RESEARCH PAPER

# PETROLEUM HYDROCARBONS IN ARCTIC URBAN LAKE SEDIMENTS (MURMANSK, RUSSIA)

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ABSTRACT. The level of pollution of lake sediments located in the city of Murmansk (Russia) with petroleum hydrocarbons was studied. The most urban lakes can be classified as slightly polluted, representing the maximum acceptable risk for human health and ecosystems; Lake Yuzhnoe – as moderately polluted with a negative impact on the aquatic environment, and Lake Ledovoe located near the highway, gas station, and car service, – as dangerously polluted, posing a threat to human health and requiring remediation. The background content of organic compounds identified as petroleum hydrocarbons in the sediments of Murmansk lakes ranges from 76 to 307 mg TPH/kg. High background concentrations of hydrocarbons can be associated with a large amount of organic matter in the sediments of lakes. The standards for the content of hydrocarbons in sediments, developed for other regions, give an ambiguous assessment of their content in the sediments of lakes in Murmansk. It is not entirely correct to use the background content of hydrocarbons in the sediments of lakes in other regions, which differ in particle size distribution, content of organic matter, and chemical composition, to assess the degree of pollution. Additional research is needed to determine the content of hydrocarbons in the sediments of background lakes in the Murmansk region and to develop standards for the content of hydrocarbons in the bottom sediments of Arctic cities.

KEYWORDS: urban lakes, sediments, total petroleum hydrocarbons, Arctic

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## INTRODUCTION

Lakes and rivers located in the city are always subject to strong negative anthropogenic impacts. Situated in the natural depressions of the area, they accumulate pollution from the adjacent territories. Petroleum hydrocarbons and heavy metals are two of the most common types of water pollution. These pollutants accumulate in sediments due to their high sorption capacity, fractionation characteristics in water and sediments, and biochemical stability.

Urban rivers and lakes often suffer from constant or regular discharges from enterprises, as well as spontaneous garbage dumps along the banks. Cienfuegos Bay (Cuba) receives up to 50 tons of high molecular weight petroleum hydrocarbons annually (Tolosa et al. 2009). In other countries, much attention was paid to studying the accumulation and transformation of TPH in sediments. In the sediments of Yuan Dan Lake (China) the TPH content is 1397 mg/kg, and in the sediments of Xiamen Harbour it is within the range of 133–943 mg/kg (Ou et al. 2004). The TPH concentrations in the samples from Lake Chao Hu, one of the most eutrophic lakes in China, varied from 11.9 to 325 mg/kg dry weight (Wang et al. 2012). In Algoa Bay (Eastern Cape Province of South Africa) the TPH content varied from 45.07 to 307 µg/L in the water and from 0.72 to 27.03 mg/kg in the sediments (Adeniji et al. 2017a), but in the Buffalo River Estuary (East London, South Africa) the TPH content varied from 12.59 to 1100 mg/kg in the sediments, with the mean values of 209.81±63.82 mg/kg (Adeniji et al. 2017b). Studies in Nigeria have shown that the TPH content in the Qua Iboe River is from 90 to 250 µg/L in the water and from 270 to 830 mg/kg in the sediments (Inyang et al. 2018), while in the sediments of Ikoli Creek the TPH content ranges from 0.001 to 0.44 mg/kg (Ighariemu et al. 2019). The transfer of TPH from the surface of Crater Lake in North America led to its accumulation in the sediments (1440 mg/kg), especially in the places of ship mooring (Oros et al. 2007). In the sediments of the middle part of the Clyde estuary (Scotland), the TPH content varied from 34 to 4386 mg/kg (Vane et al. 2011). The study of Lake Zug (Switzerland) showed that the sediments near the densely populated northern end of the lake accumulate more hydrocarbons – up to 900 mg/kg, while in the central part of the lake, with a lower population density, the content of hydrocarbons did not exceed 50 mg/kg (Giger et al. 1974).

In the Arctic, negative impacts on aquatic ecosystems have more pronounced consequences. Severe climatic conditions contribute to high vulnerability and significantly reduce the rate of natural restoration of ecosystems under technogenic impact (Nøst et al. 1997; Zubova et al. 2020).

In Russia, TPH accumulation in the sediments of lakes and rivers is studied in oil-producing regions, mainly in Western Siberia (Vorobyev and Popkov 2005; Panicheva et al. 2013; Uvarova and Zakharova 2016). The high concentrations of TPH are characteristic of lake sediments located in industrial impact zones. After a terrible environmental disaster in 1994–1997, a full study of the oil spill's effects on the Pechora River ecosystem found that TPH levels were high, ranging from 360 to 1250 mg/kg in the river's mouth and delta. In that accident, 100 to 375 thousand tons of raw oil entered the water body ecosystem (Lukin and Dauvalter 1997; Lukin et al. 2000). The TPH content in the sediments of urban lakes was also assessed. The sediments of these lakes can accumulate up to 10,000 mg TPH/kg (Belkina et al. 2008; Ivanov et al. 2011; Ivanov 2012; Guzeeva 2014).

At the same time, insufficient attention has been paid to studying urban lakes in the Arctic zone. Among the six largest cities in the world located beyond the Arctic Circle, five are in the Russian Federation, in particular on the territory of the Murmansk region, where there is a high level of urbanization and a concentration of industrialized territories.

The aim of this study was to assess the content of petroleum hydrocarbons in the sediments of urban lakes in the Arctic.

## MATERIALS AND METHODS

### Study Area

Murmansk is the largest city located beyond the Arctic Circle in the world and one of the largest ports in Russia. The population of Murmansk is approximately 290,000 people. It is located on the rocky eastern coast of Kola Bay of the Barents Sea. Murmansk has around 20 lakes. To fully assess the impact of the urban environment on lake ecosystems, seven lakes located in different parts of Murmansk and having great recreational significance for the city's residents and visitors were selected for the study.

Sediment samples were taken in July 2020 and 2021 from seven lakes located in the city of Murmansk: Severnoe (69°01'58.0"N, 33°07'04.1"E), Semenovskoe (68°59'28.8"N, 33°05'24.5"E), Srednee (68°58'51.7"N, 33°07'13.8"E), Treugolnoe (68°58'05.1"N, 33°00'10.5"E), Okunevoe (68°57'00.8"N 33°07'37.0"E), Ledovoe (68°55'58.74"N, 33°6'17.72"E) and Yuzhnoe (68°53'03.2"N, 33°04'31.2"E) (Fig. 1). All these lakes are experiencing anthropogenic impacts to a greater or lesser extent (Slukovskii et al. 2020; Slukovskii et al. 2021).

The morphometric parameters of these lakes were estimated before this study (Slukovskii et al. 2020). The water of lakes Severnoe, Semenovskoe, Treugolnoe, Ledovoe, and Yuzhnoe belongs to the chloride class and the sodium group; the water of lakes Srednee and Okunevoe belongs to the hydrocarbonate class and the calcium group. Moreover, was detected increased (compared to the background level) pH, mineralization, and concentrations of major ions, nutrients, and potentially toxic elements in the water of these lakes (Postevaya et al. 2021; Slukovskii et al. 2020; Slukovskii et al. 2021).

### Sediment sampling

The sediments of the lakes, including the cores of the sediments, were collected at the lakes' points of maximum depth using the Ekman sampling grabber (for all lakes in triplicate in 2020 and 2021) and the Limnos gravity sampler for further layer-by-layer analysis (for all lakes except Treugolnoe and Yuzhnoe in 2021). Immediately after the sampling, all the cores were divided into 5-cm layers. After collection, all the samples were placed into plastic containers, which were labelled and packed in a cooler bag.

### Laboratory analysis

The sediment samples were air-dried and powdered before extraction. A portion of the sample (1-3 grams) was placed in a 100-ml flask with a ground glass stopper. A sample in a flask was poured with 10 ml of carbon tetrachloride and stirred for 1 hour. The extract obtained was filtered through a paper filter "white tape" and poured into a bottle. The extraction was repeated two more times with new portions of carbon tetrachloride, 10 ml each. All the extracts were combined, and the total volume was recorded.

The sediment extracts were cleaned up in a chromatographic column (10 mm i.d. × 10 cm) packed with 7 g of activated aluminium oxide. A blank sample was processed the same way for quality assurance.

The TPH content in the samples was assessed by IR spectrometry based on measuring the integral intensity of absorption of C–H bonds of methyl and methylene groups of various classes of organic compounds in non-polar and low-polar solvents using the analyzer AN-2 (Quantitative chemical analysis of soils 2005).


**Fig. 1. Map of the lakes' location in Murmansk. 1 – Severnoe, 2 – Semenovskoe, 3 – Srednee, 4 – Treugolnoe, 5 – Okunevoe, 6 – Ledovoe, 7 – Yuzhnoe**

The organic carbon content was determined in air-dry samples using the Nikitin's method with a colorimetric modification. The ashing of organic compounds was carried out with a chromium mixture when heated to 150°C in an oven. The amount of carbon was determined by the amount of green trivalent chromium formed as a result of the reaction using a photocolorimeter (wavelength 540 nm). The conversion factor from organic carbon (OC) content to organic matter (OM) content was taken to be 2 (Agatova et al. 2013).

### Data analytical methods

The results were statistically processed. The arithmetic mean, standard deviation, median, and Spearman correlation coefficients were calculated. Statistical calculations were carried out in Microsoft Excel 2010.

## RESULTS

The amount of TPH in sediment samples from the 0–10 cm layer that were grabbed by the Ekman grabber ranged from 928 to 22,175 mg/kg (Table 1). Lake Ledovoe, situated near a busy highway under significant anthropogenic pressure, had the highest TPH content. Moreover, Lake Ledovoe is located 3.4 km from the Murmansk thermal power plant, which uses fuel oil. One of the streams flowing along the highway (Kolskiy Avenue), a gas station, several car dealerships, and a large bus transportation company (both on the surface and in the collector under the road) flow into Lake Ledovoe.

In the sediments of Lake Srednee and Okunevoe, the TPH content was the lowest and did not exceed 946 and 928 mg/kg, respectively. Due to their forest surrounds and distance from potential pollution sources, these lakes endure less anthropogenic pressure..

In the analyzed cores, the TPH content decreased with depth, especially in Lake Srednee (Fig. 2A). In the sediment samples from this lake, taken from a depth of 30-35 cm, the TPH content was 76 mg/kg.

There was no significant decrease in the TPH content with depth in the sediments of Lake Ledovoe, where the TPH concentrations varied from 14,116 to 25,037 mg/kg. The sediment samples from this lake contained a large amount of TPH, even in the 25-30 cm layer (Fig. 2E). The TPH concentrations in this layer served as background concentrations for the other studied lakes. In the sediments of Lake Semenovskoe, an increase in the hydrocarbon content in the 10-20 cm layer was noted (Fig. 2D).

**Table 1. TPH and organic matter content in sediments (0-10 cm) of urban lakes**

| | Severnoe | Semenovskoe | Srednee | Treugolnoe | Okunevoe | Ledovoe | Yuzhnoe |
|---|---|---|---|---|---|---|---|
| TPH content, mg/kg | 2735±322* | 2610±957 | 946±305 | 1706±295 | 928±70 | 22175±5023 | 1862±105 |
| OM content, % | 28.7±3.5 | 32.6±4.1 | 26.7±3.8 | 37.6±3.7 | 30.1±2.5 | 13.5±4.8 | 6.9±1.1 |

*Here and below the figures show the standard errors of the mean.

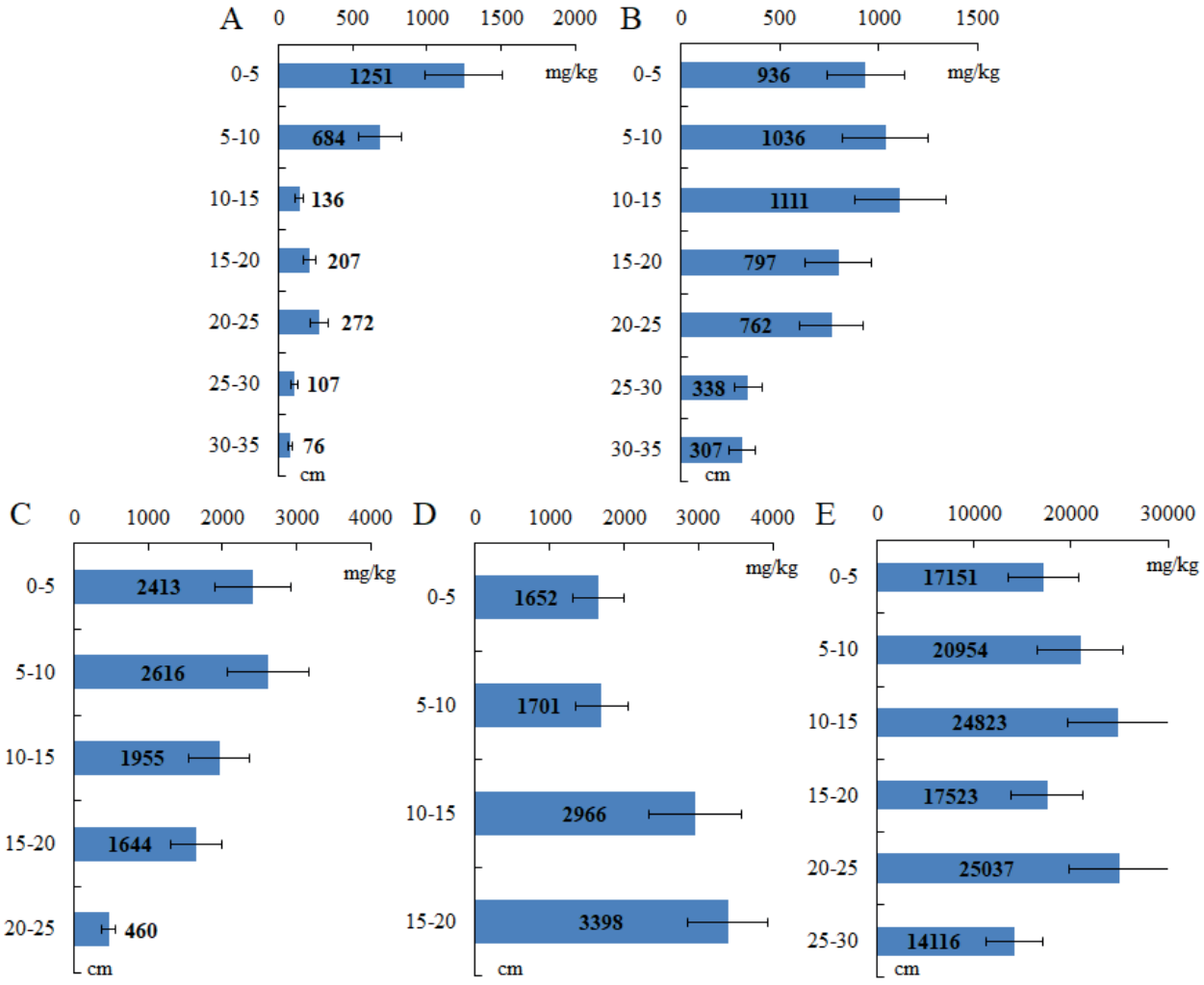


**Fig. 2. The TPH content (mg/kg) in the sediment cores from lakes Srednee (A), Okunevoe (B), Severnoe (C), Semenovskoe (D) and Ledovoe (E)**

The lake's current load is shown by the layer of sediment on the surface, and natural background concentrations are shown by the deeper part of the core (usually more than 20–25 cm) (Dauvalter 2012). Thus, the data obtained allowed us to draw a preliminary conclusion that the background content of organic compounds identified as TPH in the sediments of the lakes in Murmansk ranges from 76 to 307 mg/kg. The rate of sedimentation in the lakes of Northern Fennoscandia, including the Murmansk region, is on average 1 mm per year, and the range is from 0.3 to 3 mm/year (Dauvalter and Kashulin 2018). However, with a high anthropogenic load, the rate of sedimentation can increase due to the supply of large amounts of suspended mineral and organic substances. Taking this fact into account, we can say that the accumulation of hydrocarbons in sediments began about 70 years ago and continues today.

The surface layers of the sediments in Kola Bay (Murmansk region), which has the highest concentration in north-western Russia, previously showed similar hydrocarbon content values (average 1615 mg/kg) (Shakhverdov and Shakhverdova 2016). The highest hydrocarbon concentration (up to 2375 mg/kg) was observed in the areas located in the southern knee, as well as in the middle knee in the Severomorsk area. In general, the TPH content in the sediments of Murmansk's urban lakes is comparable to the that in Kola Bay's sediments. This indicates a high total anthropogenic load on the aquatic ecosystems in Murmansk.

To assess the degree of sediment pollution in the urban lakes of Murmansk, the data on the TPH content should be compared with the maximum permissible concentration (MPC) or the background content in the region.

The Murmansk region does not have established TPH content standards. However, there are standards for other regions of Russia and other countries. The sediments with TPH more than 15 mg/kg are considered polluted in the Persian Gulf (Massoud et al. 1996), more than 50 mg/kg in the countries of South Africa (Environmental Guidelines and Standards for The Petroleum Industry in Nigeria 2002), more than 500 mg/kg in Canada (Atlantic Risk-Based Corrective Action for Petroleum Impacted Sites in Atlantic Canada 2015). Regional documents in Russia regulate the hydrocarbon content of sediments. The regional standard «Maximum permissible level of oil and oil products in the sediments of surface water bodies on the territory of Khanty-Mansiysk Autonomous Okrug – Yugra» (dated 10.11.2004) and the regional standard «Norms and criteria for assessing sediment pollution in water bodies of St. Petersburg» (dated 22.07.1996) are among the examples. The latter document was developed based on the standards and criteria proposed by the Dutch Environmental Protection Agency (DCMR), the Center of Soil Research (TNO), and HASKONING. When assessing sediment contamination, the content of organic matter is taken into account in the regional standard for St. Petersburg.

The highest content of organic matter was observed in the sediments of lakes Triugolnoe (37.6%), Semenovskoe (32.6%), Okunevoe (30.1%), Severnoe (28.7%) and Srednee (26.7%), the lowest – the sediments of lakes Ledovoe (13.5%) and Yuzhnoye (6.9%) (Table 1).

The sediment pollution of the Murmansk urban lakes was assessed following these standards (Table 2).

The sediments of Lake Ledovoe are dangerously polluted and require prompt intervention and remediation. The sediments of Lake Yuzhnoe are moderately polluted and can have a negative impact on the aquatic environment. The sediments of the other studied lakes are slightly polluted and represent the maximum acceptable risk for human health and the ecosystem.

## DISCUSSION

The most polluted lake in Murmansk is Lake Ledovoe, in the catchment area of which there are residential zones, highways, car parking, gas stations, and a bus transportation company. A stream flows into the lake, which is considered one of the most polluted in Murmansk. Lake Yuzhnoe is the second most polluted lake. In the catchment area of the lake Yuzhnoe, there is truck parking and a gas station. There is also a thermal power plant 1.5 km from the lake. The sediments of these lakes contain the least amount of organic matter, which classifies them as dangerously polluted and moderately polluted in accordance with the Regional standard "Norms and criteria for assessing sediment pollution in water bodies of St. Petersburg". In accordance with foreign standards, all studied urban lakes can be classified as polluted.

In the absence of MPC, the level of sediment contamination can be estimated by the background TPH

**Table 2. Degree of sediment pollution in urban lakes of Murmansk**

<table>
<tr><td rowspan="2">Regulating document</td><td colspan="7">Degree of sediment pollution of lakes</td></tr>
<tr><td>Severnoe</td><td>Semenovskoe</td><td>Srednee</td><td>Okunevoe</td><td>Treugolnoe</td><td>Yuzhnoe</td><td>Ledovoe</td></tr>
<tr><td>Regional standard "Maximum permissible level of oil and oil products in the sediments of surface water bodies on the territory of Khanty-Mansiysk Autonomous Okrug - Yugra"</td><td colspan="7">polluted; depression of the bottom ecosystem</td></tr>
<tr><td>Regional standard "Norms and criteria for assessing sediment pollution in water bodies of St. Petersburg"</td><td colspan="5">slightly polluted</td><td>moderately polluted</td><td>dangerously polluted</td></tr>
<tr><td>Atlantic RBCA (Risk-Based Corrective Action) for Petroleum Impacted Sites in Atlantic Canada. Version 3. User Guidance</td><td colspan="7">polluted</td></tr>
<tr><td>Environmental Guidelines and Standards for the Petroleum Industry in Nigeria, (EGASPIN)</td><td colspan="6">polluted; attention level</td><td>polluted; intervention level</td></tr>
</table>

content. According to the study of sea silt (Kravchenko and Bibichkov 1988), the background TPH concentrations in the sediments are 10-200 mg/kg. The average TPH content in the sediment core (depth 20-50 cm) from Galifax Bay (Canada) is 18 mg/kg (Gearing et al. 1991). It is close to the TPH concentrations in the surface sediments of the Gulf of Mexico (Gearing et al. 1976; Kennicutt et al. 1987), but higher than the background TPH content in the sediments from the Scottish shelf area and other zones of the open ocean (Farrington and Tripp 1977; Keizer et al. 1978; Boehm 1984). The total TPH concentrations in the sediments of Lake Washington (USA) and Lake Zurich (Switzerland) are 20-40 and 10-25 mg/kg, respectively. These values were calculated for depths ranging from 30 to 100 cm (Wakeham 1980).

The calculated background content of hydrocarbons in the sediments of the Murmansk region is 32 mg/kg (Shakhverdov and Shakhverdova 2016), in Lake Onega it does not exceed 50 mg/kg (Belkina et al. 2008), in the water bodies of the Republic of Tatarstan, it is estimated at 50-80 mg/kg (Ivanov et al. 2011; Ivanov 2012), which is many times lower than our results.

Lake Treugolnoe, which we consider a background water body, is located on the western shore of Kola Bay. It is surrounded by forest, and there is an unused road nearby. Near the lake, there are no direct sources of pollution. At the same time, the sediment of this lake contains the largest amount of organic matter (37.6%), which likely affects the content of compounds identified as hydrocarbons (1706 mg/kg). The content of TPH in the sediments of the other background lakes of the Murmansk region and Karelia, located at a considerable distance from pollution sources, ranged from 123 to 1091 mg/kg.

High background concentrations of hydrocarbons can be associated with a large amount of slightly humified organic matter in lakes' sediments. The study of sediments of Lake Chini (Malaysia) revealed that n-alkanes primarily originate from biogenic sources, which are wax-like compounds of higher plants and microorganisms (Bakhtiari et al. 2011). The source of aliphatic hydrocarbons in the sediments of Lake Chaohu (China) was mainly higher plants (31%), fossil fuel combustion (30%), petroleum products (26%), and phytoplankton (19%) (Wang et al. 2012). Hydrocarbons in Crater Lake sediments are also of both biogenic origin (waxy compounds of terrestrial plants and algae) and anthropogenic origin (petroleum products) and enter the lake through direct input and atmospheric transport. The amount of naturally occurring n-alkanes is up to 240 mg/kg (Oros et al. 2007).

It should be noted that the lakes located in Murmansk have a high content of organic sediments (20–50%) (Slukovskii et al. 2020). The process of humification in the studied lakes is slowed down due to the climatic conditions and the significant level of anthropogenic pollution of the water bodies. Aliphatic fragments totally prevailed in the molecular structure of all sediments (Guzeva 2022).

The high content of slightly humified organic matter does not allow for accurate determination of the level of hydrocarbon pollution in urban lakes and requires additional research.

## CONCLUSIONS

The urban lakes of Murmansk are subject to the anthropogenic impact of enterprises and transport. In terms of the hydrocarbon content in the sediments, the studied lakes can be classified as slightly polluted, representing the maximum acceptable risk for human health and ecosystems; Lake Yuzhnoe – as moderately polluted; and Lake Ledovoe – as dangerously polluted, posing a threat to human health and requiring urgent intervention and remediation.

The background content of organic compounds identified as TPH in the sediments of the lakes in Murmansk ranges from 76 to 307 mg/kg. High background concentrations of hydrocarbons can be associated with a large amount of organic matter in the sediments of lakes. At the same time, the standards for the content of hydrocarbons in sediments, developed for other regions, give an ambiguous assessment of their content in the sediments of lakes in Murmansk. Also, it's not entirely accurate to judge the level of pollution by the hydrocarbons that are already present in the sediments of lakes in other parts of the world, since these sediments have different amounts of organic matter, particles of different sizes, and chemicals.

For an objective assessment of the level of urban lake pollution in the Arctic, it is necessary to develop a regional standard for the maximum permissible content of hydrocarbons in sediments, taking into account the high content of organic matter of natural origin.

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