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THE USE OF CARBON-14 AND TRITIUM FOR PEAT AND WATER DYNAMICS CHARACTERIZATION: CASE OF ČEPKELIAI PEATLAND, SOUTHEASTERN LITHUANIA

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Abstract: The present investigation conducted in Čepkeliai peatland (south-eastern Lithuania) deals with two radioisotope methods – carbon-14 (^{14}C) and tritium (^{3}H). ^{14}C was applied to peatland chronology and sedimentation rate estimation and ^{3}H to peat water dynamics characterization. According to ^{14}C data, peatland development began since 12650-11350 years cal BP and peat accumulation since 10550-9700 years cal BP with peat accumulation rate of 0.27-0.79 mm/year in the central part of the peatland. The peat water mean residence time and basic groundwater flow was estimated to be respectively about 27±6 years and 62±15 mm based on ^{3}H data. The obtained data showed relatively undisturbed natural condition of Čepkeliai peatland in the studied sites.

Keywords: Carbon-14, Tritium, Peat, Groundwater Residence Time, Holocene.

1. INTRODUCTION

Many environmental effects, including the Quaternary palaeoclimate variability, ecological changes and other changes can be studied using environmental tracers, like tritium (^{3}H) and radiocarbon (^{14}C). The peatlands are among the distinctive ecosystems containing archives of short-term and long-term environmental effects. The short-term effects can be related to peatland water system itself or bog hydrology. From geophysical point of view, bog water cycle is attributed to decades scale processes that could be characterised among others by use of short-lived radioisotope ^{3}H which was injected into the stratosphere and consequently occurred in precipitation following the nuclear weapon tests of the early sixties of the 20th century (Ekwurzel *et al.*, 1994). Well known non-monotonic ^{3}H temporal distribution in meteoric water of the last 50 years gives a possibility to evaluate groundwater residence time in the bog system, its recharge-discharge rate and water budget. The long-term environmental effects can be studied using the dated peat cores from bogs (Cole *et al.*, 1990; Shotyk *et al.*, 2000; Kim and Rejmánková, 2002). The use of the long-lived radioisotope ^{14}C as a dating tool gives time scale for assembling information on physical, chemical and biotic properties of peatlands within the context of a changing environment and as a minimum the possibility to estimate peat (and lacustrine) sedimentation rate.

The aim of the present investigation was to directly focus on radioisotope methods (^{3}H and ^{14}C) for geophysical characterization of peat and water dynamics. This study was conducted in Čepkeliai peatland (south-eastern Lithuania) which is a raised Sphagnum bog. In terms of landform morphology, hydrology, water chemistry and vegetation that determine a distinctive ecosystem with characteristic plant, invertebrate and bird communities, the Čepkeliai peatland is considered to be one of the most important territories in the Dzūkija National Park. The study included the measurements of ^{14}C in peat and gyt-

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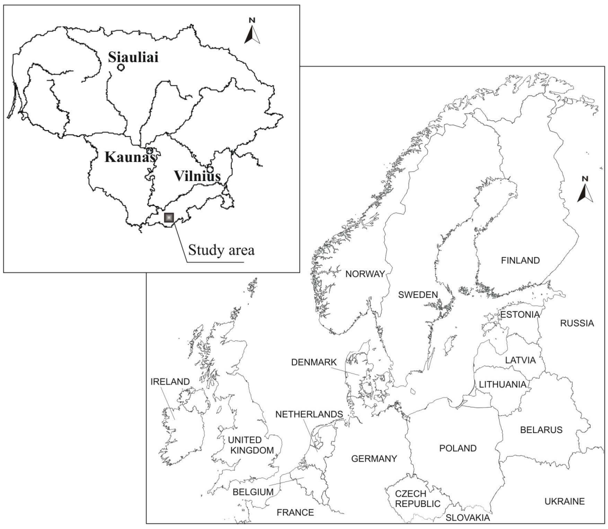

Fig. 1. *Location of studied area.*

tja, ^{3}H in peat water and in the water taken from piezometers in the period 2004-2007. The ^{3}H data were interpreted on the basis of lumped-parameter models.

2. ENVIRONMENTAL AND GEOMORPHOLOGICAL SETTINGS

Environment

The study area comprises Čepkeliai peatland (54°01'N, 24°32'E) which is located in the south-eastern part of the country, close to the Lithuanian-Belarusian border (**Fig. 1**). The peatland (total area – 5858 ha, volume of the peat – 125.6 million m³) is situated in the watershed of the Katra, Ūla and Grūda streams. The biggest part of the peatland (71%) is represented by raised bogs (Klimavičienė, 1974; Grigelytė, 1977; Tamošaitis and Grigelytė, 1977). The surface is located 134 m above the sea level in the central site of raised bogs, and 130-127 m in the margin fens.

Čepkeliai peatland is the largest one in Lithuania and one of the largest in the Baltic region. Large open raised bogs, fens, numerous small lakes prevailing in the eastern part, pools, forested islands and permanently flooded old forests cover large areas of this peatland. The Čepkeliai State Nature Reserve was established in this site in 1975, since 1993 it is Ramsar site. The total area of Reserve is 8477 hectares, of which 2725 hectares are covered by a forest and the remaining part is peatland. The aim of Čepkeliai State Nature Reserve is preserving one of the oldest and most unusual raised forest bogs in Lithuania, forested continental dunes, lakes, natural hydrological regime of the raised bog and the rare plant and animal life. The strict reserve has an enormous variety of habitats, and species of rare plants and animals abound. For example, it provides nesting grounds for the capercaillie (Tetrao urogallus).

The annual precipitation in the study area ranges from 423 (1971) to 861 (2005) mm (annual average 700 mm, 1925-2006; data of the Lithuanian Hydrometeorological Service). About two thirds of the annual precipitation falls during the vegetation season. A decrease of vegetation season precipitation and increase of cold season precipitation have been observed in the last decade. The average annual temperature is +6.2°C; the coldest month is January with average temperature of -5.2°C and the warmest month is July with +17.4°C. Hydrological conditions of the surroundings of the peatland significantly changed in the second half of 19th century, when due to the bed erosion the Ūla stream captured the upper reaches of the Katra stream. High groundwater level in the peatland stipulates the intensive evotranspiration. About 78% of precipitation evaporates in May – September. Before and after this season evotranspiration decreases by 2-3 times, and in October it does not exceed 6.7-21.8 mm (Dilys and Gikytė, 1977). The annual average evotranspiration makes approximately 500 mm (Dilys and Gikytė, 1977). Consequently, the annual average run-off from the catchment area including groundwater basic flow is 200 mm. The average spring flood run-off is 60 mm (Dilys and Gikytė, 1977).

Geomorphology

The Čepkeliai peatland is situated in the glaciolacustrine plain occupying the lowest part of the outwash plain which distal part marks the very margin of the Last Glaciation (**Fig. 2**). Further to the south-east flat or slightly undulated morainic plateau is 20-30 m higher than the glaciolacustrine plain. To the north-west glaciolacustrine plain is surrounded by the well expressed continental dunes. The glaciolacustrine plain of the Last Glaciation lies at about 127-132 m a.s.l. and is nearly everywhere composed of fine-grained sand. The lowest marshy parts of this plain were occupied by former lakes throughout late glacial and Holocene time (Stančikaitė *et al.*, 2002) which later turned into bogs and marshy areas. The Čepkeliai peatland is the largest among them.

Investigated sites

Two cores from different locations of Čepkeliai peatland (location of Core No 1 – 54°00’55”N, 24°26’08”E, location of Core No 2 – 54°01’21”N, 24°3’29”E) were taken using a peat corer with a tube 1 m in length and 5 cm in diameter in July 2003. The distance between the two sites is about eight km (**Fig. 1**).

The 10-15 cm long samples (4 samples from each core) were taken for ^{14}C dating. The 5-10 cm long samples (13 samples from Core No 1, 12 samples from Core No 2) were taken for the measurements of ^{3}H volumetric activity in peat water.

The 5 cm thick slices (11 samples from Core No 1) were taken in order to determine the depth distribution of main components (biogenic, mineral – ash, carbonates) of peat material.

In 2004, five piezometers (0.8, 1.0, 2.0, 3.0 and 3.94 m depth) for groundwater level observation and ^{3}H sampling were installed very close to the Site No 1 in the north-western part of the peatland (54°01’11”N, 24°26’14”E). Four piezometers screened the peat body and one piezometer (the deepest) screened confining from below sandy layer. The groundwater level observations were continued every ten days in the vegetation periods of 2004-2006 (20th May 2004 – 10th December 2004;

10th April 2005 - 30th October 2005; 10th April 2006 - 20th April 2006). Groundwater samples for ^3H measurements from the piezometers have been taken several times per year (35 samples in total).

3. METHODS OF INVESTIGATIONS

^{14}C dating

The eight bulk samples including peat and gyttja for ^{14}C dating were more or less evenly spaced along the two sediment sequences. Peat and gyttja samples were dried in an oven (at 45°C). After drying, acid-alkali-acid based physicochemical pre-treatment of organic material was implemented for peat samples. The carbonates from gyttja samples were transferred to CO_2 dissolving them with hydrochloric acid without any chemical pre-treatment. The specific activity of ^{14}C in peat and gyttja samples was measured as described in (Gupta and Polach, 1985; Arslanov, 1985; Bowman, 1995). A conventional method for synthesis of benzene was applied (Kovaliukh and

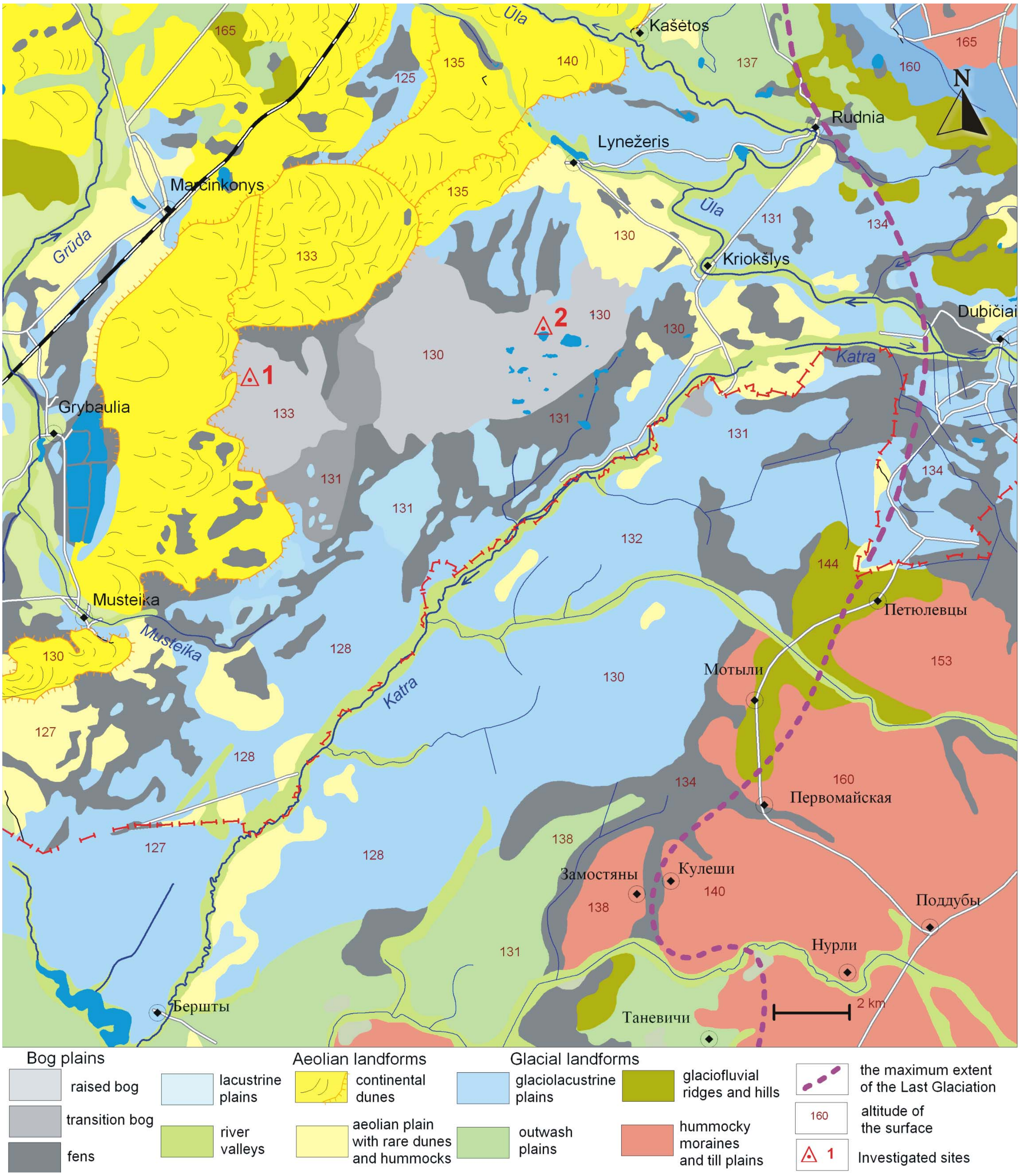

Fig. 2. *Geomorphological map of the Čepkeliai peatland area (Guobytė* et al., *2005).*

Skripkin, 1994). The ^{14}C specific activity of benzene was determined by liquid scintillation spectrometry. The main performance parameters of liquid scintillation analyzer (Tri-Carb 3170 TR/SL) for ^{14}C in benzene put into 3 ml Teflon vials were as follows: background count rate of 0.41±0.04 CPM; counting efficiency of 71.3±0.8% ; and Figure of Merit (Efficiency2/Background) of 12400±700.

The primary ^{14}C results were expressed as specific activity (1 pMC = 2.27 Bq kg^{-1} C) (Stuiver and Polach, 1977; Stuiver, 1983) and as uncalibrated ^{14}C age (years BP±1 σ). The uncertainty of results was reported at 1-sigma level. The radiocarbon dates were converted to calendar years and sediment accumulation rate was estimated using the radiocarbon calibration program OxCal v3.10 (Bronk Ramsey, 2001) with the IntCal04 data set (Reimer *et al.*, 2004).

The quality of ^{14}C determinations was periodically tested through participation in various intercomparisons. The results of FIRI, organized by the University of Glasgow in 2000, indicated that most of the ^{14}C results of Radioisotope Research Laboratory at Institute of Geology and Geography, Vilnius, Lithuania are of normal (3%), while a part is of high (1.5%) precision.

Component-based analysis

Percentage of main components of sedimentary matrix was estimated as described in (Bengtsson and Enell, 1986). First, the samples were slowly dried at 50-80°C and later at 105°C until a constant weight was reached. Carbonate content was estimated by titration with HCl, organic matter content was estimated using loss of weight on ignition at 550°C, and mineral (ash) component of sediment matrix was calculated. The water content and dry bulk density of sediments were estimated by weighting standard volume samples dried at 105°C.

^{3}H measurements

The ^{3}H specific activity in water evaporated from peat by the laboratory distiller and in the water taken from piezometers was measured by the same liquid scintillation analyzer using scintillation cocktail with 12 ml of OptiPhase "TriSafe" and 8 ml of original water, mainly without electrolytic enrichment, as described in (IAEA, 1981). Only the last set of samples taken on 25th June 2007 underwent electrolytic enrichment. The main performance parameters of analyzer for ^{3}H with 20 ml plastic vials were as follows: background count rate of 1.01±0.08 CPM; counting efficiency of 22.0±0.4%; and Figure of Merit of 480±20 (Efficiency2/Background). The ^{3}H results were presented in Tritium Units (TU), the latter by definition is equivalent to a concentration of $^{3}H/^{1}H = 10^{-18}$ or to specific activity of ^{3}H in water 0.118 Bq l^{-1}. The uncertainty of results was reported at 1 sigma-level.

The normal precision of ^{3}H measurements in water was recognized in the comparison, organized by the Latvian Environment Agency Laboratory (Jurmala), National Veterinarian Laboratory of Lithuania and Radioisotope Research Laboratory of Institute of Geology and Geography in 2004, in the Seventh IAEA intercomparison of low-level tritium measurements in water (TRIC2004) and in the NKS LABINCO Intercomparison Exercise 2004-2005.

Lumped-parameters (box) models for determining the mean residence time of groundwater

Maloszewski and Zuber (1982) were the first to apply lumped-parameters models to groundwater systems in order to evaluate the age or mean residence time of groundwater. Environmental tracers such as ^{3}H enable the use of models for aquifers with young (i.e. <50 years) water. Detailed information on the use of lumped-parameter models in the calculation of mean residence time of groundwater can be found in Maloszewski (1996). The lumped-parameter models are represented by several computer codes, namely FLOWPC (Maloszewski, 1996), BOXMODEL (Zoellmann *et al.*, 2001) and TRACER (Bayari, 2002). In this study, BOXMODEL for ^{3}H data evaluation developed by Zoellmann and Aeschbach-Hertig was used. The tritium input function (variation of activity in precipitation water of studied area) based on tritium measurements in precipitation in the East Lithuania since 1999 and their correlation with the data of Global Network of Isotopes in Precipitation (GNIP Vienna Hohe Warte station) was compiled. The comparison of both data sets for overlapping period is shown in Fig. 3 and the excerpt of the input function is shown in Fig. 4.

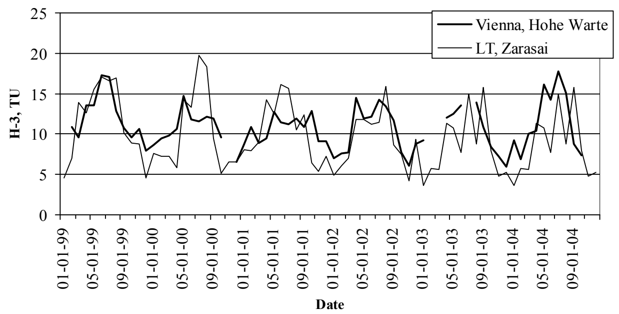

Fig. 3. *^{3}H variation in atmospheric precipitation of East Lithuania (monthly samples of period 1999-2004) compared to that in Vienna (GNIP data, WEB site: http://isohis.iaea.org, accessed 2009 January 15). Correlation coefficient between two data sets is 0.59 (number of observations is 66).*

Linear regression equation $y_{LT} = 0.88x_{Vienna} + 0.36$.

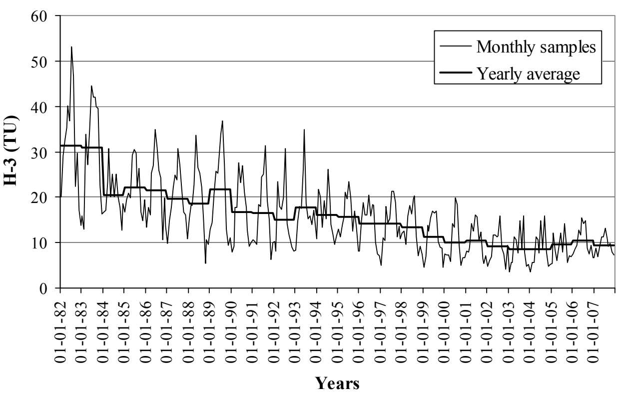

Fig. 4. *^{3}H variation in atmospheric precipitation in East Lithuania (Zarasai station). Continuous measurement data are available since January 1999.*

For a given ^{3}H input function convoluted theoretical output depending on transfer function and on its parameters was calculated using BOXMODEL. The transfer functions are known for most common piston flow, exponential and dispersion models (PM, EM, DM). The parameters of the transfer function can be: mean residence time τ and dispersion parameter δ. By comparison of the observed and calculated ^{3}H concentration, these parameters can be identified if the considered groundwater system is linear and steady state in its flow properties. The choice of flow system model depends on general aquifer situation which governs scale of groundwater flow pathlines mixing.

4. RESULTS AND DISCUSSION

Radiocarbon dates of the peat and gyttja samples as conventional and converted to calendar ones are given in **Table 1**.

The Core No 1 contains peat all the way down to the mineral basement of the peatland. The calibrated ^{14}C age of the peat ranges from 750-560 years cal BP within the depth interval 0.85-1 m to 10550-9700 years cal BP at the depth interval (peat and sand interface) 4.35-4.50 m. Component-based data of Core No 1 (**Table 2**) revealed that peat contains carbonate material admixture (up to 6.8%) only in top (0-40 cm) and bottom (435-445 cm) slices of the core. The lower layer consists of fen peat where remains of mollusc shells can be found. Comparatively high content of carbonates in the upper layer probably is due to aerosols with high amount of $CaCO_3$. In the environment of Čepkeliai peatland lies a large area of drained fens which is used for agriculture and gyttja is observed to be ploughed in the upper layer of fens. Mineral (ash) fraction of the peat varies between <0.1 and 3.4% with maximal value at a 40-50 cm depth interval. The carbonate phase can be sensitive to climatic and drainage basin conditions. The mineral (ash) fraction can be related to aeolic processes in drainage basin and forest fires. Core No 2 is presented by binary sequence including peat layer of 4.9 m thickness overlying a 1.1 m thick gyttja layer. The calibrated ^{14}C age comprises time span between 1570-1340 years BP for 0.85-1 m depth (peat) and 12650-11350 years BP for depth interval of 5.85-6.00 m (gyttja) very close to bog mineral base. The transition from gyttja to peat accumulation occurred in Late Boreal time (8900-7800 cal years BP).

Table 2. *Components of Core No 1 taken from Čepkeliai peatland (1σ uncertainties for $CaCO_3$ component – ±1.9%, for biogenic component – ±1.0, for mineral component – ±0.4).*

No	Depth of sampled interval (cm)	$CaCO_3$ (%)	Mineral (ash) (%)	Biogenic (%)
1	0-10	6.1	<0.1	93.8
2	50-40	6.8	3.4	89.8
3	80-90	<0.1	1.4	98.6
4	130-135	<0.1	1.7	98.3
5	185-180	<0.1	1.8	98.2
6	235-230	<0.1	1.1	98.9
7	285-280	<0.1	1.7	98.3
8	335-330	<0.1	1.1	98.9
9	385-380	<0.1	1.9	98.1
10	435-430	3.9	<0.1	96.0
11	450-445	4.2	1.1	94.7

The oldest dates representing the first organic or carbonate material laying on the mineral bottom of peatland, yielded calendar ages 10550-9700 years BP for Core No 1 and 12650-11350 years BP for Core No 2. These dates fairly well correspond to the Preboreal and Younger Dryas chronozones of Holocene time scale (Kabailienė, 1998).

The ^{14}C age / sediment depth relationships are shown in **Fig. 5**.

Based on the calibrated (1σ) ^{14}C age minimum and maximum values and on the depth of sampled interval midpoint the sediment accumulation rates were calculated. The range of sediment accumulation rates in the area of Core No 1 were as follows: between depths

Table 1. *Results of radiocarbon (^{14}C) dating for cores taken from Čepkeliai peatland (location of Core No 1 – 54°00'55"N, 24°26'08"E; location of Core No 2 – 54°01'21"N, 24°33'29"E).*

No.	Lab. index	Sample description	Material analysed	^{14}C activity (pMC±1σ)	^{14}C age (BP±1σ)	Calibrated ^{14}C age with 68.2% confidence level (cal BP)
1	Vs-1461	Core No 1, peat, sampled interval 0.85-1.00 m	Bulk organics	91.3±1.6	730±90	750-560
2	Vs-1454	Core No 1, peat, sampled interval 1.85-2.00 m	Bulk organics	91.3±1.6	1750±100	1820-1550
3	Vs-1462	Core No 1, peat, sampled interval 3.35-3.50 m	Bulk organics	80.4±1.7	6110±130	7170-6800
4	Vs-1453	Core No 1, peat, sampled interval 4.35-4.50 m	Bulk organics	46.41±0.99	9020±300	10550-9700
5	Vs-1465	Core No 2, peat, sampled interval 0.85-1.00 m	Bulk organics	32.5±1.6	1570±110	1570-1340
6	Vs-1466	Core No 2, peat, sampled interval 2.85-3.00 m	Bulk organics	82.3±1.5	3800±90	4390-4000
7	Vs-1467	Core No 2, peat, sampled interval 4.50-4.75 m	Bulk organics	62.29±0.96	6930±90	7850-7670
8	Vs-1480	Core No 2, gyttja, sampled interval 5.85-6.00 m	Carbonates	42.22±0.59	10260±450	12650-11350

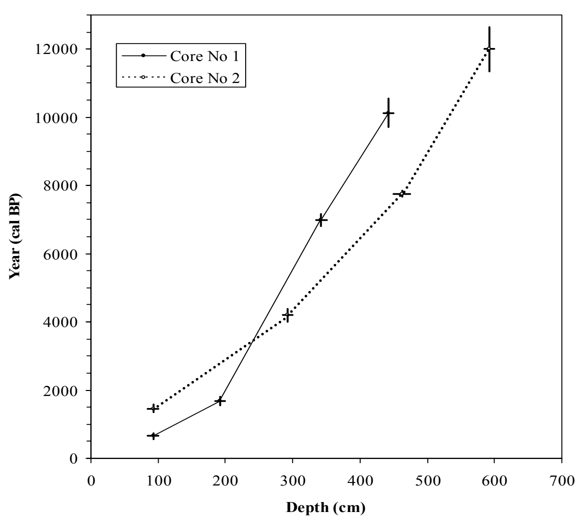

Fig. 5. *Peat age versus depth for Čepkeliai peatland cores (Core No 1 and Core No 2). Radiocarbon dates with standard deviations have been converted to calendar years using OxCal v3.10 (Bronk Ramsey, 2001). Vertical bars represent calibrated age ranges, horizontal ones – thickness of samples. Based on linear interpolation of age – depth distribution the transition from gyttja to peat accumulation in area of Core No 2 occurred in Late Boreal time (~8600 cal BP).*

93 and 193 cm – 0.91-1.25 mm/year, between depths 193 and 343 cm – 0.27-0.29 mm/year, and between 343 and 443 cm – 0.29-0.38 mm/year. The range of sediment accumulation rates in the area of Core No 2 were as follows: between depths 93 and 293 cm – 0.70-0.79 mm/year, between depths 293 and 463 cm – 0.45-0.50 mm/year, and between 463 and 593 cm – 0.27-0.37 mm/year. Thus, based on the dates of the lower part of the sequence (2-6 m) in both sites sediment accumulation rate varies between 0.27-0.38 mm/year. Compared to the lower part of the core almost threefold higher sediment accumulation rate is peculiar to the top part of sequence of Core No 1 (0.91-1.25 mm/year) located in the marginal part of peatland. Similarly, compared to the lower part of the core two times higher sediment accumulation rate is estimated for the top part of the sequence of Core No 2 (0.70-0.79 mm/year) located in almost central part of the peatland.

^{3}H activity in the water evaporated from peat samples showed pulse-shaped distribution of ^{3}H with depth (**Fig. 6**).

Based on ^{3}H data in the site of Core No 1, more active groundwater circulation in vertical profile was observed compared to site of Core No 2. In spite that the original ^{3}H activity in peat water of certain depth interval could be diluted by water from the top during coring, the noted fact stipulated an installation of piezometers in order to obtain ^{3}H in groundwater time series and to observe the groundwater level in the different depth piezometers. Groundwater level observation in the piezometers group illustrated downward hydraulic head gradient and recharge conditions (**Fig. 7**).

Water level variations in the piezometers at the end of August 2005 are related to heavy rains of 8-10 August 2005 what had influenced flood in the studied region.

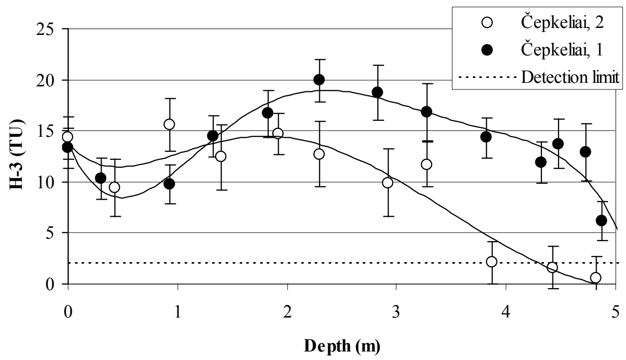

Fig. 6. *^{3}H activity in water evaporated from peat samples versus depth. Smoothed bomb-pulse distribution is observed and however can be disturbed due to dilution by recent water during coring. ^{3}H detection limit for samples without enrichment is about 2 TU. Polynomial trendlines of order 5 fitted to the data points (R-squared value for Core No 1 – 0.91, for Core No 2 – 0.88).*

^{3}H in the groundwater time series comprise period of 2004-2007 and are based on 7 sampling campains. For ^{3}H data interpretation BOXMODEL multiple runs were done. After computational exercises, it was derived that an agreement of the piston flow model (PM) with measured samples from deepest piezometers occurs; meanwhile no agreement of such model with measured samples from shallow piezometers is observed. On the other hand, the exponential model (EM) was satisfactory with the data of shallow piezometers and was inappropraite for the data of deeper piezometers. Dispersion model (DM) yielded the best correlation between all the experimental ^{3}H data and theoretical output (**Fig. 8**).

Basically, ^{3}H activity concentration in water from two top piezometers is constant in the course of time what is an indication of water short residence time. It is evident from the ^{3}H input function (see **Fig. 4**) that yearly averaged ^{3}H activity concentration in atmospheric precipitation in past seven years is about 10 TU. However ^{3}H activity concentration in water from lower piezometers shows a decreasing trend despite large analytical uncertainties. The decreasing trend of ^{3}H

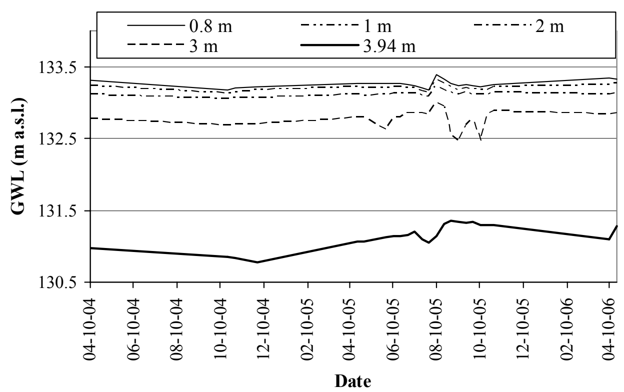

Fig. 7. *Altitude of groundwater level (m above sea level) in piezometers of Čepkeliai peatland. Surface altitude is 133.28 m a.s.l. At the piezometers site the minimum water level is at the depth of 11 cm below surface and the maximum water level is at the high of 11 cm above surface.*

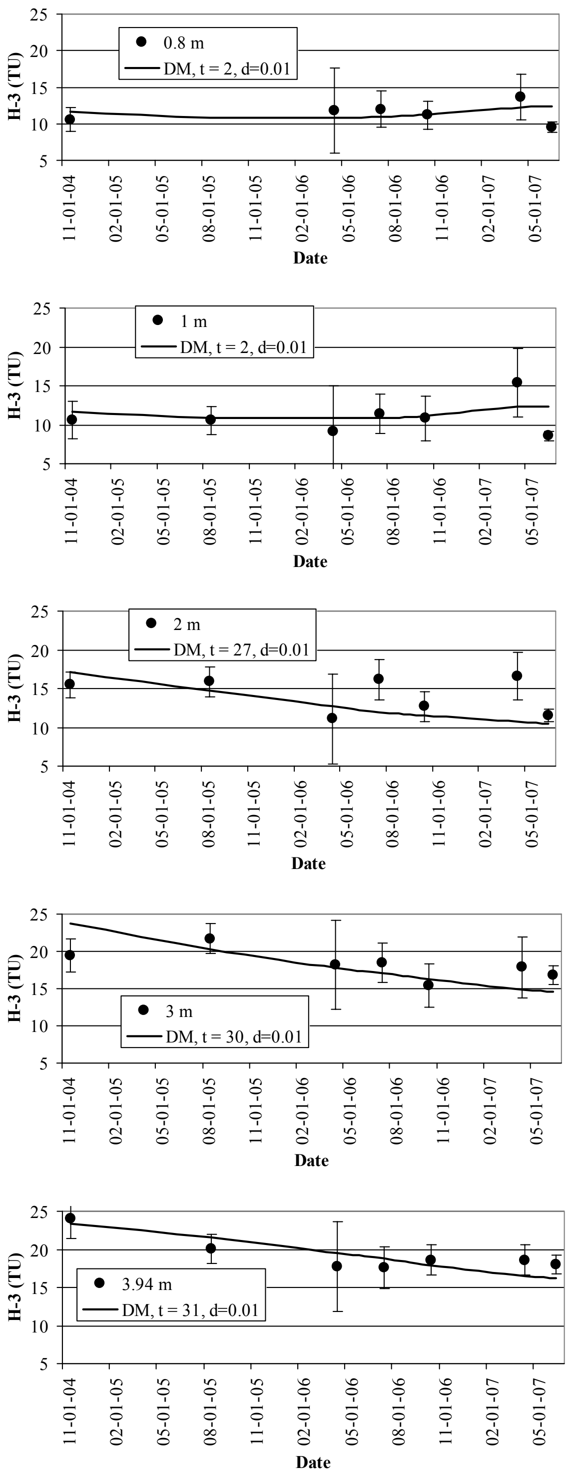

Fig. 8. *Output activity of ^{3}H as a function of time for different dispersion models. t – mean residence time, d – dispersion parameter. Time series of ^{3}H measurement with 1σ uncertainty are given for different depths of piezometer.*

activity concentration in water is an indication of water long residence time.

If the investigated groundwater system is best described with a dispersion model (DM with δ = 0.01), a realistic mean residence time would be as follows: about 2±2 years (or even less) for 0.8 m and 1 m depth, 27±6 years for 2 m depth, 30±6 years for 3 m depth and 31±6 years for 3.94 m depth.

Based on groundwater mean residence time robust water budget estimates can be substantiated. If we assume dominant thickness of the peat layer to be about 2.3 m, the groundwater residence time for this depth 27±6 years, volume porosity of peat 0.78, difference between precipitation and evotranspiration (700±20)–(500±10)=200±22 mm, the annual steady state groundwater recharge and discharge rates or basic groundwater flow can be estimated as 62±15 mm. Consequently the annual average run-off can be found as (200±22)-(62±15)=138±27 mm what is consistent with conventional hydrologic pattern.

5. CONCLUSION

The estimated ^{14}C chronology of bog development and calculated peat accumulation rate are consistent with the geological history of the Čepkeliai peatland environs. The little varying long-term linear peat accumulation rate (0.27-0.79 mm/year) in central part of the peatland indicates a relatively undisturbed natural condition of Čepkeliai peatland in the studied sites. The ^{14}C age data obtained in this study can be used to estimate the timing of environmental changes in Čepkeliai peatland during the last 12000 years.

The observed ^{3}H in groundwater time series enabled evaluating groundwater residence time and, based on it, specifying peatland water main budget elements. For dominant thickness of peat layer groundwater residence time is about 27 years what shows natural resistance of whole peatland hydrological system to very short time climatic variations. However, the top layer of peatland (~1 m) with active water circulation is sensitive to short time climatic variations.

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