ISSN 0001-4338, Izvestiya, Atmospheric and Oceanic Physics, 2023, Vol. 59, No. 8, pp. 982–989. © Pleiades Publishing, Ltd., 2023.
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Mercury in Frozen Quaternary Sediments of the Spitsbergen Archipelago

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Received March 14, 2023; revised May 4, 2023; accepted May 10, 2023

Abstract—The climate warming–related degradation of permafrost can lead to the entry of climatically and biologically active substances, including mercury, into the biosphere; this work focuses on the analysis of the total content of mercury and organic carbon in 15 cores drilled in frozen Quaternary deposits of the Arctic Archipelago of Spitsbergen. The mercury content was additionally analyzed in bedrock samples, because the studied Quaternary deposits are formed by the weathering of the bedrock of the area. The results show that mercury concentrations in 157 studied samples of frozen Quaternary deposits range from 21 to 94 ng/g, with an average value of 40 ng/g. The expected correlation of mercury content with organic carbon content is not revealed. There are no trends in the accumulation of mercury depending on the lithological facies, geomorphological position, the time of sedimentation, or the freezing conditions. The average content of mercury in bedrock is relatively low, with a mean value of 8 ng/g. This means that the main source of mercury in frozen Quaternary deposits is not bedrock, but the formation of organic matter complexes or sorption on clay particles. In terms of the ongoing discussion about mercury input from permafrost to ecosystems, the results obtained from boreholes can be considered preindustrial background values.

Keywords: mercury, permafrost, organic carbon, Spitsbergen

DOI: 10.1134/S0001433823080054

INTRODUCTION

Other than its obvious negative impact on engineering infrastructure, the climate warming–related degradation of permafrost is important in terms of its potential impact on global biogeochemical cycles. An analysis of the content of climatic and biologically active substances in permafrost, as well as an estimate of the rate of their involvement in the general cycle of substances, is an urgent problem at the intersection of permafrost science and biogeochemistry. Significant reserves of organic matter preserved in permafrost are believed to be involved in the modern cycle when it thaws. The decomposition of organic matter with the participation of microorganisms must lead to its mineralization with the release of greenhouse gases (CO_2 and CH_4), i.e., an increase climate warming (Schuur et al., 2015). The thawing of frozen rocks must be also taken into account from the viewpoint of the possibility of entering the biosphere for significant amounts of mercury forming complexes with organic matter (Schaefer et al., 2020).

Attention to mercury is understandable since, along with As, Cd, Se, Pb, and Zn, it is one of the most toxic metals. Considering the potential danger of mercury, as well as the high content of organic matter in the soils of the permafrost zone and the ability of mosses and lichens to accumulate mercury from the atmosphere, attempts were made to study it in soils and peatlands to a depth of ~3 m (Olson et al., 2018; Schuster et al., 2018; Lim et al., 2020). However, an increase in the depth of seasonal thawing, lowering of the upper permafrost boundary, thermokarst, and thermal abrasion mobilize organic matter and the associated mercury both from surface soil horizons and from underlying frozen deposits. To date, the mercury content in frozen sediments located at depths of up to several tens of meters was estimated in only one work using the example of Yakutia (Rutkowski et al., 2021). This is clearly not sufficient to understand the basic patterns of the mercury distribution in permafrost and answer the question of whether permafrost can be considered a significant storage of mercury and whether permafrost degradation can lead to a sharp increase in the supply of this element to ecosystems.

In order to supplement the currently sparse information about mercury in permafrost strata, in this paper, studies were carried out on the Arctic Archipelago of Spitsbergen, where the thickness of permafrost rocks is more than 200 m. Due to climate warming, permafrost is at risk of degradation (Boike et al., 2018; Christiansen et al., 2021). In the area of the village of Barentsburg, permafrost was studied by domestic scientists since the 1930s in connection with the exploitation of coal deposits. Research continued since 2016 as a part of the study of the Russian scientific expedition to the Spitsbergen archipelago (RAE-S). In recent years, thermometric wells and sites for monitoring the seasonally thawed layer (STL), which were included in the GTNP international observation network, were installed here (Demidov et al., 2016). General features of the permafrost–hydrogeological structure of the territory were established (Demidov et al., 2020), cryogenic phenomena were studied (Demidov, Demidov, 2019; Demidov et al., 2019, 2021, 2022), the dating of sediments was made, and microbiological characteristics of permafrost were given (Karaevskaya et al., 2021a, b). The mercury content in cores was analyzed parallel with the listed studies in the period from 2016 to 2023 directly in the chemical analytical laboratory of RAE-Sh in Barentsburg.

In the abovementioned sources, a detailed description of the temperatures, cryolithological structure, and composition of the water extract from 10 of the 15 wells, the studied materials of which served as the basis for writing this paper, can be found. What makes the area of Barentsburg a useful testing ground for research is both the high degree of its geocryological diversity and knowledge and the availability of information on the mercury content in modern bottom soils of the Grönfjord Bay, on the shore of which it is located (Lebedeva et al., 2018), and in the soils of the neighboring Adventdalen valley (Halbach et al., 2017). Earlier in Spitsbergen, the mercury content in the modern bottom soils of three more bays (Horsund, Vijdefjord, and Diksonfjord) was analyzed, and deglaciation of the archipelago was assumed to lead to increased removal of organic matter and associated mercury (Kim et al., 2020).

This study is aimed at determining the average mercury content in frozen Quaternary sediments and possible variations in its content at a depth depending on the type of sediment and the amount of organic carbon. The task was also to compare the obtained data with previously published results for Spitsbergen and other regions on the mercury content in soils and permafrost. Since wells in Spitsbergen uncovered frozen marine Late Pleistocene and Holocene sediments, comparing the mercury content in them with published data on the mercury content in current bottom soils of the Spitsbergen fjords was a separate task.

NATURAL CONDITIONS OF THE TERRITORY AND OBJECT OF RESEARCH

The natural conditions of the studied area are discussed in detail in (Demidov et al., 2020). The studied area is located at 78° N on the Arctic Archipelago Spitsbergen and is limited to the part of Nordenskiöld Land between the Gröndalen and Hollander valleys (Fig. 1). These two deeply incised valleys give the flat bedrock a mountainous topography. The absolute heights of the peaks reach 800 m above sea level. The average annual air temperature according to the Barentsburg weather station was – 2.5° C in 2022. The soils on the peaks and mountain slopes are primary and desert–arctic with very sparse grass–moss–lichen groups. In the valleys and on sea terraces, typically arctic and arctic-tundra soils are found under slightly sparse willow–forb–moss groups.

Over the last century, there has been the widespread degradation of glaciers in the western part of Nordenskiöld Land due to climate warming. Only in the upper reaches of the Gröndalen and Hollander valleys are small hanging and mountain–valley glaciers preserved. Geologically, the area is confined to the West Spitsbergen grabenlike trough. Paleogene sedimentary rocks come to the surface here as packs of alternating sandstones, siltstones, and mudstones. Quaternary deposits are represented by slope, alluvial, fluvioglacial, and glacial deposits characteristic of mountainous countries, as well as marine deposits. The cover of Quaternary deposits, with the exception of areas where taliks develop, is completely frozen. Near the coastal zone, the thickness of cryogenic rocks is ~100 m, increasing at watersheds to 540 m. The average annual temperatures of rocks at the base of the annual heat turnover layer range from –3.6 to –2.2°C.

The object of our research was frozen Quaternary sediments uncovered by wells (Figs. 1, 2), which make up the staircases of accumulative marine terraces on the southern shore of the Isfjord Bay (wells 2, 3, 4, and 5); the terrace of Cape Finneset (well 7); the intermountain valleys of Gröndalen (wells 8, 8a, 10, 11, 16, 17, 18, and 19) and Hollander (well 20) filled with marine, slope, and river sediments; and a perennial heaving mound in the Gröndalen valley (well 9). The maximal exposed thickness of Quaternary deposits is 25 m (well 8a on a terrace in the Gröndalen valley).

In most wells, the upper part of the section is represented by coarse material with a predominance of gravel, gruss, and pebbles with or without loamy and sandy loam filler. In genetic terms, sediments are products of the destruction of bedrock and their further processing under beach conditions and river processing of slope and fluvioglacial deposits in valleys. They are characterized by massive cryotexture. The average moisture content of sediments is 25%.

The bottoms of wells that are composed of marine sediments are characterized by a predominantly loamy composition. The cryogenic texture is schlieren, and

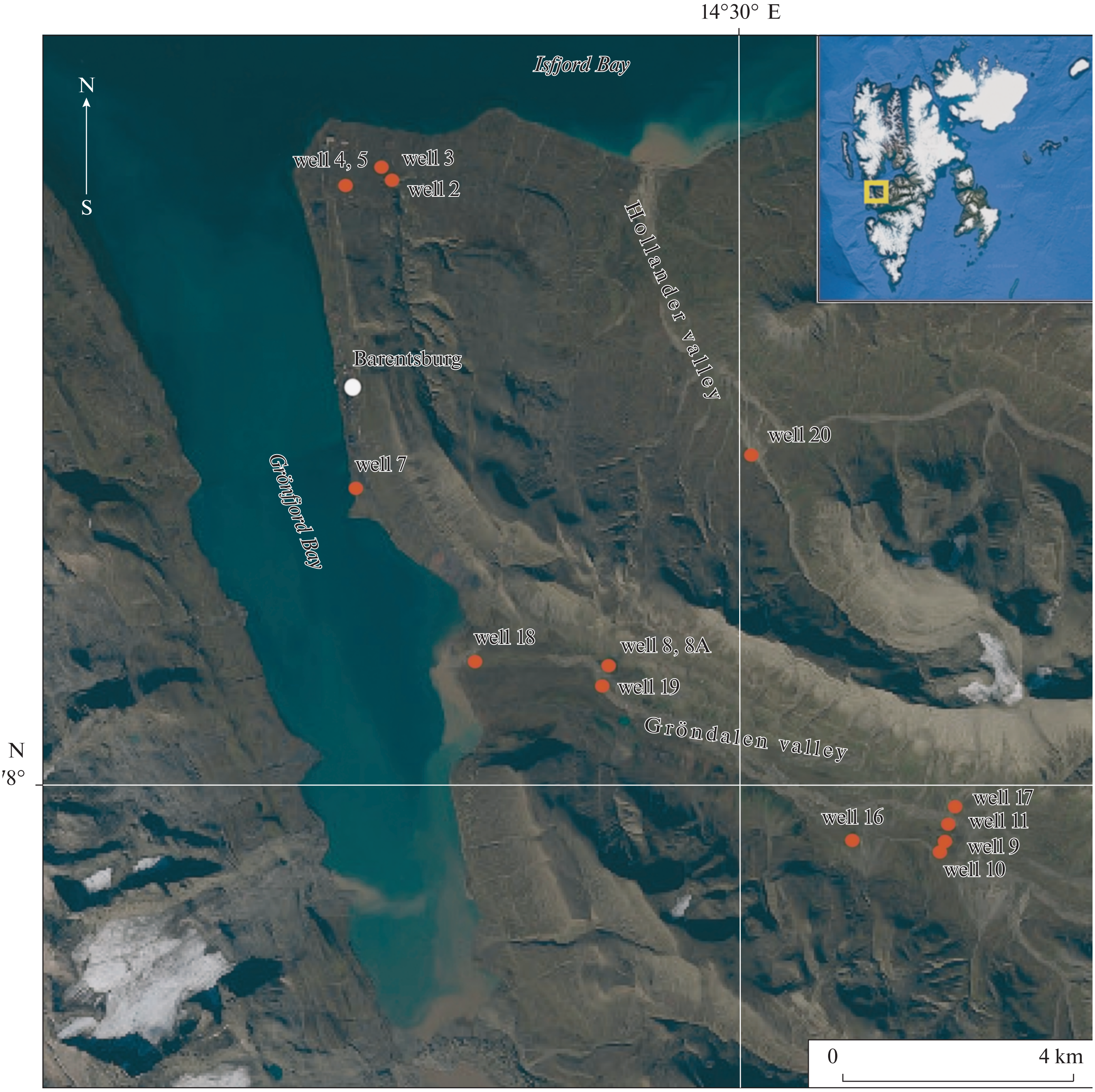

Fig. 1. Aerial photo of the studied area with indication of the drilling locations (image from the website of the Norwegian Polar Institute (https://toposvalbard.npolar.no) with additional information). The inset shows the location of the studied region on the territory of Spitsbergen.

the average moisture content of the sediments is 38%. According to the type of freezing, the loamy member is epicryogenic, while the coarse clastic member could freeze through both the epicryogenic and syncryogenic scenarios. According to the results of radiocarbon dating of samples from terrace deposits at Cape Finneset, the studied deposits were formed at the end of the Late Pleistocene–Holocene (Karaevskaya et al., 2021a).

Since the above-described Quaternary deposits are formed by weathering of rocky soils, it was of interest to study the mercury content in sandstones, siltstones, and mudstones that make up the bedrock of the territory. The object of study of the mercury content in rocky soils were samples taken from outcrops on the slopes of the Gröndalen valley.

MATERIALS AND METHODS

The gross content of mercury in frozen sediments was determined in the chemical–analytical laboratory

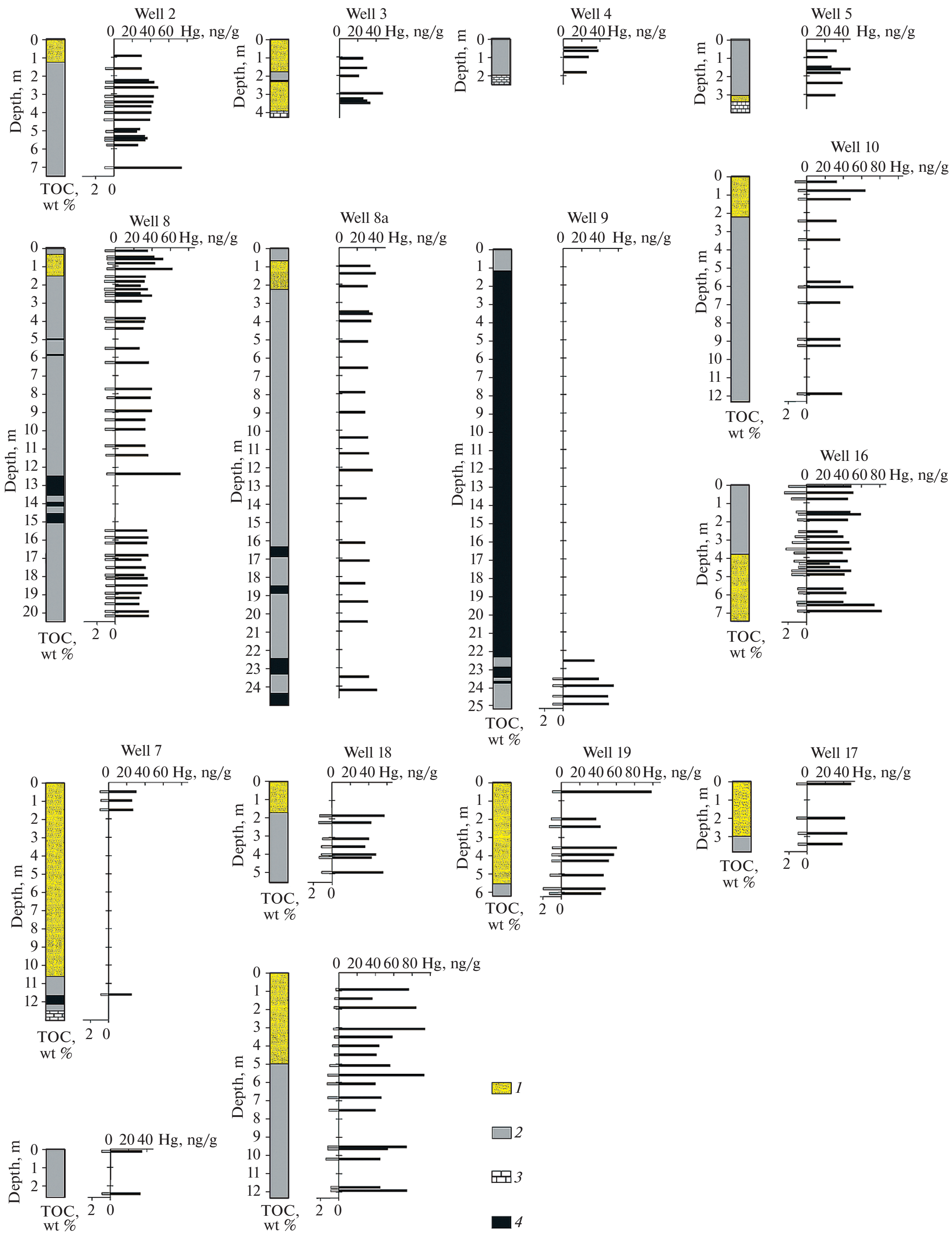

Fig. 2. Lithological well columns and results of determining the content of mercury and organic carbon (TOC) in frozen sediments of the studied region. (*1*) Coarse sediments with a predominance of gravel, gruss, and pebbles with loamy and sandy loam filler, (*2*) loams and clays, (*3*) rocky soil, and (*4*) ice.

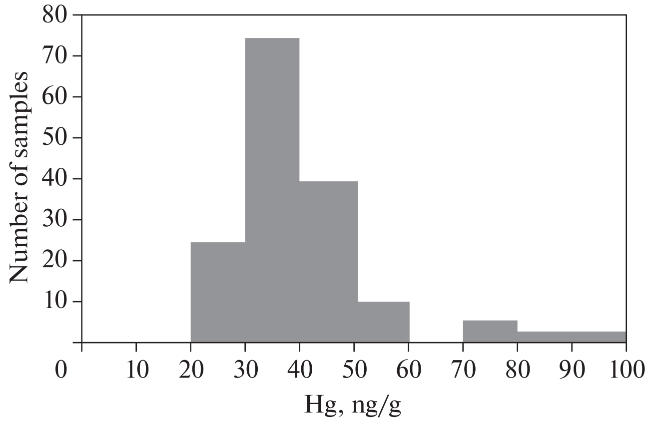

Fig. 3. Histogram of measured values of the mercury content in frozen Quaternary sediments of Spitsbergen.

of RAE-Sh in Barentsburg. After drilling, frozen cores were delivered to the laboratory, where they were dried to an air-dry state. Next, large pebbles and crushed stone were removed from the sample, after which the soils were crushed in a porcelain mortar and sifted through a 0.25-mm sieve. A weighed portion of the crushed sample, with accuracy of to 0.002 g, in a quartz glass boat was placed in an atomizer. Atomization of the mercury contained in the sample was carried out in the pyrolizer of the PIRO-915+ attachment, followed by the transportation of the resulting vapors to an analytical cell. Atomization conditions: air pumping rate was 0.8–1.2 L/min, and evaporator temperature was 680–740°C.

The mercury content was determined by the flameless atomic absorption method with Zeeman correction using a RA-915M analyzer (Lumex, Russia). Each sample was analyzed at least in duplicate, and the discrepancy between measurements was no more than 20%. The integration of the analytical signal and calculation of the gross mercury content were carried out using RAPID software. The calibration coefficient was obtained using a SChT standard sample of the soil composition. Calibration stability was monitored every 30 min or before each new series of samples. A mercury content in rocky soils was analyzed using a similar method.

The content of total organic carbon (TOC) was determined using an Elementar VARIO MAX C elemental analyzer at the Alfred Wegener Institute of the Helmholtz Center for Polar and Marine Research (Germany). The analytical accuracy of measurements was ±0.1 wt %. The samples were predried using lyophilization, homogenized, and the fraction larger than 2 mm was removed.

RESULTS

The results of determining the mercury content in sediments exposed by wells are shown in Fig. 2. No patterns in the distribution of mercury in depth and depending on whether the samples belong to a coarse-grained or fine-grained rock member are observed. There are no significant differences in the mercury content between the STL sediments and the underlying frozen sediments. If we conventionally take 1.5 m as the depth of seasonal thawing (Demidov et al., 2016), then the mercury content varies from 25 to 95 ng/g at an average value of 45 ng/g for 23 samples in STL sediments and from 21 to 94 ng/g at an average value of 40 ng/g for 157 samples in lower lying frozen deposits (Fig. 3).

The mercury content of ten rock samples was also analyzed. The mercury content turned out to be significantly lower compared to that in Quaternary deposits: from 3 to 14 ng/g at an average value of 8 ng/g.

The organic carbon content was also analyzed in 19 samples from STS and 112 samples from frozen sediments, in which the mercury content was determined (see Fig. 2). The average content of organic carbon in STL is 1.1 wt % with a range of values from 0.3 to 2.4 wt %. The average content of organic carbon in frozen sediments is 1.1 wt %, and the range of values is from 0.5 to 2.5 wt %. The pairwise linear correlation coefficient between the mercury content and the amount of organic carbon was –0.04 for STL samples and –0.05 for samples of underlying frozen sediments. The lack of correlation is confirmed by the chaotic distribution of points on the scatter diagram (Fig. 4).

DISCUSSION

Obtained in this study, the average mercury content of 40 ng/g in the frozen sediments of Spitsbergen turns out to be several times higher than the average mercury content of 9.7 ng/g in the frozen sediments of Yakutia given in (Rutkowski et al., 2021). It is not yet possible at this stage of study to explain this difference. One of the explanations could be the increased content of organic carbon in the permafrost of Spitsber-

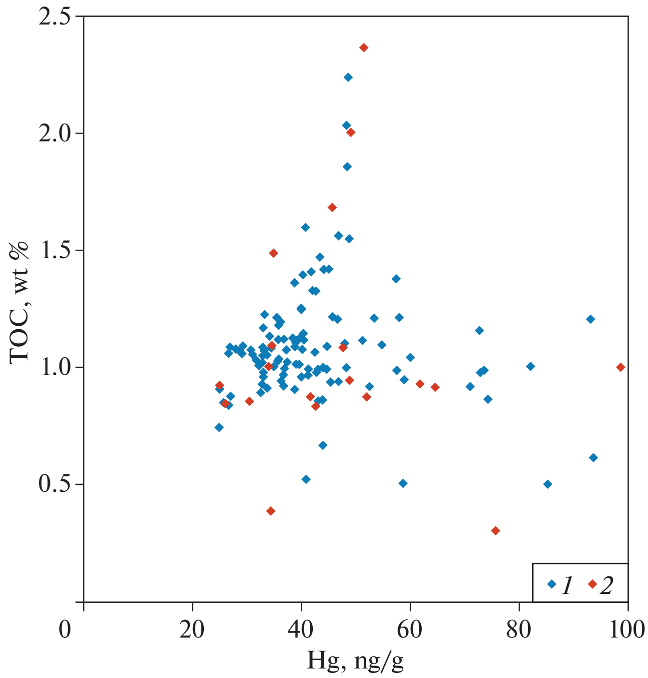

Fig. 4. Scatter diagram of the mercury content versus organic carbon content in (*1*) frozen sediments and (*2*) STS of the studied region.

gen. However, the organic carbon concentration in the studied samples was lower than that in the samples of (Rutkowski et al., 2021). It should be also noted that, earlier in Spitsbergen, the mercury content in mineral soil samples taken from a depth of 20 cm in the Adventdalen valley and in the area of the village of Ny-Alesund was estimated (Halbach et al., 2017). It turned out to be equal to 25 ng/g, which is less than the average mercury content in STL and frozen sediments in the area of Barentsburg.

Mercury concentrations similar to those we obtained in the Spitsbergen STS were previously obtained in Alaska when studying samples from soils and subsoil sediments from depths of up to 3 m, where the average mercury content was 43 ng/g (Schuster et al., 2018). The average mercury content in the three studied frozen peatlands of Western Siberia increased with latitude from 47 to 109 ng/g (Lim et al., 2020). The mercury content in these peatlands turns out to be significantly higher than that in the frozen deposits of Spitsbergen. Moreover, this excess does not turn out to be proportional to the content of organic carbon. The content of the latter in the frozen peat bogs of Siberia is more than an order of magnitude higher than that in the permafrost of Spitsbergen, while the excess in mercury reaches a factor of 2.5 only.

The uniform distribution of mercury along the depth of the wells indicates the absence of patterns in its content depending on the facies environment of the sedimentation accumulation. Therefore, the nature of freezing does not affect the mercury content in sediments. According to previous radiocarbon dating of samples from the well 7 (Karaevskaya et al., 2021a), the sediments were formed during marine isotopic stages 1–2, while their age range exceeds 10 000 years. Consequently, the global climatic restructuring that occurred at the boundary of the Pleistocene and Holocene also did not affect the characteristics of mercury accumulation in the sediments of Spitsbergen.

Previous studies showed a correlation between mercury content and organic carbon in Arctic soils, bottom sediments, and permafrost (Kim et al., 2020; Rutkowski et al., 2021; Tarbier et al., 2021). In a number of studies, however, this correlation was not found (Olson et al., 2018). The lack of correlation between the mercury content and the amount of organic carbon in the case of the studied frozen deposits and STS in Spitsbergen is due possibly to the fact that, against the background of the low and consistent organic carbon contents characteristic of these rocks, there are some other unknown factors that neutralize this dependence. At the same time, analyses of the mercury content in bedrock showed an average value of 8 ng/g, which is significantly lower than that in Quaternary sediments. Thus, the main part of mercury entered the studied Quaternary sediments not from bedrock during weathering, but, probably, during sedimentation by forming complexes with organic matter or was adsorbed on clay particles.

Of interest is a comparative analysis of the mercury content in the studied frozen Quaternary sediments of marine origin and its content in current sea bottom soils of Spitsbergen according to literature data. In (Lebedeva et al., 2018), the mercury content was measured in five samples of bottom soils of Grönfjord Bay; a range of values was obtained from 7.1 to 42.3 ng/g. In other fjords of Spitsbergen, the average values of the mercury content in bottom soils were 52 ng/g in Horsund, 30 ng/g in Viidefjord, 16 ng/g in Diksonfjord (Kim et al., 2020), and 22 ng/g in Kongsfjord (Jiang et al., 2011). Only in Viidefjord was the mercury content higher than that in the studied frozen Quaternary sediments of marine origin. In all other fjords, including Grönfjord, current marine sediments are characterized by the lower mercury content than MIS 1-2 frozen preindustrial marine sediments.

Thus, if MIS 1-2 frozen marine sediments in the area of Barentsburg are characteristic representatives of preindustrial sediments for Spitsbergen, then the comparative analysis calls into question the effect declared in some studies of the increasing mercury content in current marine bottom soils of the archipelago that are caused by anthropogenic pollution and deglaciation.

CONCLUSIONS

The mercury content in 157 analyzed samples from permafrost Quaternary sediments in the area of Barentsburg (Spitsbergen) varies from 21 to 94 ng/g at an average value of 40 ng/g. The absence of significant differences in the mercury content depending on the geomorphological position of the wells and the uniform distribution over depth indicates the absence of patterns in its accumulation depending on the facial situation, the time of sediment formation, and the nature of freezing. There is no correlation between mercury content and organic carbon. At the same time, the average content of mercury in bedrock was 8 ng/g, which indicates the entry of mercury into frozen Quaternary sediments during sedimentation through the formation of complexes with organic matter or through absorption on clay particles.

A comparative analysis of the values we obtained for mercury content in frozen deposits of Spitsbergen near Barentsburg with previously published data showed the following. Similar mercury concentrations were obtained in Alaska when studying samples from soils and subsoil sediments with depths of up to 3 m. There was more mercury in the frozen sediments of Spitsbergen than in the permafrost of Yakutia. Preindustrial frozen sea sediments in the area of Barentsburg contains more mercury than current bottom soils of most of the fjords of Spitsbergen, including Grönfjord, on the shore of which Barentsburg is located.

ACKNOWLEDGMENTS

We express our gratitude to the RAE-Sh logistics center for supporting field work and to the staff of the RAE-Sh chemical analytical laboratory in Barentsburg, namely, N.N. Fateev, V.M. Mikhailova, and V.V. Boyko for determining the mercury content in the samples, as well as specialists from the Carbon and Nitrogen Laboratory of the Alfred Wegener Institute of the Center for Polar and Marine Research (Potsdam, Germany) for analyzing the organic carbon content.

FUNDING

This work was supported by ongoing institutional funding. No additional grants to carry out or direct this particular research were obtained.

CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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Translated by A. Ivanov

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