl	15	<dl
		32-36	<dl	15	17±3
		36-40	<dl	15	16±3

Bq/kg ^{210}Pb. Measurement errors mostly did not exceed 10% and 20%, for ^{137}Cs and ^{210}Pb, respectively.

The samples of deeper lying peat and gyttja from the same locations were taken for ^{14}C dating using a Russian corer with a tube 1 m in length and 5 cm in diameter. The 10–15 cm long sub-samples were used for ^{14}C dating by conventional liquid scintillation counting (Gupta and Polach 1985). Two methods of sampled bulk organics preparation were applied: (1) acid-alkali-acid based physical–chemical pre-treatment of sampled material; (2) synthesis of benzene – the final counting liquid for beta decay counting. A routine method for synthesis of benzene was applied (Arslanov 1985; Skripkin and Kovalyukh 1994; Petrošius, Mažeika 2004). The following operations were carried out for this purpose: (1) reaction of C with Li in the high temperature reactor at 550–600°C receiving Li_2C_2, (2) hydrolysis of Li_2C_2, receiving C_2H_2, (3) synthesis of benzene by cyclo-trimerisation of C_2H_2 using a catalyst containing aluminous-silicate and activated with V_2O_5, (4) purification of benzene from moisture remains by adding of metallic sodium and distillation, (5) adding of scintillation admixtures PPO and POPOP to the benzene sample. The beta decay counting was undertaken in the Radioisotope Research Laboratory at the Institute of Geology and Geography, Vilnius, using a LS Analyzer *Tri-Carb 3170 TR/SL* which provides ^{14}C background count rate of 0.41 cpm with Teflon-copper vials of 3 ml, efficiency for ^{14}C counting of 71.3% and upper ^{14}C age limit of 50 000 years using 48 hours counting.

Peat accumulation rates

The peat accumulation rate was determined through a combination of ^{210}Pb dating for younger samples and radiocarbon dating for older material. The ^{210}Pb chronology of the last two hundred years was determined through the activity of 10 and 12 depth intervals for respective cores according to constant rate of supply (CRS) model (Appleby, Oldfield 1978), with confidence intervals calculated by first-order error analysis of counting uncertainty (Binford 1990). Assumption of Pb immobility in peat column was taken into consideration. Dry mass accumulation rates were calculated according to Binford *et al.* 1993. Unsupported ^{210}Pb was calculated by subtracting supported ^{210}Pb (estimated from asymptotic activity at depth) from the total activity at each level. Rough values of supported ^{210}Pb activity were also derived from ^{226}Ra, which however in most of the samples was detected with very low activities approaching the detection limits for the used gamma spectrometric system. The radiocarbon dates were converted to calendar years using 2-D dispersion calibration procedure with computer software CALPAL_A (Jöris, Weninger 2000) and calibration curve CALPAL 2004 (http://www.calpal.de), which for Holocene time is basically derived from INTCAL98 (Stuiver *et al.* 1998).

RESULTS AND INTERPRETATION

The experimental results of activity concentrations of ^{210}Pb, ^{137}Cs and ^{226}Ra in sliced samples of peat cores taken from the upper part of the Aukštumala bog in 2003 are presented in Table 1 and in Figs 3 and 4.

^{210}Pb activity decreases down core in an exponential manner (R^2=0.85 for core #1, and R^2=0.97 for core #2), reaching supported values (10–15 Bq/kg) below 32 cm for core #1, and below 20 cm for core #2 (Figs. 3, 4). However, the core #1 showed some departures from a simple monotonic decline, possibly reflecting young episodic variations in the peat accumulation rates or perhaps manifestation of ^{210}Pb mobility. The core #1 is located in significantly damper conditions compared to core #2. The activity of ^{210}Pb in the upper 16 cm interval of core #1 ranges between 1970 and 1350 Bq/kg; while below at an interval of 16–28 cm, the activity of ^{210}Pb evenly decreases until it reaches a constant level of 10–15 Bq/kg. The activity of ^{210}Pb in the core #2 evenly decreases from 980 Bq/kg until it reaches a constant level of 10–15 Bq/kg.

^{137}Cs activity decreases down core in an exponential manner (R^2=0.96 for core #1, and R^2=0.97 for core #2) for the upper 18–20 cm interval. Below this interval, the ^{137}Cs activity rises again and reaches 5 or even 17 Bq/kg (Table 1). This may imply an elevated ^{137}Cs mobility in the bog environs that reduces the possibility of dating by ^{137}Cs (Frissel and Pennders 1983). Peat bog waters are naturally acid and organic-rich and may vary between oxic and anoxic, depending on the depth of the water table fluctuating seasonally. These circumstances

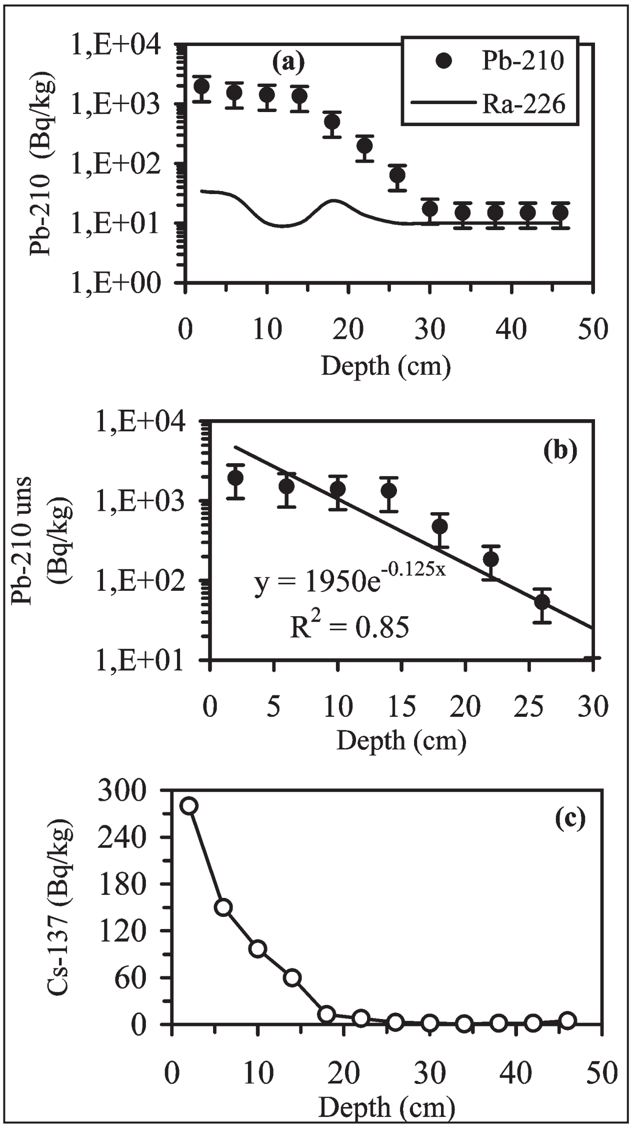

Fig. 3. Depth profile of total ^{210}Pb and ^{226}Ra (a), unsupported ^{210}Pb (b) and ^{137}Cs (c) for core from borehole Aukštumala 1.

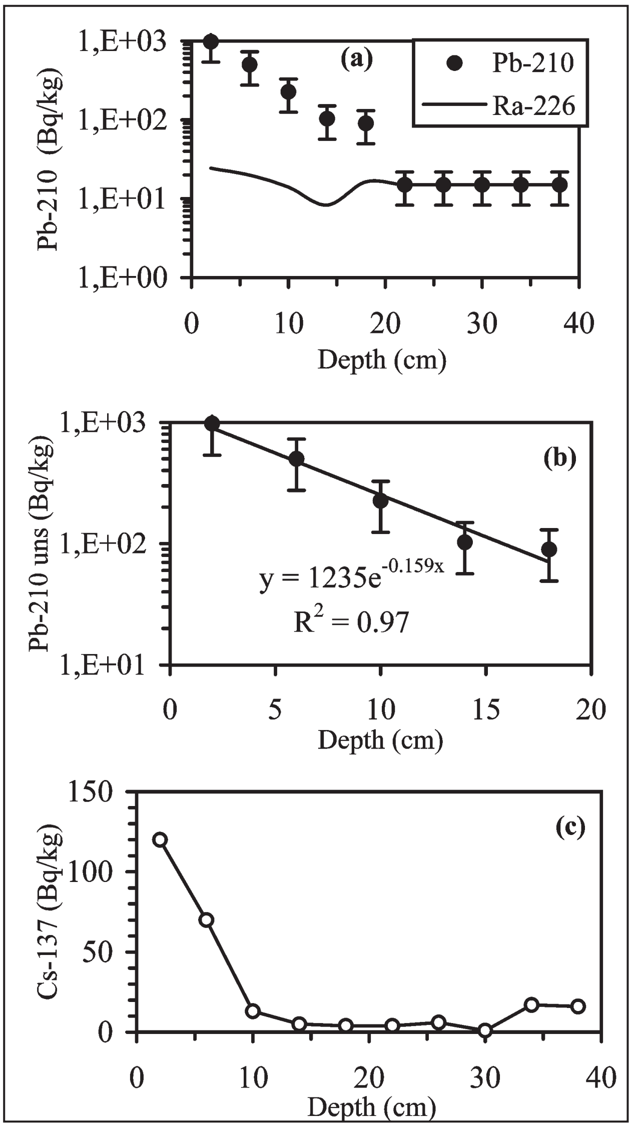

Fig. 4. Depth profile of total ^{210}Pb and ^{226}Ra (a), unsupported ^{210}Pb (b) and ^{137}Cs (c) for core from borehole Aukštumala 2.

Table 2. Results of radiocarbon (^{14}C) dating for peat cores taken from the Aukštumala bog in 2003.

Borehole, No.	Latitude N; longitude E	Uncalibrated ^{14}C age BP, ±1σ (Calendar age with 68% confidence limits, BP (0=AD1950)	Examined interval, cm	Dated material	Laboratory code
1	55°23'00" N 21°22'34" E	2760±190 (2890±260) 5720±160 (6530±160)	385-400 800-810	Peat Sandy gyttja	Vs-1458 Vs-1463
2	55°24'18" N 21°19'43" E	2280±200 (2330±270) 5560±140 (6350±150)	285-300 735-750	Peat Sandy gyttja	Vs-1455 Vs-1470

may give rise to a variety of chemical and biochemical transformations of metals, which could liberate them from the solid phase (peat) and facilitate their removal by diffusive or advective flow (Coleman 1985; Urban *et al.* 1990; Shotyk *et al.* 2000).

^{226}Ra activity, conditioning the activity of supported ^{210}Pb, was measured only approximately because gamma count rates of ^{226}Ra from the samples were within the range of background variations of used gamma spectrometric system. Yet orientationally, ^{226}Ra may be assumed to range within an interval of 10–30 Bq/kg, according to ^{214}Pb, the daughter product of ^{226}Ra (Table 1).

Net peat accumulation expressed in terms of mean dry mass accumulation rates and derived from constant rate of supply (CRS) calculations for the last years is relatively constant for both cores, varying between 0.006 and 0.011 g/cm^2/yr for core #1, and between 0.008 and 0.014 g/cm^2/yr for core #2.

Radiocarbon dates of older strata as conventional and converted to calendar ones are given in Table 2. The oldest dates representing the first organic material laying on the mineral bottom of bog, yielded calendar ages of 6530±160 years BP for core #1 and 6350±150 years BP for core #2. These dates fairly well correspond to the Litorina Sea stage of the Baltic Sea development and to the middle of the Atlantic chronozone (8000–5000 yr BP) using the Holocene time scale (Kabailienė 1998).

The age/depth relationships and mean linear peat accumulation rates derived from both ^{210}Pb and ^{14}C dating methods are shown in Fig. 5. The least-squares regression through the lead-210 and radiocarbon dates shows rather smooth transition between ^{210}Pb and ^{14}C chronology. The error bars representing 1 standard deviation propagated from ^{210}Pb counting uncertainty are not to scale in Fig. 5. However, the error terms become notably large for the sampling intervals below depth of 20 cm, reflecting the uncertainty of estimating small activity remaining in the older peat. Radiocarbon dates with standard deviations have been converted to calendar years and are shown with their 68% confidence limits.

Mean linear peat accumulation rates during the last 200 years in the two cores #1 and #2 were not significantly different (0.17 and 0.19 cm/yr, respectively). Between the oldest ^{210}Pb dates and youngest ^{14}C dates, peat accumulation rates in both cores are more similar: 0.13 and 0.11 cm/yr. For the oldest ^{14}C dates peat accumulation rates in both cores are exactly the same: 0.11 cm/yr. A general trend of the past somewhat lower peat accumulation rates, gradually increasing to higher recent accumulation rates of the ^{210}Pb chronology, is noticeable. This apparent

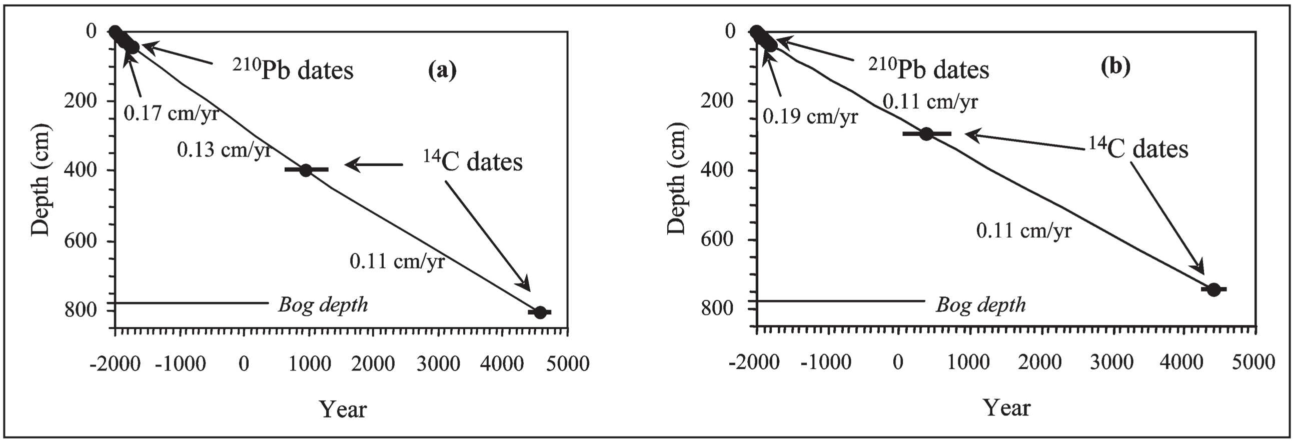

Fig. 5. Peat age versus depth for cores Aukštumala 1 (a) and Aukštumala 2 (b). Radiocarbon dates with standard deviations have been converted to calendar years and are shown with their 68% confidence limits. Numbers indicate peat accumulation rate.

increase in peat accumulation probably is an artefact of decaying organic matter or an indication of ^{210}Pb mobility in peat. As time goes on, the organic matter representing a span of time should become more decayed and compacted, giving the appearance of a lower rate of accumulation in the older strata (Cole *et al.* 1990).

CONCLUSIONS

The possibilities of evaluation of peat accumulation parameters by radioisotope methods are shown on the example of Aukštumala raised bog located in western Lithuania. Positive results have been obtained by combining ^{210}Pb and ^{14}C dating methods. The estimated radioisotope chronology and calculated peat accumulation rates are consistent with the geological history of the Aukštumala bog environs and the whole Lithuanian maritime region.

Mean linear peat accumulation rates during the last two hundred years for the two cores from Aukštumala bog were as following: core #1 – 0.17 cm/yr, core #2 – 0.19 cm/yr. Recent mean peat accumulation rates during the last 200 years were similar to long-term linear peat accumulation rates (0.11–0.13 cm/yr) that indicate the relatively undisturbed condition of Aukštumala bog in the studied sites.

The values obtained in this study can be used to estimate timing of environmental changes in Aukštumala bog during the last 6000 years.

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Mr. G. Berlinskas from Environment Protection Agency of the Ministry of Environment, Lithuania, provided expert gamma ray assay of upper samples of peat cores for ^{210}Pb, ^{137}Cs and ^{226}Ra activity estimations. Financial support provided by the Lithuanian State Science and Studies Foundation (contract No. C-19/2003) is gratefully acknowledged. The author thanks Dr. R. Petrošius and Mr. G. Davainis for the assistance in the laboratory and the team of Department of Climate and Water Systems at the Institute of Geology and Geography, Vilnius, for the assistance in field trips. The reviewers Dr. Helmar Kunzendorf, Denmark, and anonymous are thanked for useful critical comments that resulted in a significantly improved manuscript.

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