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MARINE CHEMISTRY

Hydrogen Sulfide Contamination of Coastal Lakes at Different Stages of Isolation from the White Sea

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Abstract—The results of studies of four coastal lakes with unique hydrological and hydrochemical structure at different stages of isolation from Kandalaksha Bay of the White Sea are reported for 2012–2019. All waterbodies are characterized by a permanent or seasonal stratified structure; as a result, bacterial sulfate reduction and hydrogen sulfide accumulation, toxic for aerobic life forms, become more active under anaerobic conditions. The highest amount of hydrogen sulfide was recorded in Lake Trekhtsvetnoe, which is at the meromictic stage of isolation.

Keywords: White Sea, isolation of waterbodies, anaerobic conditions, sulfate reduction, stratification, meromictic stage, hydrogen sulfide

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INTRODUCTION

The Karelian Coast of the White Sea is heavily indented and has many bays and lagoons. Such bays are often isolated from the sea by small rocky bars. Isostatic postglacial uplift of the coast at a rate of 3–4 mm/year results in isolation of small bays and lagoons from the sea [7]. This is a long-term process: with increasing isolation, sea bays pass the stage of lagoons with asymmetric tides and seasonal stratification; then, the stage of waterbodies with periodic inflow of seawater (most often during spring tides); then, the meromictic stage, which may proceed for several centuries and ends with their transformation into "ordinary" freshwater lakes [6]. During this time, significant changes take place in the hydrochemical and hydrobiological structure of such waterbodies: the influence of the sea and inflow of saline water decrease, the washing regime changes, the upper horizon of the waterbody is desalinated due to atmospheric precipitates and surface water from the land, and seasonal or permanent stratification is established. There is a change of aquatic organisms and microbiological communities. Oxygen of bottom water is completely consumed for oxidation of autochthonous and allochthonous organic matter under stratification conditions. The activity of sulfate-reducing bacteria intensifies under anaerobic conditions, which results in the occurrence and accumulation of hydrogen sulfide. Among the most famous water areas with a stratified structure and hydrogen sulfide contamination are the Black Sea [3], Lake Mogil'noe (Russia) [17], Lake Doroninskoe (Russia) [1], lakes Shira and Shunet (Russia) [16], Framvaren Fjord (Norway) [34], Lake Medusa (Palau) [26], as well as lakes in Canada (Lake Ogac) [32], France [25], Finland [27]. Such waterbodies can form under the natural conditions, as well as due to human economic activity, when small sea bays are isolated and their natural connection with the sea is disrupted (construction of dams, bridges, and roads).

Several such waterbodies occurring at different stages of natural isolation from the White Sea were discovered near the White Sea Biological Station, Department of Biology, Moscow State University (Karelian coast, Kandalaksha Bay of the White Sea). Since 2012 [7], they have been comprehensively monitored [30]. The investigated water areas represent a model for the study of short-term intrawater basin processes, without waiting hundreds of years for successive changes in stages. A specific feature of the surveyed waterbodies is the presence of hydrogen sulfide in bottom water, generated during sulfate reduction. This component is toxic, as well as a powerful reducing agent. It plays the role of a major regulator of redox conditions and influences the general direction and efficiency of processes occurring in the waterbody. The presence or absence of free hydrogen sulfide in water is controlled by the ratio of the intensity of sulfate reduction, on the one hand, and the presence of oxidants and elements capable of binding the newly formed hydrogen sulfide, on the other.

Our aim was to study the formation and evolution of hydrogen sulfide contamination in lake waters at

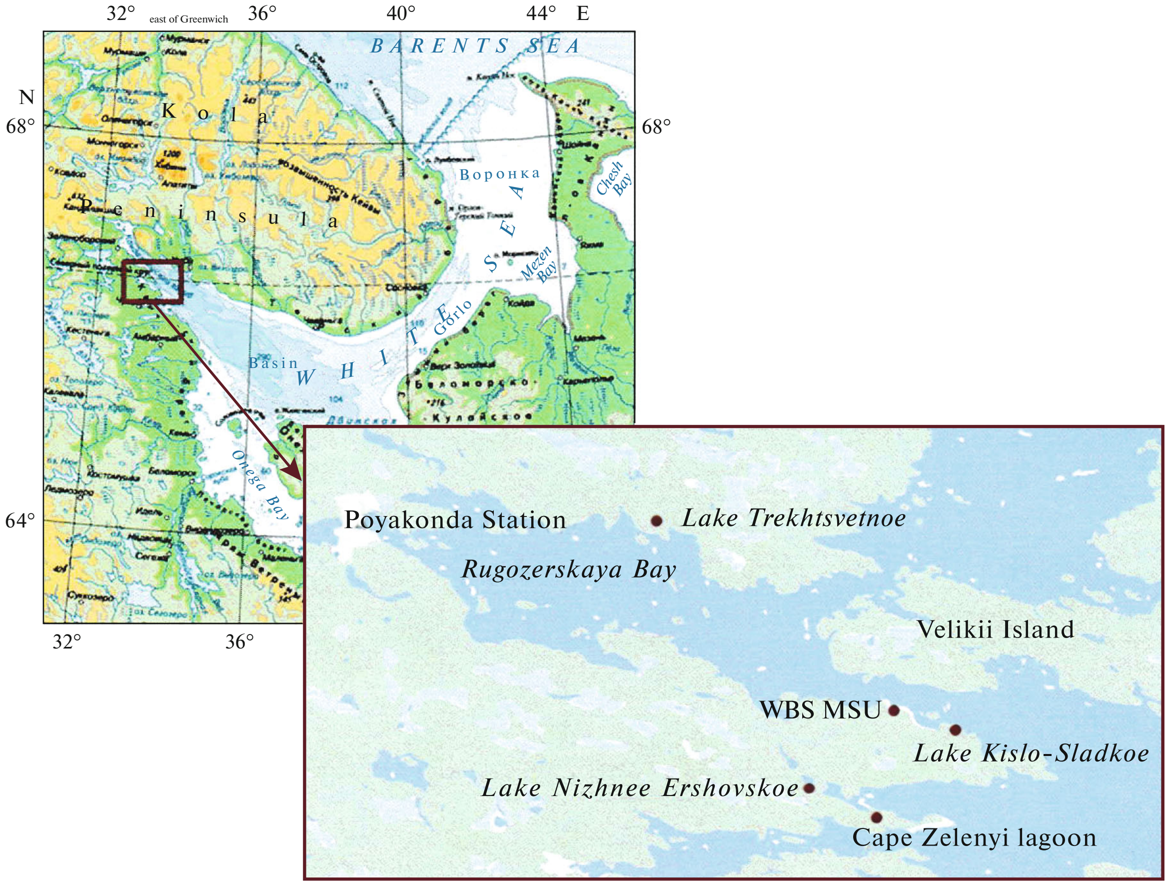

Fig. 1. Sketch map of lakes.

different stages of isolation from Kandalaksha Bay of the White Sea.

Four lakes were selected for the study; two of them (Nizhnee Ershovskoe and Trekhtsvetnoe) are the most isolated from the sea. In the third waterbody (Lake Kislo-Sladkoe), seawater is supplied when the high tide coincides only with a storm surge. The fourth waterbody is the so-called Cape Zelenyi Lagoon, which is at the initial stage of isolation. It is most affected by the sea (daily tides and ebbs). These waterbodies, among several others found in this area of the White Sea, are unique in their hydrochemical structure and attract the attention of scientists in different fields [30].

MATERIALS AND METHODS

A sketch map of the studied waterbodies is shown in Fig. 1. The coordinates of sampling stations and the main morphometric characteristics of lakes are reported in Table 1. Sampling was carried out at the deepest

Table 1. Morphometric parameters of studied water waterbodies

Parameter	Lake Nizhnee Ershovskoe 66°32.14′ N 33°03.74′ E	Lake Trekhtsvetnoe 66°35.53′ N 32°58.85′ E	Lake Kislo-Sladkoe 66°32.87′ N 33°08.14′ E	Cape Zelenyi lagoon 66°31.80′ N 33°05.65′ E
Waterbody area, km^2	0.081	0.032	0.016	0.012
Catchment area, km^2	1.23	0.64	0.16	0.19
Length, m	500	340	196	120
Width, m	245	135	147	120
Maximum depth, m	2.5	7.5	4.7	6.0
Average depth, m	1.0–1.5	2.5	1.0–1.5	2.0

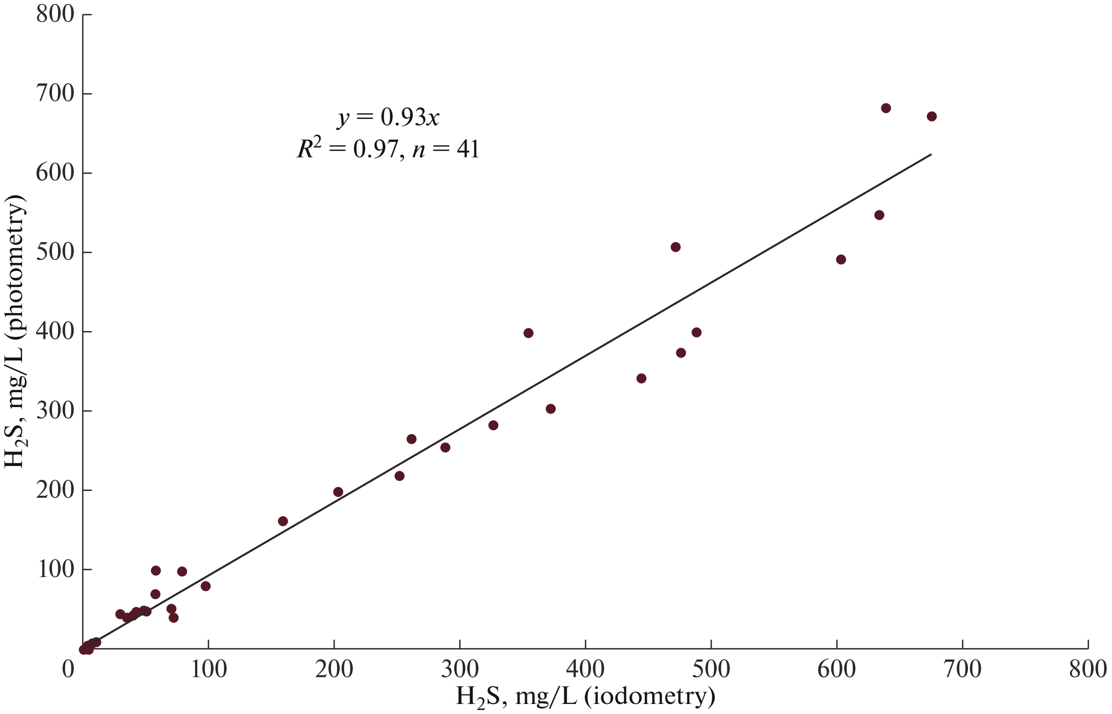

Fig. 2. Comparison of analyses of hydrogen sulfide concentration by iodometric (*X* axis) and photometric (*Y* axis) methods.

point of the water area of the lakes from different depth levels in different seasons using a silicone tube fixed on a calibrated cable, with a submersible Whale Premium Submersible Pump GP1352 (United States) or a horizontal polycarbonate sampling bottle (United States) with volumes of 2 and 5 L. To fix hydrogen sulfide (H_2S), zinc acetate and cadmium acetate solutions were used for photometric and iodometric analytical methods, respectively [4, 15, 18].

Since analysis of such samples is associated with methodological difficulties, it is necessary to confirm the correctness of the high values of the concentrations of sulfides obtained in analysis of the samples with a high (more than 100 mg/L) hydrogen sulfide content. For example, a high sulfide content (>10 mg/L) may partially or completely suppress the reaction of formation of a colored compound during photometric analysis, and dilution of samples after the appearance of color leads to a nonlinear change in optical density depending on the concentration. A solution to this problem was proposed in [4]: the authors suggested dilution of samples with a high hydrogen sulfide content directly during sampling for analysis of water samples from the hydrogen sulfide zone of the Black Sea.

To analyze the samples with a high hydrogen sulfide content, water samples with a volume of 0.2–5 mL, depending on the expected concentration, were taken by an automatic dispenser directly from the pump hose or a bottle and placed in glass cylinders with a solution of zinc acetate (for photometric analysis) or cadmium acetate (for iodometric analysis); then the volume of the sample was brought to 50 mL with distilled water. The samples diluted in this way were analyzed by the photometric and iodometric methods, as this was previously done for the analysis of water samples from the hydrogen sulfide zone of the Black Sea [4]. We carried out extra studies of some lake water samples by two methods in triplicate to estimate the reproducibility of the results when analyzing high hydrogen sulfide concentrations (Table 2). Analysis of the reported data shows that the reproducibility of the results for samples with a high hydrogen sulfide content by both methods can be estimated as acceptable.

In this study, we carried out 41 parallel hydrogen sulfide analyses using two methods in a wide concentration range (from 1.8 to 687 mg/L). Figure 2 shows the results. The scatter of the data on both sides of the trend line indicates that there is no systematic deviation of the results, and the high level of correlation ($R^2 = 0.97$) between them confirms the correctness of the concentration values obtained for a high hydrogen sulfide content in lake water.

The validity of such an approach (analysis by two methods and dilution of samples during sampling) was substantiated in March 2018, when the hydrogen sulfide content in samples diluted duing sampling was analyzed by the photometric method alone. For the bottom depths of Lake Trekhtsvetnoe, the data were overestimated by an average of 30% (900 mg/L) in comparison with the earlier and later results (~600 mg/L). We

Table 2. Hydrogen sulfide concentrations (mg/L) in samples of lake water analyzed by two methods (photometric and iodometric)

No.	Sample volume, mL	Method of analysis	Results of analysis	Mean	Standard deviation	Relative standard deviation, %
1	0.5	Iodometry	600 630 636	622	15	2.4
2	5.0	Iodometry	570 600 577	582	12	2.0
3	5.0	Iodometry	583 532 583	566	22	4.0
4	5.0	Iodometry	549 579 564	564	10	1.8
5	0.2	Photometry	687 676 673	679	6	0.8
6	0.5	Photometry	565 560 552	559	4	0.8
7	1.0	Photometry	221 257 242	240	13	5.2
8	1.0	Photometry	101 102 119	107	8	7.4

obtained a similar error in an analysis of waters with high hydrogen sulfide concentrations from other lakes. These data are not considered in the current study. Thus, we used both analytical methods with dilution of samples with distilled water during sampling in order to obtain an accurate data of analysis for the samples with high hydrogen sulfide concentrations.

We used an Expert 003 photometer (Russia) for photometric analysis of hydrogen sulfide. Water temperature and salinity were measured with a WTW Condi 315i conductometer (Germany); the oxygen content was measured during sampling with a MARK 302E oximeter (VZOR, Russia).

RESULTS AND DISCUSSION

Lake Nizhnee Ershovskoe. This is the lower of the two lakes formed at the place of an ancient strait, connected by a channel to the freshwater Lake Verkhnee Ershovskoe, from which it receives fresh water. This lake is isolated from the sea by a rocky bar, along which a fresh stream flows out of it. The inflow of a small amount of water from the sea occurs no more than once a year, during the strong spring tides and storms only. Most of the water column is fresh or slightly saline (0.1–0.7 PSU). Seawater entered it, as well as all other studied water waterbodies in autumn 2011, during a storm. The salinity increased up to 1.1 PSU in the surface layer of the lake and up to 15 PSU in the bottom layer. This returned the waterbody to an earlier stage of its isolation from the sea and set a new point of monitoring.

In March 2012, the first comprehensive studies of the waterbody were carried out. The water became saline up to 0.9 PSU at the surface (0.1 m), while at a depth of >1 m, the salinity increased sharply up to 16.4 PSU (Fig. 3). In March 2013, the salinity decreased slightly; it was 5.9 and 8.0 PSU at depths of 1.5 and 2.5 m, respectively. Fresh water predominated in other monitoring periods (from July 2014 to March

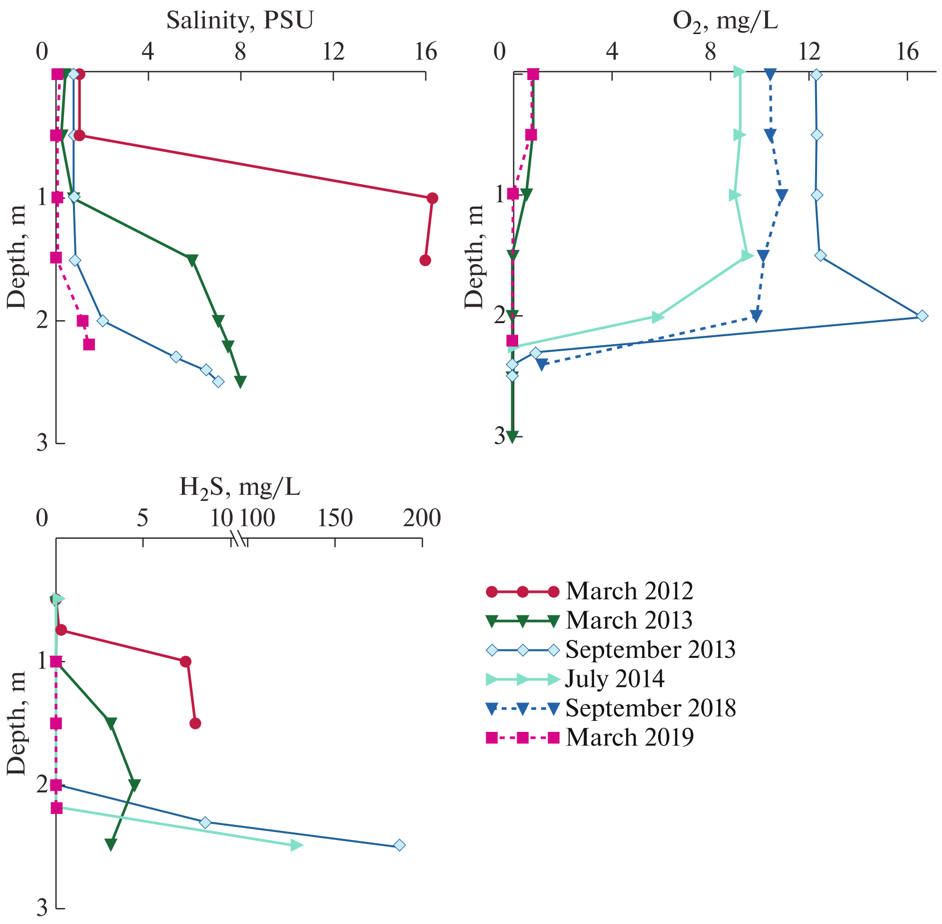

Fig. 3. Hydrochemical characteristics of Lake Nizhnee Ershovskoe.

2016), while brackish water (1.4–4.7 PSU) occurred in bottom lows only. In 2018, the lake became freshwater in its entire area.

The temperature in the lake varies along the water column depending on the climatic season. Since the depth of the lake is small (maximum 2.5 m), it warms to the bottom (up to 19°C) in summer [6]. In general, the oxygen content corresponds to the climatic season as well [6], but in all cases, it decreases almost to zero in periods with salinity stratification in the bottom water layer.

A layer of green water appeared in January 2013 in Lake Nizhnee Ershovskoe at the boundary of the aerobic and hydrogen sulfide water columns, the color of which is due to the presence of green sulfur bacteria [28, 29]. Extensive development of sulfur bacteria is controlled by hydrogen sulfide, which appeared in the bottom layer upon activation of sulfate reduction after the influx of seawater.

Sulfate reduction in the lake proceeds only in brackish bottom horizons, where the hydrogen sulfide concentration was 8.1 mg/L in March 2012, 4.5 mg/L in March 2013, and 9.0 mg/L in July 2013 (Fig. 3) [12]. The activation of sulfate reduction is associated with the creation of favorable conditions: the development of hypoxia in the lake, intake of sulfates, and presence of sufficient organic matter content. The highest hydrogen sulfide concentration in the bottom water mass in Lake Nizhnee Ershovskoe was detected in autumn. The maximum hydrogen sulfide content in 2013 was recorded in September (188 mg/L). In July 2014, the hydrogen sulfide layer, as well as in previous years, was thin at the bottom; its thickness did not exceed 20–30 cm, whereas the hydrogen sulfide content was very high (up to 130 mg/L). The persistence of such high hydrogen sulfide concentrations from September 2013 (in January 2014, this indicator was slightly less than 100 mg/L) suggests the presence of vertical stability of the water column at that time, despite its shallow depth. However, the hydrogen sulfide content decreased significantly (0.01–14.4 mg/L), often to trace amounts, from March 2018 to March 2019. This was probably due to the return of the lake to its original state, before the inflow of seawater (a freshwater waterbody completely isolated from the sea). Sulfate reduction in water can only take place during seasonal stagnation with a saline lens in a bottom low.

Lake Trekhtsvetnoe is considered a waterbody completely isolated from the sea, with a stable stratified structure and all meromictic signs [7]. The lake got its name for the different colors of its three layers: the upper freshwater layer is yellowish due to humic substances coming from swamp runoff. These are underlain by saline waters and include the bright green layer, then, hydrogen sulfide water with a dull yellow color. The green color is the result of extensive development of green sulfur bacteria [21, 29].

The features characteristic of meromictic water columns appeared in Lake Trekhtsvetnoye from the very beginning of research in 2012. The absence of water exchange between the surface depth layer (mixolimnion) and bottom layer under permanent stratification conditions leads to the formation of a chemocline with a sharp change in redox conditions, as well as a static anaerobic zone (monimolimnion). Such conditions make meromictic waterbodies unique for studying the geochemistry and microbiology of anaerobic processes. A fresh mixolimnion is found to a depth of 1 m in Lake Trekhtsvetnoye, while a stagnant saline hydrogen sulfide water column (monimolimnion) starts at a depth of 1.5–1.8 m. There is a narrow pycnocline with sharp physicochemical gradients between them. A layer of green water with a thickness of 15–20 cm is located directly above the chemocline. Microbiological studies at the boundary of the aerobic and anaerobic zones showed an abundance of anoxygenic phototrophic bacteria, which are the green sulfur bacteria responsible for the bright green color of the water [13, 33].

Saline water penetrated into Lake Trekhtsvetnoe, as well as into other studied waterbodies, during the spring tide, which coincided with a strong wind surge in November 2011. This resulted in salinization of the mixolimnion and most likely slightly increased the salinity of underlying waters [6]. Its meromictic structure was not disturbed, despite the infiltration of hydrogen sulfide waters of the monimolimnion into the surface layers. Since there were no earlier observations, it is difficult to say to what extent the hydrogen sulfide concentrations recorded in March 2012 in the monimolimnion are a consequence of dilution of deep lake waters with fresh seawater, and what the hydrogen sulfide content was in lake waters earlier. The surface layer was cleaned of hydrogen sulfide by September 2012, but its content in anaerobic deep (below 4 meters) water continued to increase over the next several years and became relatively stable only by 2016 (Fig. 4). Since then, the hydrogen sulfide concentration at bottom depths has remained at a level of ~600 mg/L [11, 12, 31], which significantly exceeds its content in such well-known waterbodies with anaerobic conditions as the Black Sea (12.3 mg/L) [3], Lake Mogil'noe (160 mg/L) [17], and Framvaren Fjord, Norway (200 mg/L) [34].

At present, the waters of the mixolimnion are completely fresh; below the chemocline, oxygen is completely absent and the waters of the monimolimnion are contaminated with hydrogen sulfide. The temperature and salinity are constant at a depth below 4 m and reach 6–7 °C and ~22 PSU, respectively, at any time of the year. Thus, we have a classic example of a meromictic waterbody (Fig. 4).

The accumulation of hydrogen sulfide in the anaerobic layer of Lake Trekhtsvetnoe occurs against a decrease in sulfate content: there is a decrease in the SO_4/Cl ratio with depth [2].

Lake Kislo-Sladkoe was formed as a result of isolation of the sea bay from the sea area by an island with two rocky shallows on the sides. As a result of land uplift, one of the shoals turned into a bridge, now overgrown with meadow grasses, and the other is a rocky sill through which water from the sea enters the lake during spring tides and surges. The rest of the time, the current is directed from lake to. Fresh water can enter the waterbody with snowmelt and runoff from a small freshwater stream in summer only [6]. In 2001 and 2002 Shaporenko et al. [22–24] described the three-layer structure of this lake: (1) the desalinated upper layer to a depth of 1 m with a salinity of 13 PSU (epilimnion); (2) the layer of water saturated with oxygen with a temperature higher than that of the surface layer to a depth of 3.2–3.75 m (hypolimnion); (3) the bottom anaerobic layer with a high sulfide content.

Desalination of the water column, as well as wind mixing, affect only the upper water layer to a depth of 1 m. It separates underlying more saline saltier from contact with the atmosphere, which results in the greenhouse effect and appearance of the phenomenon of a deepened temperature maximum in summer, which is typical of heliothermal waterbodies: the temperature of the subsurface layer is slightly higher than that of the overlying one. Despite the low depth of the waterbody (~4.5 m), the temperature of the bottom layers does not exceed 11°C even in the summer months, although the surface layers can warm up to 18–28°C [6]. Among the hydrochemical patterns of the lake, we can note the uniquely high concentrations of dissolved oxygen in the middle layer reaching 26–28 mg/L (200–300% saturation) at some stations. This is explained by algal bloom and the presence of a pycnocline, which prevents subsurface waters from mixing with overlying layers [22].

In summer, a pink layer may appear in the lake at the boundary of the aerobic and anaerobic layers (redox zone), due to the extensive development of Cryptophyceae *Rodomonas* sp. [9, 10, 14].

Long-term monitoring on Lake Kislo-Sladkoe showed that the processes described above, as well as vertical stratification and three-layer structure, are characteristic of only the summer period. The waterbody can be flushed during high autumn surges and spring tides, which was observed several times, and it became homogeneous throughout the entire water column. However, vertical stratification is restored in spring, after ice and snow melt, which results in stag-

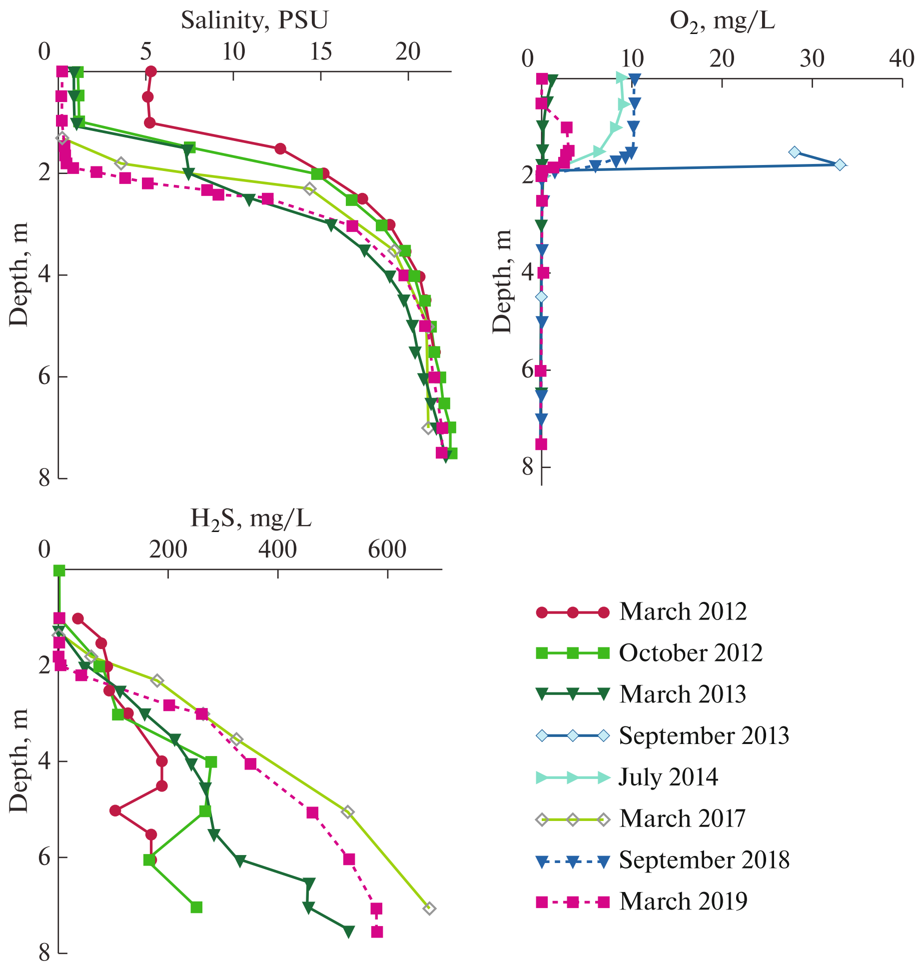

Fig. 4. Hydrochemical characteristics of Lake Trekhtsvetnoe.

nation in the bottom layers and the formation of hydrogen sulfide contamination.

A large volume of seawater entered the lake, and the salinity at the surface reached 26.2 PSU in March 2012 as a result of a strong surge in autumn 2011 (Fig. 5). Subsequently, the surface layer was freshened due to precipitatation and snowmelt (the minimum salinity of ~6 PSU was recorded in March 2013). In September 2013, the salinity gradient was low (21.9 PSU at the surface and 23.8 PSU at the bottom), which is most likely associated with seasonal seawater floods during high autumn tides. In July 2014, the indicators returned to the January 2013 values and the salinity of surface waters was 13 PSU. In March 2019, the highest salinity values were recorded throughout the entire depth of the lake (~30 PSU), which may indicate winter flushing of the waterbody with saline and cold water, which most likely came from the central parts of the White Sea.

In March 2012, a small hydrogen sulfide content (28.3 μg/L) was recorded only in the bottom layer of the lake after the first autumn flushing, which is significantly lower than the values obtained earlier for this waterbody by Shaporenko et al. (5.6–10.3 mg/L) [22–24]. However, in October 2012, hydrogen sulfide was detected in the lake throughout the entire water column with a maximum content of 18.9 mg/L in the bottom layer (Fig. 5). A similar distribution persisted in the lake during the subsequent cold period (January and March 2013). In autumn and summer 2013 and 2014, hydrogen sulfide was detected only in the deep water area (below 2.5 m). However, whereas in the summer months its content below this horizon was 2.3–16.7 mg/L, in September it became many times higher, reaching 101 mg/L. Such a high hydrogen sulfide content in autumn is most likely associated with the development of favorable conditions for sulfate reduction. These include combination of anaerobic conditions during seasonal stagnation with the accumulation of organic matter in bottom saline waters as a result of withering away of the biomass formed in the growth season. In March 2019, the hydrogen sulfide content in the deep layers of the lake was minimal (4.3 μg/L) due to the influx of oxygen-bearing seawater with negative temperatures and, consequently, attenuation of sulfate reduction.

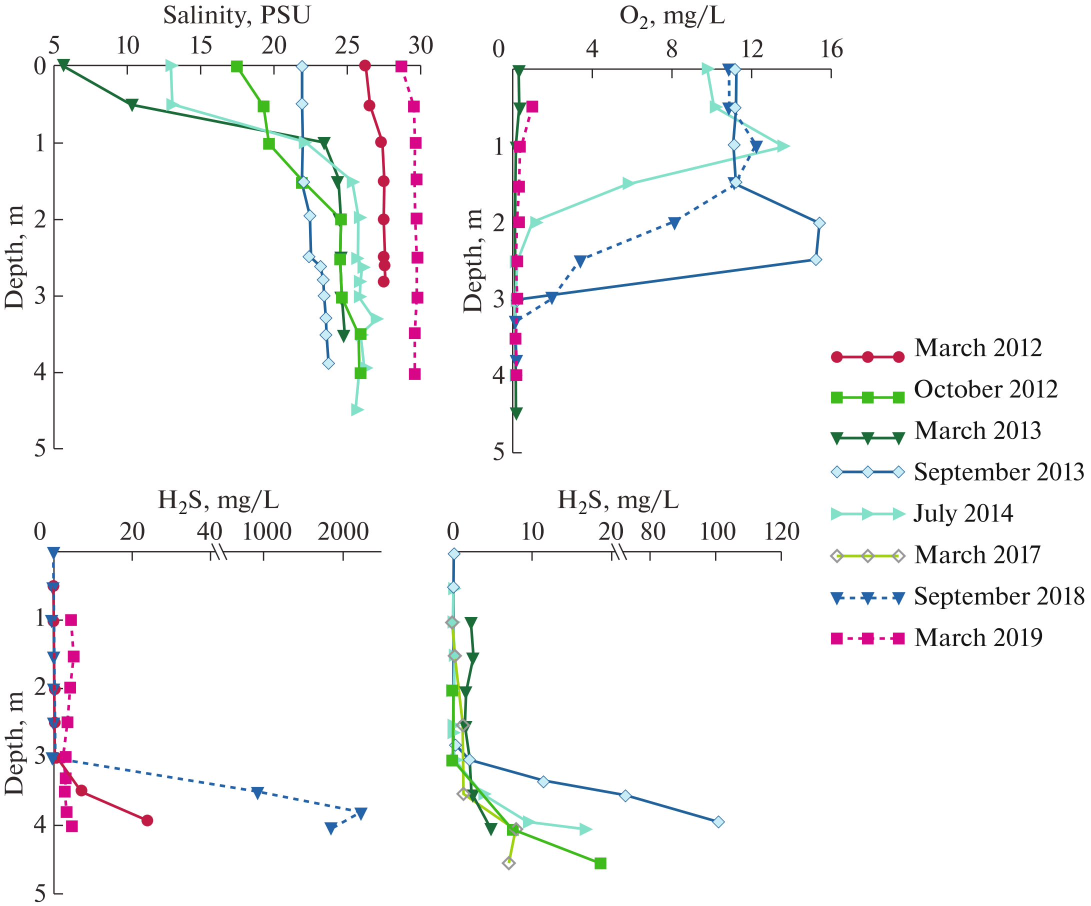

Fig. 5. Hydrochemical characteristics of Lake Kislo-Sladkoe.

This waterbody differs from the other three in the greatest instability of all hydrochemical characteristics, the values of which were controlled by the climatic season and degree of flushing of the lake with seawater during high tides. The hydrogen sulfide content in the bottom layers of the waterbody varied widely depending on the season and predominant hydrological conditions. The lake is characterized by periodic stratification and hydrogen sulfide accumulation in the bottom layers during the stagnation period.

Cape Zelenyi Lagoon is located at the base of the Cape Zelenyi Peninsula and is connected to Kislaya Bay by a small sill, over which seawater enters at each tide; the surface layer of water flows from lake to sea at low tide. The lake was formed at the place of an ancient strait, which isolated an island from the mainland, later turned into the Cape Zelenyi Peninsula [6, 8].

The seasonal vertical stratification in the lagoon, as well as in Lake Kislo-Sladkoe, depends on autumn flushing of the waterbody. The water in the lagoon is saline througout the water column. Slight desalination of the upper layer occurs only after the ice melts, but much less than in other studied water waterbodies. The salinity in the near-bottom horizons is higher than that in the neighboring sea area. This may be due to the very small drainage basin of this waterbody, as well as to the ice salting out upon seawater freezing [5]. Despite the slight difference in the density of saline water on the surface and at the bottom, the greenhouse effect is recorded in the warm season in the Cape Zelenyi lagoon, as well as in Lake Kislo-Sladkoe. The lower horizons keep constant temperature and salinity in summer. The stratified structure of the waterbody may persist for many years, if the lagoon is not flushed in autumn or winter.

In summer, there is a layer of pink water at the boundary of the aerobic and anaerobic zones in the Cape Zelenyi Lagoon, as well as in Lake Kislo-Sladkoe, which is due to the development of Cryptophyceae *Rodomonas* sp. [9, 10, 21, 28]. It appears only in summer and occurs upon prolonged stagnation. In winter, anaerobic brown-colored green sulfur bacteria, rather than algae, give a reddish tint to the water [9].

After the autumn flushing of 2011, the hydrogen sulfide content did not exceed 200 μg/L in March 2012, even in the bottom water [31], which is significantly lower than the concentrations recorded by

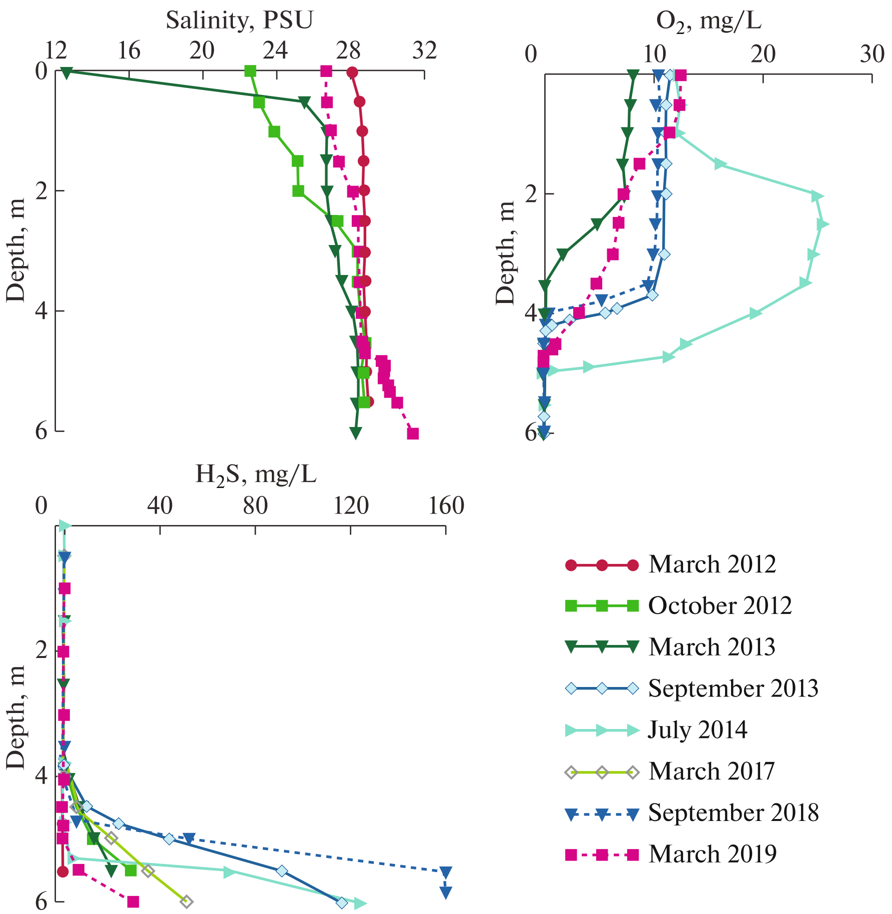

Fig. 6. Hydrochemical characteristics of Cape Zelenyi lagoon.

Shaporenko with coauthors in summer of 2001–2002. [22]. The following autumn (October 2012), its content in the bottom layers increased to 12.6 mg/L at a depth of 5 m and to 28.7 mg/L in the bottom waters (5.5 m), which is three times lower than the previously recorded maximum (90 mg/L) (Fig. 6). The amount of hydrogen sulfide increased up to 117 mg/L by autumn of 2013, and up to 125 mg/L in July 2014. In March of 2016 2017, and 2018, the maximum hydrogen sulfide content in the bottom layers was estimated at the level of 50 mg/L. The highest H_2S concentration of ~160 mg/L was recorded for this waterbody in September 2018; the value of only 29 mg/L was recorded in March 2019. According to Shaporenko [22], the high hydrogen sulfide concentrations characteristic of the bottom waters of this waterbody are caused by the extensive development of filamentous algae in the lagoon, which, dying off, accumulate at the bottom and become a substrate for sulfate-reducing bacteria. In general, an increase in the bottom hydrogen sulfide concentration during the growing season of the year, with maximum values reached in autumn, is typical of the Cape Zelenyi Lagoon, as well as of Lake Kislo-Sladkoe. The winter vertical stratification in this area depends on the height of the autumn spring tides and on the permeability of the ice barrier at the bar, which affect the degree of flushing of the waterbody and, as a consequence, the distribution of hydrogen sulfide.

CONCLUSIONS

Study of four lakes at different stages of isolation from Kandalaksha Bay of the White Sea shows that all of them have a unique hydrological and hydrochemical structure, which is formed at a certain stage of their evolution. After a storm discharge of water from the sea at the beginning of winter 2011–2012, the lakes returned to the earlier stages of isolation, setting a new starting point for monitoring of the restoration of stratification and the formation of hydrogen sulfide contamination in them.

Hydrogen sulfide contamination in the shallow freshwater Lake Nizhnee Ershovskoe developed after the overflow of sulfate-bearing seawaters. The hydrogen sulfide concentration in the bottom salty lens reached 130–190 mg/L. After the lake became fresh again, the concentration decreased and the current hydrogen sulfide content in the bottom horizons is comparable to that in similar lakes with a significant influx of bog waters [19, 20].

Meromictic Lake Trekhtsvetnoe is characterized by the highest hydrogen sulfide content among all investigated waterbodies. This content reached 500–680 mg/L in the last few years, which is significantly higher than the hydrogen sulfide concentration in other known water waterbodies with a euxinic zone.

The appearance and accumulation of hydrogen sulfide in Lake Kislo-Sladkoye and in Cape Zelenyi Lagoon is due to vertical stratification, which depends on the degree of their flushing with seawater. Both lakes are characterized by increased bottom hydrogen sulfide concentration during the growing season, with maximum values in autumn. The hydrogen sulfide content in Cape Zelenyi Lagoon (up to 120–160 mg/L) is higher than in Lake Kislo-Sladkoe (≤20 mg/L), which is most likely due to the extensive development of filamentous algae, which, dying off, accumulate at the bottom and become a substrate for sulfate-reducing bacteria.

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