

Transformation of the Group Composition of Organic Matter from the Channel Deposits of a Small River Affected by Anthropogenic Activity

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Abstract—The group composition of organic matter (OM) was studied in the channel sediments of the Pakhra River under natural condition and in the zone affected by the town of Podol'sk, Moscow oblast. It was found that the natural alluvium is poor in OM ($C_{\text{org}} = 0.65\%$), the composition of which is dominated by humus acids (81.8% of C_{org}) at minor fractions of residual OM (16.7%) and lipids (1.5%). Anthropogenic muds formed in the river channel in the zone affected by urban pollution are conspicuous in high OM content ($C_{\text{org}} = 1.26\text{--}2.60\%$), the composition of which is enriched in lipids (up to 10–20%) and residual OM (up to 27.3–48.6%), whereas the fraction of humus acids decreases to 29.6–57.1%. The muds are most significantly enriched in lipids (their specific concentration increases by factors of 6–59 compared with the natural alluvium) and residual OM (by factors of 3–11). The amount and character of the group composition of OM from anthropogenic muds are controlled by the specific features of the sources of sedimentary material in the river and the character of the environment of alluvium sedimentation in the zone affected by an industrial town.

Keywords: organic matter, small river, industrial impact, natural alluvium, anthropogenic mud

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INTRODUCTION

Organic matter (OM) transported into small rivers of anthropogenic landscapes with surface runoff and sewage plays an important role in the formation of the composition of modern alluvium deposits and concentration and behavior of various chemical elements in them. The analysis of available data shows that most previous studies focused on the estimation of the accumulation rate of individual organic compounds in river sequences [1]. The group composition of OM from river sediments, especially affected by anthropogenic activity, is poorly known. On the other hand, this factor controls to a large extent the physicochemical conditions of the environment of alluvium deposition and the direction and intensity of various geochemical, biochemical, and physical processes [2–4]. It can be suggested that the proportions of the main groups of OM characteristic of river sediments in the zones of anthropogenic pollution may be different from those under natural (background) conditions. This was first supposed by Vernadsky [5], who noted that one of the most profound geochemical effects in natural waters associating with anthropogenic activity is a change in the composition of its organic component, which is manifested both in an increase in total OM content and in a transformation of its qualitative structure. The goal of this study was to determine the group composition of OM from the

channel deposits of a small river and the character of its transformation in an urban industrial environment.

REGION AND METHODS

Our investigations were conducted on the Pakhra River in the vicinity of Podol'sk, a large industrial center in Moscow oblast (Fig. 1). The Pakhra is 135 km long, its catchment area is 2720 km², and the mean water discharge estimated during many years in the region of Podol'sk is 9.95 m³/s. Under natural conditions, the regime and water content of the Pakhra, which is an East European-type river recharged mainly by snowmelt, are typical of small rivers in central Russia [6]. During the past decades, industrial and domestic sewage has played an important role in the water budget of the Pakhra River; it supplies significant amounts of peculiar sedimentary materials to the river resulting in the formation of a new type of channel deposits known as anthropogenic muds [7]. The main input of wastewater from Podol'sk to the Pakhra comes from the municipal sewage treatment plant (STP) through the Chernyi Brook. An additional source of anthropogenic sedimentary material in the river is runoff from cultivated areas.

The channel deposits of the Pakhra River (layer 0–20 cm) were sampled using a TBG-1 drill at the follow-

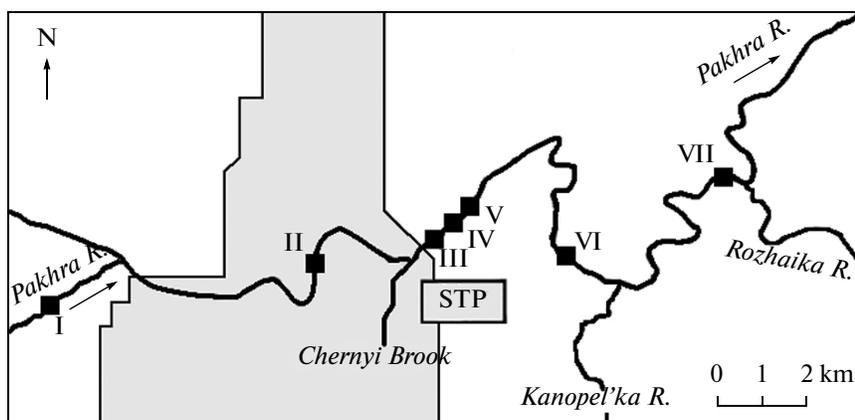


Fig. 1. Location of the studied segments of the Pakhra River in the vicinity of Podol'sk. (I)–(VII) Sites of sampling of channel deposits, STP is the sewage treatment plant, and the industrial urban territory is indicated by shading.

ing key segments: I, at the entrance to Podol'sk; II, center of Podol'sk; III–VII, segments 2, 2.2, 2.4, 9, and 15 km downstream of the of the Chernyi Brook mouth, respectively; and VIII, upper reaches of the river (local background). Within each key segment, no less than three particular samples (macroscopically identical sedimentary materials) were collected near the target site (2–3 m from the water level), and a combined sample (~1 L in volume) was prepared from them. Samples were air-dried in shadow, and the material of each sample was carefully mixed, sieved (1 mm sieve), and quartered to obtain aliquots for subsequent analysis.

The petrochemical composition of sediments was examined using routine techniques (combination of gravimetric, volumetric with complexon, potentiometric, flame photometric, and photocolometric methods); Ni, Cu, Zn, Mo, Ag, Cd, and Pb (as indicators of anthropogenic impact) were measured by atomic absorption; and total organic carbon content (C_{org}) was determined by the method of I.V. Tyurin. In order to sequentially extract the main groups of OM from the sediments, the following protocol of phase analysis was utilized. (1) Alcohol (C_2H_5OH)–benzene (C_6H_6) mixture (1 : 1 in volume, Soxhlet extraction for 20 h at ambient temperature). It is believed that lipids (fats, waxes, and resins) are mainly extracted from sediments [8]. (2) Sodium pyrophosphate solution (0.1 M $Na_2P_2O_7 \cdot 10H_2O$ with 0.1 *n* NaOH, extraction for 12 h, pH ~ 13; samples were treated 3–6 times until the complete discoloration of the solution). Such a treatment of sediments extracts mainly humus acids bound to calcium and nonsilicate compounds of iron and aluminum [9]. Humic and fulvic acids (HA and FA, respectively) were separated using the method of [10], and organic carbon was determined using the method of Tyurin modified by Tsyplov [11]. The amount of organic carbon in the insoluble residue (C_{rom} , characterizing residual OM, including clay–humus humin, lignin, and anthropogenic organics in polluted areas) was calcu-

lated by subtracting the sum of carbon in the alcohol–benzene (C_{lip}) and pyrophosphate ($C_{HA} + C_{FA}$) extracts from the bulk C_{org} content in the sample. The results of all analyses are normalized to the air-dry mass of samples.

RESULTS AND DISCUSSION

Within the background segment, the channel of the Pakhra River is made up mainly of well sorted sands with a small amount (0.4–0.6%) of clay particles. The average (median) size of particles in the background alluvium is ~0.1 mm, and the content of clay grain size fraction (<0.01 mm) ranges from 0.9 to 1.3% [12]. In the zone of Podol'sk influence, where alluvial deposits contain considerable amounts of anthropogenic sedimentary material transported with sewage and surface runoff from cultivated areas, the Pakhra channel is covered by poorly sorted sandy, fine silty, and coarse silty anthropogenic muds dominated by the silt fraction (35.1–53.8%), whereas the relative contents of clay particles and physical clay are 1.5–7.4 and 5.9–16.1%, respectively [12]. The median size of particles in the muds is 0.031–0.075 mm. The anthropogenic muds differ from the background alluvium in a peculiar petrochemical composition and high contents of heavy metals (Table 1).

The background alluvium shows rather low content of OM ($C_{org} = 0.65\%$), which is dominated by humus acids (81.8% of C_{org}), whereas the fractions of residual OM is minor (15.4%) and that of lipids is negligible (1.5%). A characteristic feature is higher HA contents compared with mobile FA, which indicates a very high degree of OM humification in the background sediments (Table 2).

The anthropogenic muds differ from the background alluvium in significantly higher (by a factor of 2–4) total OM content and fundamentally different balance of main OM groups (Table 2, Fig. 2). The muds

Table 1. Chemical composition of the channel deposits of the Pakhra River

Component	Sampling segments							
	VIII	I	II	III	IV	V	VI	VII
Major components, %								
SiO ₂	79.39	73.64	65.54	69.47	67.97	68.04	71.52	75.61
TiO ₂	0.41	0.39	0.57	0.34	0.42	0.40	0.31	0.42
Al ₂ O ₃	4.34	6.72	6.84	6.97	6.81	6.20	5.39	4.92
Fe ₂ O ₃ + FeO	2.01	2.34	3.28	2.99	3.01	3.21	2.98	2.12
MnO	0.06	0.06	0.08	0.05	0.09	0.08	0.06	0.05
MgO	0.98	0.89	1.40	0.61	0.82	0.42	0.86	0.63
CaO	4.10	4.58	6.23	4.79	5.05	5.44	4.88	4.11
Na ₂ O	0.77	0.61	0.74	0.61	0.74	0.72	0.55	0.61
K ₂ O	1.62	1.37	1.74	1.32	1.61	1.97	1.49	1.37
P ₂ O ₅	0.22	0.28	0.30	0.41	0.68	0.69	0.55	0.39
H ₂ O ⁻	0.64	0.77	1.03	0.67	1.09	1.12	0.93	0.89
H ₂ O ⁺	1.57	2.37	3.70	3.58	3.69	3.98	3.24	1.61
CO ₂	3.17	3.22	5.58	3.81	3.64	3.11	3.38	3.11
LOI*	1.31	3.41	3.58	3.89	4.93	4.98	3.97	3.69
Total	100.59	100.65	100.61	99.51	100.55	100.36	100.11	99.53
Trace elements, ppm								
Ni	21	31	38	60	80	80	50	30
Cu	29	42	70	500	500	600	300	100
Zn	50	60	75	300	300	300	200	80
Mo	0.8	1.1	1.3	1.8	4.1	3.2	2.2	1.1
Ag	0.05	0.08	0.09	3.11	5.12	4.09	3.12	1.10
Cd	0.1	0.2	0.3	4.6	5.8	6.2	2.1	1.1
Pb	23	40	70	300	400	400	300	100

* Loss on ignition.

show very high enrichment factors for residual OM (3–11) and, especially, lipids (6–59) (Table 3). In turns, the relative fraction of lipids increases in the muds up to 10–20% (as compared with 1.5% in the background alluvium), and that of residual OM is up to 27.3–48.6% (versus 15.4%). Simultaneously, the anthropogenic muds show a decrease in the relative fraction (at a slight increase in specific content) of humus acids (from 81.8% in the background alluvium to 29.6–57.1% in the muds).

Away from Podol'sk, the anthropogenic muds are progressively depleted in total OM (at the expense mainly of poorly soluble organics and HA) and show an increase in the specific content and relative fraction of FA. This results in a change in the type of humus and the degree of OM humification in the channel deposits. For instance, as was noted above, the background alluvium is characterized by a very high degree of OM humification (owing to its oxidation transformation), which is typical of rivers and water reservoirs of the

humid zone [4, 13], whereas the anthropogenic muds, especially in the zone of their maximum abundance (segments III–V), show a lower degree of OM humification, which indicates the predominance of reducing processes in the zone of anthropogenic impact (Table 4). In turn, while the background alluvium is characterized by a fulvate–humate type of humus, the anthropogenic muds from the zone adjacent to the pollution source contain fulvate-type humus (segments II and III), which is changed downstream by humate (segments IV and V) and, then, humate–fulvate (segments VI and VII) types, which evidently reflects the existence of spatial differentiation of physicochemical conditions and sedimentation processes in the channel. In particular, it cannot be excluded that humatogenesis [16] occurs in the Pakhra River within the proximal zone of urban pollution (segments IV and V), where the anthropogenic muds show $C_{FA}/C_H < 1$, and calcium is most abundant among the adsorbed bases; i.e., the least mobile stable organomineral derivatives of humic sub-

Table 2. Group composition of OM in the channel deposits of the Pakhra River

Segment	C_{org} , % of sediment	% of C_{org}				residual OM
		lipids	humus acids			
			total	FA	HA	
I	1.38	4.4	43.5	22.5	21.0	52.1
II	1.52	6.6	50.0	34.2	15.8	43.4
III	1.71	9.9	32.2	21.1	11.1	57.9
IV	2.46	13.4	36.2	16.3	19.9	50.4
V	2.60	22.6	29.6	13.1	16.5	47.7
VI	1.65	20.0	46.7	26.7	20.0	33.3
VII	1.26	15.9	57.1	33.3	23.8	27.0
Mean (II–VII)	1.87	14.7	41.9	24.1	17.9	43.3
VIII (background)	0.65	1.5	81.8	39.4	42.4	16.7

stances, calcium humates, are formed and (to a higher degree) accumulated in the muds owing to the hydraulic settling of suspended materials in sewage. The compositional peculiarity of OM from the anthropogenic muds and their difference from the background alluvium are clearly seen in the values of geochemical coefficients (Table 5). It is characteristic that the concentration of organic carbon (C_{org}) in the anthropogenic muds is significantly higher than that of carbonate carbon (C_{carb}) (in contrast to the background alluvium and other sediments). For instance, C_{carb}/C_{org} is 7.5 in Phanerozoic sediments, 5.4 in the sedimentary shell of the Earth, 5.3 in the sedimentary layer of the continen-

tal crust, 2.9 in Cenozoic sediments [17], 1.3 in the background alluvium, and 0.7 (on average) in the anthropogenic muds. This indicates an important role of anthropogenic muds in the local geochemical cycle of organic carbon.

It is well known that practically any phase method for the determination of the group composition of OM in sediments is arbitrary to some extent [14, 15]. However, in our case, the most important problem was to establish the systematic tendency of a sharp increase in the total content of OM and a significant change in the distribution of the group composition of OM in riverine deposits formed in the zones of anthropogenic pollution rather than to identify accurately (qualitatively and quantitatively) organic components.

For instance, the relatively low C_{org} content (0.65%) in the background alluvium of the Pakhra is due to its accumulation in an environment with an active geodynamic regime, which promotes the removal of organic detritus and pelitic particles from the sediments and formation of the so-called lithogenic facies of channel deposits, the composition of which is dominated by sandy fractions and silica. It looks likely that the contents and the structure of the group composition of OM determined in the background alluvium are typical of small plain rivers under natural conditions. For instance, sands (even silty) from the channel shoals of rivers of the central Russian Plain contain 0.10–0.34% C_{org} [2]. The mean OM content in the riverine deposits of the humid zone is 1% [13]. The mean C_{org} content in continental sedimentary rocks was estimated as 0.62% [18]. The qualitative composition of OM from the channel alluvium of small plain rivers is controlled under natural (background) conditions mainly by the input of allochthonous material from the catchment, whereas the contribution of autochthonous material is much smaller. The main sources of OM that can be accumulated in the channel deposits of small rivers are catchment soils (main source of humus acids) [19, 20]

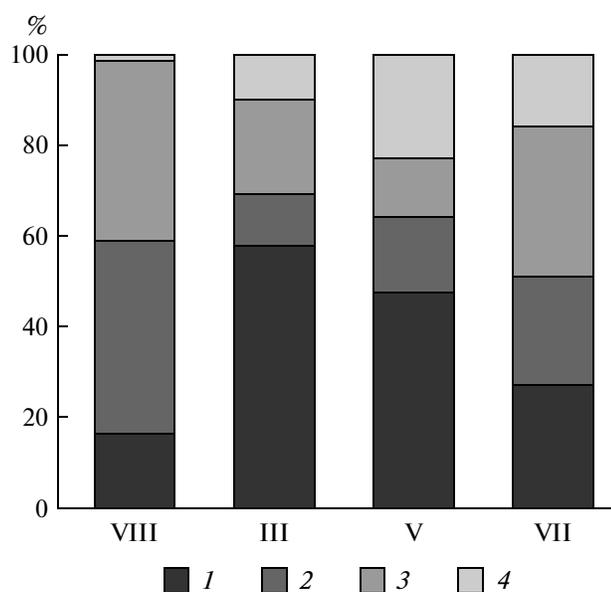


Fig. 2. Group composition of OM from anthropogenic muds (segments III, V, and VII) and background alluvium (segment VIII). (1) Residual OM, (2) humic acids, (3) fulvic acids, and (4) lipids.

Table 3. Degree of OM concentration in anthropogenic muds (concentration factor relative to content in the background alluvium)

Segment	C _{org}	Lipids (C _{lip})	Humus acids			Residual OM (C _{rom})
			total (C _{sol})	FA (C _{FA})	HA (C _{HA})	
I	2.1	6	1.1	1.2	1	6.5
II	2.3	10	1.4	2	0.9	6.0
III	2.6	17	1.0	1.4	0.7	9.0
IV	3.7	33	1.6	1.5	1.8	11.3
V	3.9	59	1.4	1.3	1.5	11.3
VI	2.5	33	1.4	1.7	1.2	5.0
VII	1.9	20	1.3	1.6	1.1	3.1
Mean (II–VII)	2.8	28	1.4	1.6	1.2	7.6

Table 4. Type of humus and degree of humification of OM in the channel deposits of the Pakhra River

Segment	Humus type		Degree of humification	
	C _{FA} /C _{HA}	after [14]	(C _{HA} /C _{org}) × 100%	after [15]
I	0.93	Fulvate–humate	43.5	Very high
II	0.46	Fulvate	50.0	Very high
III	0.53	Fulvate	32.2	High
IV	1.22	Humate	36.2	High
V	1.26	Humate	29.6	Medium
VI	0.75	Humate–fulvate	47.7	Very high
VII	0.71	Humate–fulvate	57.1	Very high
VIII (background)	1.08	Fulvate–humate	81.8	Very high

and, to a much smaller degree, plant litter and metabolic products of aquatic organisms (main source of lipids) [21]. It is known that the OM of the sod–podzolic soils (C_{org} = 1.2–2.3%) occurring in the Pakhra basin is dominated by humus acids (up to 68–69% of total OM) [14], which evidently results in their prevalence in the background alluvium. It is characteristic that even the bottom sediments of uncontaminated freshwater bodies (reservoirs, ponds, and lakes) whose sedimentation is significantly contributed by autochthonous biogenic matter are enriched in humus acids (usually, mainly HA) accounting for up to 40–70% of total OM [4]. The composition of lipids from the background alluvium is probably dominated by stable compounds (hydrocarbons and free fatty acids), and their moderate concentration results from their minor input into the water channel and reflects the transformation of the labile part of OM during early diagenesis. In particular, the specific concentrations of lipids are approximately 0.1% in the arable layer of sod–podzolic soil and 0.06–0.07% in horizon B [22], which is consistent in our case with their low content in the background alluvium. The main component of residual OM, the concentration of which in riverine sediments ranges usually from hun-

Table 5. Spatial variations in geochemical coefficients in the channel deposits of the Pakhra River

Segment	C _{carb} /C _{org}	Fe oxides/C _{org}	Al ₂ O ₃ /C _{org}	CaO/C _{org}
I	0.6	1.7	4.9	3.3
II	1.0	2.2	4.5	4.1
III	0.9	1.8	4.1	2.8
IV	0.4	1.2	2.8	2.1
V	0.3	1.2	2.4	2.1
VI	0.6	1.8	3.3	3.0
VII	0.7	1.7	3.9	3.3
Mean (II–VII)	0.7	1.7	3.5	2.9
VIII (background)	1.3	3.1	6.7	6.3

dredths to a few percent [19], is evidently the products of lignin destruction and clay–humus humin.

The abundance and distribution of OM groups in the anthropogenic muds are also systematic and con-

trolled primarily by the character of sediment sources for the Pakhra River in the zone of Podol'sk influence. It was shown [23] that anthropogenic riverine muds from the areas affected by industrial urban activity are based mainly on sedimentary material delivered to water streams with industrial and domestic sewage, and sludge produced in sewage treatment plants during purification operations is a peculiar geochemical analog of this material and, correspondingly, anthropogenic muds. According to the available data, sewage sludge contains benzene compounds (up to 50–90% of total OM), fats (7–17%), alpha-cellulose (2–12%), hemicellulose (3–25%) [24], and considerable amounts of lipids [25] and is distinguished by minor relative fractions of humus acids (~20% of total OM) [26]. According to [27], the composition of sludge (recalculated to dry mass) includes 4.5% lignin, 2% cellulose, and 1.8% humic acids. Low content (<0.03%) or even absence of humus acids is typical of industrial sewage sludges, which contain more than 100 other organic compounds [28]. The fraction of humus acids in domestic wastewater is much lower (30.1–41.3% of total dissolved OM) [29] than in natural surface waters (60–80%) [30]. It is known that an increase in the specific and relative contents of the group of resistant (poorly oxidized) organic compounds is typical of effluents discharged from municipal sewage treatment plants [31]. For instance, according to Sinel'nikov [32], the fraction of resistant compounds in the total amount of OM in sewage and contaminated waters is up to 60–65%. In polluted environments, anthropogenic muds may accumulate higher fatty acids (for instance, components of synthetic surfactants) and petroleum products (delivered to the water stream with runoff from urbanized areas) with relatively low decay rates, which results in high contents of residual OM in the deposits. Humates of Ca, Al, Mn, and Fe can also be formed in muds; these compounds show low solubility and high resistance against microbial decomposition. The elevated OM content in the unhydrolyzed residue of anthropogenic muds could be related to the sorption of a number of organic compounds on the surface of highly dispersed inorganic fractions, which are very abundant in muds. It should be noted that an increase in the content of poorly oxidized OM in the deposits of polluted water streams was noted long ago by Bunch et al. [33]. Therefore, the sedimentary material transported to rivers by sewage and runoff from urbanized areas is rich in lipids and poorly hydrolyzed OM and poor in humus acids, which is the main reason for the peculiar group composition of OM in anthropogenic muds.

CONCLUSIONS

Under natural (background) conditions, the distribution of organic matter (OM) in the channel deposits of a small river is controlled mainly by the mechanical differentiation of introduced allochthonous sedimentary material and, to a smaller degree, by the superim-

posed accumulation of autochthonous organic materials. This results in a low OM content in the background alluvium ($C_{\text{org}} = 0.65\%$) and predominance of humus acids (81.8% of C_{org}) in its composition at minor fractions of residual OM (16.7%) and lipids (1.5%). The background alluvium contains OM of the fulvate–humate type with a very high degree of humification, which indicates that oxidation processes prevail in natural environments.

Anthropogenic riverine muds formed in the zone of industrial urban impact are distinguished by higher OM contents (C_{org} is 1.26–2.60%, averaging 1.87%), and the most significant increase was observed for the specific concentrations of lipids (by factors of 6–59 relative to the background alluvium) and insoluble OM (by factors of 3–11). The specific contents of humus acids, which are dominated by FA, increase to a much smaller extent (by factors of 1.3–1.6). The muds are distinguished from the background alluvium by a fundamentally different structure of the group composition of their OM: the relative fractions of lipids and residual OM increase up to 10–20% and 23.7–48.6%, respectively, whereas that of humus acids decreases to 29.6–57.1%. In general, the OM of anthropogenic muds is characterized by medium and high degrees of humification and fulvate or humate types of humus, which indicates the prevalence of reducing processes under the conditions of anthropogenic pollution. Away from the town, the total content of OM in the muds decreases mainly at the expense of humic acids and poorly soluble organic compounds. The anthropogenic muds are significantly enriched in organic carbon relative to carbonate carbon, which distinguishes them from the background alluvium and other sedimentary materials. The peculiarity of the group composition of OM in the muds is caused by the specific features of anthropogenic sources of sedimentary material in the river and the character of the medium of anthropogenic alluvial sedimentation.

The OM that is accumulated in anthropogenic muds definitely controls their essential physicochemical properties and plays an important role in the behavior of many heavy metals. The high OM content in muds results in an increase in biochemical oxygen demand, which is favorable for the formation of anaerobic (gleyed) environments in the river channel; as a result, the migration capacity of metals and their exchangeability between sediments and water increase. Lipids occurring in significant amounts in anthropogenic muds and representing the most labile part of OM may promote the formation of mobile geochemically active species of metals, and the elevated content of poorly hydrolyzed OM is favorable for the extensive formation of their strongly bonded forms. Therefore, anthropogenic muds are significant as long-term secondary sources of pollution of water and aquatic organisms.

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