Polycyclic Aromatic Hydrocarbons and Carbon Isotopes in a Palsa Peat (Bol’shezemel’skaya Tundra)

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Abstract—The content of polycyclic aromatic hydrocarbons (PAHs) and carbon isotope composition in the peat of a palsa near Eletsky settlement, Vorkuta urban district, Komi Republic are analyzed. The carbon isotope composition of peat varies from –28.05 to –30.05‰ (average –29.15‰). The total PAH content varies from 11 to 360 ppb, with an average of 63 ppb and a median value of 34 ppb. Heavy compounds, such as benzo(a)anthracene and benzofluoranthenes, are prevalent among PAHs. The presence of PAHs in the peat is determined by three main factors: technogenic impact, wildfires, and biogeochemical soil processes. The prevalence of benzo(a)anthracene in the upper part of the palsa down to the bottom of the active layer suggests an anthropogenic impact (the influence of transport and domestic fuel combustion). The share of benzo(a)anthracene decreases with depth, while the share of benzofluoranthenes increases. Two sharp peaks of PAH content (260 and 360 ppb) are observed; they coincide with a local increase of carbon isotope values, most likely resulting from wildfires. The minimums in PAH content are presumably determined by the biogeochemical factor and the input of polyarenes generated by decomposition of plant residues. Carbon isotope composition of peat mainly reflects the isotope composition of vegetation, the degree of peat moistening, and the influence of pyrogenic factor.

Keywords: polycyclic aromatics, paleoenvironment, wildfires, stable carbon isotopes, Dystric Histosols

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INTRODUCTION

The permafrost peat mounds are commonly referred to as palsa, a Lappish word denoting a peat or a peat—mineral convex mound formed by peat or typically fine mineral sediments covered with peat and having a frozen core. In Russian studies, these mounds are also referred to as “migrational” mounds since their ice core formed of schlieren and thin lenses of segregated ice develops during soil moisture migration from an adjacent bog or the atmospheric precipitation filtering to the freezing front, i.e., to the formed core. The peat of palsa can be regarded as an archive of the data reflecting the natural conditions at the time they were formed and the corresponding technogenic impacts. The carbon isotope composition of peat as well as polycyclic aromatic hydrocarbons (PAHs) and other biomarkers of peat layer are the particular indicators of these conditions [26, 30].

The products of biomass combustion [27] and the plant residues transformed by soil processes are the likely sources of PAHs in the palsa peat. For example, natural paleofires were identified according to the presence of heavy PAHs (pyrene, fluoranthene, benzofluoranthenes, and some other compounds) in buried soils [36]. The effect of pyrogenic factor is frequently assessed using diagnostic PAH ratios (for example, see [17]) based on different energy expenditures during formation of individual compounds.

A natural biogeochemical origin of PAHs is also possible. Although our knowledge about this phenomenon is rather vague, polyarenes are abundant in the landscapes unaffected by anthropogenic activities [8, 9, 12, 13, 16]. As has been shown [11], light polyarenes (3-ring phenanthrene and its alkyl derivatives) are prevalent in the PAH composition of peat bodies under natural conditions. The authors [11] proposed an indicator of the natural biotransformation potential of soils towards PAHs as the ratio of phenanthrene to its derivatives.

The carbon isotope composition of peat is in many respects inherited from vegetation [14, 29] and depends on the temperature during the vegetation season as well as on the water abundance in bogs and on the specific features in the decomposition of plant residues. The isotope signal of the peat-forming plants mainly varies from –20‰ in oligotrophic mosses to –29‰ in plants of sedge family (Cyperaceae Juss.) [21]. The leaves of Cyperaceae Juss. plants are enriched in 13C as...
ERS. Thus, the 12C and 13C distributions in Histosols down of the microbiological processes in deeper layers or remain constant because of the slowing down of the microbiological processes in deeper layers. Thus, the 12C and 13C distributions in Histosols and peat deposits reflect the total effect of stable isotope fractionation processes during photosynthesis and microbiological decomposition. Commonly, uniform distribution or slow increase in δ13C values with the depth indicate young soils [19, 22–24, 28].

The goal of this work was to find specific features in the distributions of 13C isotopes and PAHs in the palsa peat and to clarify the genesis of these phenomena. The following tasks were set: (1) to analyze the specific features in the contents of PAHs and carbon isotopes in palsa profile and (2) to find the sources of PAHs entering the palsa peat and to clarify the dynamics of carbon isotope composition.

OBJECTS AND METHODS

Location and natural conditions of the study area. A peat palsa located in the northeastern part of the Bol’shezemel’skaya tundra, within a palsa mire near Eletsky settlement (Fig. S1) at a distance of 1.5 km to the east of the settlement and 500 m from the railroad was examined in detail; its height is 3.5 m; diameter is 10 m; and slope gradients are up to 15 degrees (Fig. S2). The palsa is covered with moss—grass vegetation with dwarf shrubs (sphagnum, common haircap moss, sedges, and dwarf birch). According to radiocarbon dating, the age of the palsa is over 6400 years [3, 4].

A large part of the territory is hilly with low flat-topped mounds, hill chains, and shallow hollows separating them; the hollows have gentle slopes and wide bottoms, which are typically occupied by lakes, bogs, and peatlands with palsas.

The climate is subarctic moderately continental; summer is short and cold; and winter is abundant in snow, long, and severe. The climate is formed under conditions of low solar radiation in winter and the influence of northern seas and intensive western airflow. The warm sea air outflow caused by Atlantic cyclones and frequent invasions of the air from the Arctic Ocean makes the weather unstable throughout the year [10].

The annual temperature amplitude is 32.7°C. July is the warmest month of the year (mean monthly temperature is +12.4°C) and the coldest is January (mean monthly temperature is −20.3°C). The mean annual temperature is −6.0°C. The number of days with mean daily temperatures exceeding 0°C is 125.

The snow cover thickness increases from November to January (these are the months with the most frequent cyclones). The thickest snow cover is reached by the second decade of March and amounts to 75 cm [10].

The landscapes of the territory belong to the southern tundra undulating hilly plains with a silt loam cover and moraine bedding and the southern tundra and forest—tundra upland and undulating foothills of the Subpolar Urals [10].

The vegetation is composed of tundra plant species, mosses, herbs, and dwarf shrubs, such as sphagnum, common haircap moss (Polycyllium commune), European blueberry (Vaccinium myrtillus), bog bilberry (V. uliginosum), cloudberry (Rubus chamaemorus), dwarf birch (Betula nana), and various sedge (Carex) species.

The prevalent soil types are Cryosols and Histosols. The study territory resides within the Pechora Plain in the southern tundra subzone, Bol’shezemel’skaya province, and Vorkuta district of the tundra surface-gley and peat-gley frozen soils [1, 6]. Stagnant overwetting by surface water with formation of Histosols over large areas is frequently observed on flat ridges. Better surface drainage leads to formation of various soil associations and mosaic complexes of semihydromorphic and automorphic soils [1, 6].

Field methods. The study was performed in the late September of 2019 and 2020, when the seasonal thawing reached the maximum depth of 80 to 100 cm. A 100-cm-deep soil profile was excavated on the top of the palsa. The soil there belongs to oligotrophic peat soils (Dystric Histosols). The thawed peat was sampled from the profile with intervals of 1—3 cm and sampled for the stable carbon isotope and PAHs analyses. Five samples of frozen peat to the depth of 140 cm were taken by drilling (using a Bosch electric ice drill).

Stable carbon isotope measurements were performed during 1—6 months after sampling. Peat samples were dried at temperature of approximately 25—30°C, comminuted, and sieved (mesh, 0.25 mm). The isotope composition was determined for total peat carbon [5, 33, 34].

Carbon isotope composition was determined at the Isotope Laboratory with the Faculty of Geography, Moscow State University, in a Delta-V Plus isotope ratio mass spectrometer with a standard EA 1112 HT O/H-N element analyzer. The IAEA-CH-3, IAEA-CH-6, IAEA-600, and USGS-24 international standards were used for the calibration. The measurement accuracy of δ13C was ±0.2‰.

PAHs were analyzed by high performance liquid chromatography. The preparation of samples started from the extraction with dichloromethane (20 mL per 1 g dry peat). The resulting extract was filtered, evaporated, re-extracted with hexane, and purified using modified silica gel Diapak-C cartridges. The air-evaporation was performed to dryness. The purified extracts were also evaporated and re-extracted with
acetonitrile. PAHs were analyzed in an Agilent 1260 at the Analytical center of the Faculty of Soil Science, Moscow State University. In total, 13 PAHs were identified: naphthalene, fluorene, acenaphthene, phenanthrene, anthracene, pyrene, chrysene, fluoranthene, benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, and benzo(ghi)perylene.

Data processing. The indicator ratios anthracene/phenanthrene and fluoranthene/fluoranthene + pyrene [17] were calculated for PAHs. Both ratios demonstrate the degree of pyrogenicity or petrogenicity of the studied PAH associations. The values >1 and >0.5, respectively, suggest a prevalently pyrogenic character; otherwise, prevalently petrogenic one.

Cluster analysis of the PAH concentrations was used for clarifying the affinity of different compounds for one another. The analysis was performed in R environment using agglomerative algorithm (cluster package).

RESULTS AND DISCUSSION

The average PAH concentration in peat amounts to 63 ppb with a median value of 34 ppb. Most PAH concentrations fall into the range of 10 to 40 ppb. The PAH content is elevated (42 ppb) at the palsa surface and somewhat decreases (to 33 ppb) with the depth. Sharp peaks of PAH concentration, amounting to 360 and 260 ppb, are observed at depths of 60–63 and 84–87 cm, respectively. In general, the average PAH content in the peat of the lower part of the palsa is higher as compared with its upper part (36 ppb in the lower part versus 28 in the upper part). A minor local increase in the concentration is also observable at depths of 114–117 and 141–144 cm. The minimum PAH concentrations were detected at depths of 6–9 and 129–132 cm (Table 1).

The prevalent individual PAH is benzo(a)anthracene (average content, 26 ppb and median, 8 ppb) followed by benzo(k)fluoranthene and benzo(b)fluoranthene (average content for both, 8 ppb and median, 3.4 ppb for the former and 2.6 ppb for latter). The concentrations of heavy compounds display a distinct depthwise trend: the share of benzo(a)anthracene is dominant to the depth of 60–63 cm, whereas benzo-fluoranthenes account for the major PAH share in deeper layers (Fig. 1).

The carbon isotope composition in peat displays a weak variation. The maximum δ13C value is –28.05‰, being observed at the depth of 63–66 cm and the minimum δ13C value is –30.5‰, being observed at the depth of 33–36 cm (Fig. 2). The average δ13C value is –29.15‰. Any distinct trend in the change of 13C content with depth is unobservable. However, lower car-

| Sample ID | Depth, cm | Total PAHs, ppb | Anthracene/phenanthrene | Fluoranthene/ [fluoranthene + pyrene] |
|-----------|-----------|-----------------|------------------------|--------------------------------------|
| 1         | 0–3       | 42.39           | 0.14                   | 0.69                                 |
| 2         | 6–9       | 9.73            | –                      | 0.97                                 |
| 3         | 12–15     | 33.38           | 0.25                   | 0.84                                 |
| 4         | 15–18     | 12.33           | 0.82                   | 0.57                                 |
| 5         | 18–21     | 28.44           | 0.02                   | 0.67                                 |
| 6         | 24–27     | 34.44           | –                      | 0.80                                 |
| 7         | 33–36     | 31.95           | –                      | 0.76                                 |
| 8         | 51–54     | 33.51           | –                      | 0.75                                 |
| 9         | 59–62     | 67.66           | 43.19                  | 0.00                                 |
| 10        | 60–63     | 359.12          | 0.30                   | 0.26                                 |
| 11        | 78–81     | 36.06           | –                      | 0.00                                 |
| 12        | 84–87     | 256.61          | 3.29                   | 0.13                                 |
| 13        | 87–90     | 65.84           | –                      | 0.74                                 |
| 14        | 96–99     | 36.42           | –                      | 0.37                                 |
| 15        | 105–108   | 23.88           | –                      | 0.66                                 |
| 16        | 114–117   | 51.81           | –                      | 0.19                                 |
| 17        | 123–126   | 17.38           | –                      | 0.40                                 |
| 18        | 129–132   | 11.97           | 0.03                   | 0.00                                 |
| 19        | 141–144   | 50.96           | –                      | 0.00                                 |
bon isotope values are recorded at depths of 36–39, 57–60, and 120–123 cm and higher ones, at depths of 6–9, 63–66, 87–90, and 102–105 cm.

An insignificant positive correlation between naphthalene concentration and peat carbon isotope composition ($r = 0.5$) is observed. The carbon isotope composition displays a weak negative correlation with benzo(a)pyrene. No correlation of the total PAHs with the isotope composition is observed; however, local higher PAH content in the lower part or the profile
correspond to higher isotope values (Fig. 2). An opposite situation is characteristic of the upper part of the profile: higher δ13C values correspond to lower PAH concentrations.

Cluster analysis distinguishes three PAH groups according to their affinity for one another and similar variation: the first cluster comprises anthracene, chrysene, pyrene, and benzo(ghi)perylene; the second cluster contains benzo(a)pyrene, phenanthrene, naphthalene, acenaphthene, fluorene, and fluoranthene; and the third one, benzo(k)fluoranthene and benzo(b)fluoranthene. Benzo(a)anthracene displays no affinity for any of the studied PAHs. A similar clustering was earlier reported [25]; however, a specific position of benzo(a)anthracene and its prevalence in the PAH association have not been earlier observed.

The determined specific features in the distribution, composition, and concentrations of PAHs and carbon isotopes in peat are the result of a joint influence of different factors, including vegetation, matter redistribution during freezing–thawing cycles, climatic effects, and anthropogenic impact.

A comparison of our data to the relevant literature demonstrates that the PAH content in the examined peat palsa is rather low. For example, the PAH concentrations of 160 to 8655 ppb were recorded in the peat palsa near the village of Seida [8] versus the values of 10–360 ppb observed in the palsa near Eletsky settlement. The values recorded in the palsa site are more characteristic of the mineral soils near the village of Seida [7]. In addition, the authors [7] report the prevalence of 2–4-ring PAHs versus the palsa near Eletsky settlement with an absolute prevalence of 5-ring compounds. In the examined palsa, a considerable increase in the PAH content below the active layer bottom was observed, which is analogous to the specific features previously recorded in the peat soil near the town of Inta [9].

The studied territory is near the objects with anthropogenic influence, such as the railroad and an urban-type settlement with furnace heating. These factors affect the PAH content in the palsa sites mainly in the upper peat layers, in particular, the near-surface maximum is most likely determined by this effect, which is confirmed by the presence of a maximum fluoranthene (regarded as a “technogenic” PAH [8]) concentration in the near-surface sample. Most researchers regard benzo(a)anthracene as a technogenic product; note that this compound in the examined palsa was also the prevalent PAH in the peat at the depth of 0 to 60 cm, i.e., to the active layer bottom. Its prevalent content in the entire studied peat layer is most likely associated with the vertical matter transfer mainly determined by the freezing–thawing pro-

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**Fig. 2.** Carbon isotope composition and total PAH concentration in the peat layer.
cesses. The indicator ratios at these depths do not unambiguously suggest a pyrogenic origin of the PAH association in the analyzed peat material, indicating a mixed effect of the factors in this case. The deeper peat layers are rather unaffected by the anthropogenic impact and the PAHs in them are of a natural origin.

This also refers to the sharp peaks in PAH concentration (260 and 360 ppb) at depths of 60–63 and 84–87 cm. This may result from natural paleofires, as the concentrations of pyrene, benzo(b)fluoranthene, and benzo(k)fluoranthene are increased in these layers (according to [36], their production is a characteristic feature of biomass combustion).

The indicator ratios of anthracene/phenanthrene and fluoranthene/(fluoranthene + pyrene) [17] in most samples of the overall peat layer are <1 and <0.5, suggesting a petrogenic (low temperature) origin of the PAHs. On the other hand, the anthracene/phenanthrene ratio at depths of 60–63 and 84–87 cm amounts to 43 and 3, respectively, thereby confirming a pyrogenic origin of the PAHs at these layers.

Presumably, lower PAH concentrations in peat at a depth of 60 cm are of biogeochemical nature and are to a considerable degree determined by the transformation of plant residues determined by soil processes. As is noted above, several researches agree that PAHs, including the heavy compounds, such as benzo(ghi)perylene and benzo(a)pyrene, can be of a plant origin [12, 15, 18]. However, the indicator ratio of fluoranthene/fluoranthene + pyrene in certain (at a depth of over 60 cm) depths in the peat frozen layer suggest a potential contribution of wildfires.

The variation in the PAH composition in the palsa peat in a deeper layer can be also associated with the variation in the composition of plant residues during the second half of the Holocene (as is mentioned above, the radiocarbon age of the peat in the examined palsa at a depth of 0.5 m is 6400 ± 60 years [3]).

The δ13C values in the examined palsa are variable along the profile to a lesser degree than PAHs. In general, these values are rather low (–28 to –30‰) as compared with, for example, the carbon isotope composition of the peatbogs of Central Europe (–26 to –28‰) [20]. Presumably, this is explainable with the specific climatic features: the organic matter of a peatland in cold climate is considerably weaker transformed and, thus, the carbon isotope composition does not become enriched with time. The absence of a depthwise trend of the changes in carbon isotope composition according to [2, 31, 32] suggests that the examined palsa is not currently subject to degradation and remains in a stable state. In addition, the factor of vegetation can also contribute to the situation since the species composition and the efficiency of photosynthesis change following the specific climate features.

Carbon isotope composition of the peat in the examined palsa displays a considerable variation in the δ13C values. Presumably, certain increase of the δ13C values suggests some episodes of lower moistening and aerobic conditions. On the contrary, the episodes of a decrease in the δ13C values may demonstrate an increase in the δ13C values of –28 to –30‰ have been mainly formed with at the expense of grasses.

A comparison of the specific features in a vertical distribution of PAH concentrations and δ13C values in a peatland allows more unambiguously identify the role of paleofires in the formation of peat layer of the palsa. The episodes of paleofires may increase the abundance of 13C relative to the initial matter as well as the concentrations of heavy PAHs, especially pyrene, benzo(a)pyrene, and benzo(a)anthracene. A drastic increase in the PAH content as well as an increase in the 13C abundance is observed at the depth of 60–66 cm in the examined palsa profile, which confirms the assumption on the effect of a paleofire at this depth. On the other hand, note also a vertical migration of the matter caused by freezing–thawing processes, which enhances a relatively uniform distribution of the matter up to the upper boundary of the permafrost.

The carbon isotope composition of palsa peat is determined by the isotope composition of the peat forming vegetation, moistening conditions, and the degree of organic matter degradation.

The carbon isotope composition of the examined palsa peat is relatively depleted (δ13C values vary between –28 and –30‰). These values are close to the isotope composition of herbaceous plants and most likely suggest that peat had been formed mainly of this vegetation type. The variation of isotope composition is noticeable. Presumably, the episodes of an
increased carbon isotope values suggest the periods with poorer moistening and aerobic conditions, while the episodes of decreased isotope values may suggest the prevalence of anaerobic conditions and anaerobic destruction of organic matter.

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**CONFLICT OF INTEREST**

The authors declare that they have no conflicts of interest.

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**SUPPLEMENTARY INFORMATION**

The online version contains supplementary material available at https://doi.org/10.1134/S1064229321070139 and accessible for authorized users.

Fig. S1. Study region on the Google Earth map.

Fig. S2. The examined palsa near Eletsky settlement (photo by N.A. Budantseva).

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