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Original article

Vertical Distribution of Oxygen and Hydrogen Sulfide in the Deep Part of the Black Sea Based on the 2017–2019 Expedition Data

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Abstract
Purpose. The purpose of the work is to analyze the features of vertical distribution of the dissolved oxygen and hydrogen sulfide in the deep part of the Black Sea in the modern period.
Methods and results. The data obtained in 11 expeditions of Marine Hydrophysical Institute (MHI) of RAS in the Black Sea within the economic zone of Russia in 2017–2019 were used. These surveys included more than 200 deep-sea stations at which the hydrochemical samples were taken at specific isopycnal surfaces by means of a cassette of 12 bathometers of the Sea-Bird 911 plus CTD Seabird-Electronics INC device; as a rule, it was a series of σ_t values equal to 16.30; 16.20; 16.10; 16.00; 15.95; 15.90; 15.80; 15.60; 15.40; 15.20; 15.00 and 14.60 kg/m^3. Such a scheme made it possible to determine both general vertical distribution of oxygen in the oxycline and depth of hydrogen sulfide occurrence with an accuracy of up to 0.05 kg/m^3 in the conventional density scale.
Conclusions. All surveys showed a decrease in oxygen content with depth (and occurrence of oxycline, respectively) starting below isopycnal surface σ_t = 14.5 kg/m^3. The position of sub-oxygen zone upper boundary defined by the isooxygen 10 μM was not strictly isopycnal, but fell on the range of isopycns σ_t = 15.7–15.85 kg/m^3. However, it was not possible to identify a relation between the change in the position of the upper boundary and a certain hydrological season. For example, the deepest occurrence of the upper boundary below σ_t = 15.8 kg/m^3 was observed both in November and December 2017 and August 2018. The lowering of isooxygen 10 μM to σ_t = 15.9 kg/m^3 in the Kerch shelf area is related to a more voluminous and colder intermediate layer above the shelf in December 2017. Only one of 11 surveys showed the position of hydrogen sulfide upper boundary determined by isosulfide 3 μM raised almost to σ_t = 16.0 kg/m^3 in April 2017. In all other cases (including the one in August 2017, i.e. six months after its rising), it was invariably found within range σ_t = 16.10–16.15 kg/m^3. The concentration of hydrogen sulfide at the depths of 1750–2000 m has remained unchanged at level 383 ± 2 μM over the past 25 years.

Keywords: Black Sea, vertical distribution of oxygen and hydrogen sulfide, sub-oxygen zone, field data

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Introduction

The Black Sea is the largest euxinic basin in the world where hydrogen sulfide occurs at a depth of 90–150 m depending on the area (the greatest contrast in the depths of its occurrence is represented by the centers of cyclonic and anticyclonic gyres, shelf edge and continental slope) [1, 2]. The main cause for hydrogen sulfide formation is weakened vertical exchange when oxygen-containing surface water can penetrate downward only to a certain depth.

The vertical exchange weakening is due to the constant Black Sea stratification by salinity and density at which a constant halocline among isohalines of 18.5–21.5 [3, 4] and a pycnocline among isopycnals of 14.5–16.5 kg/m^3 take place, as well as a cold intermediate layer (CIL) with a temperature of < 8 °C and a core located at isopycnal surface depth σ_t = 14.5 kg/m^3 [4]. This structure makes it possible for oxygenated surface waters to sink only to a certain depth during winter aeration. As a rule, winter aeration reached isopycnal surface depths within the range of 14.2–14.5 kg/m^3 in the last decades of the past century [5, 6]. Recent years showed the gradual decrease of the penetration depth to σ_t = 14.2–14.3 kg/m^3 due to warm winters [7, 8].

In such a situation, below the isopycn σ_t = 14.5 kg/m^3, only CIL waters become the source of oxygen required for the oxidation of the particular organic matter (POM) descending from the overlying waters. This results in the oxygen content gradual decrease with the depth and occurrence of the oxycline which gradually moves to the so-called sub-oxygen zone. Its upper boundary is defined by the isooxygen 10 μM (or 20 μM according to some researchers) [9] and its lower boundary is the isopycnal surface at which oxygen disappears (i.e. its content becomes less than a defined minimum of 3 μM). In fact, the lower boundary is the upper one of the hydrogen sulfide occurrence, below which sulfates become the POM oxidizer instead of dissolved oxygen. The reduction of sulfates results in the hydrogen sulfide formation.

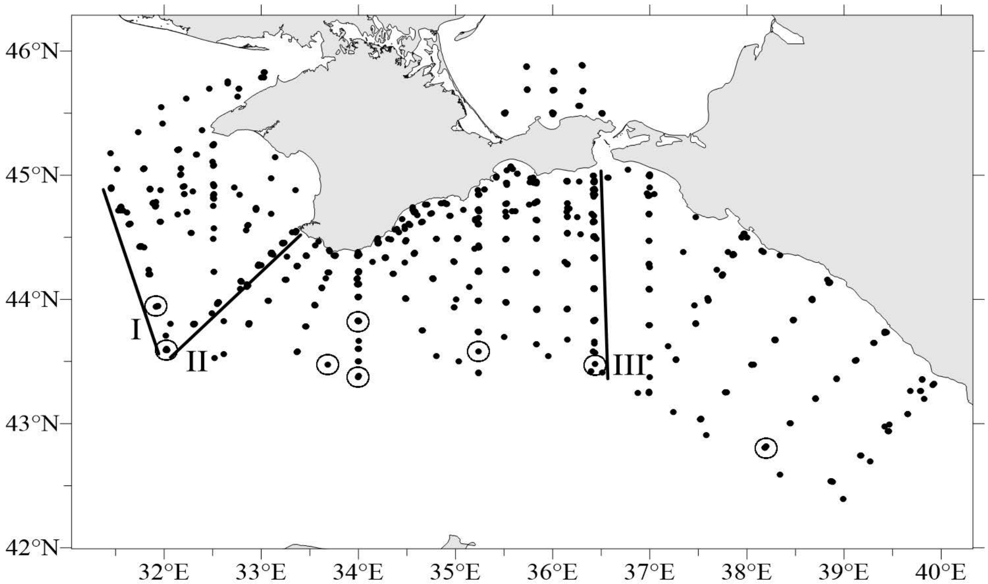
One of the most important tasks of the Black Sea expeditionary research in the modern period is monitoring the position of the upper boundary of hydrogen sulfide zone where the oxidizer for the POM decomposition process changes when the sulfate anion becomes an acceptor of electrons instead of dissolved oxygen. The presence of oxygen in the Black Sea surface waters provides the usual trophic chain typical for any ocean: phytoplankton formed during the photosynthesis from inorganic (and also organic) forms of nutrients serves as food for zooplankton which in turn provides food for larger marine organisms. Only certain types of bacteria can exist in the hydrogen sulfide zone with no oxygen. A shift in the boundary of the hydrogen sulfide zone, namely its rise to the surface, threatens the life of surface water organisms.

Since November 2015, MHI has carried out systematic comprehensive expeditionary studies of the Black Sea within the economic zone of the Russian Federation, in particular, monitoring the lower boundary of the sub-oxygen zone. The results obtained made it possible to compare these data with the results of individual short-term MHI expeditions of past years [10] and start a detailed analysis of the state of this zone. The features of its upper and lower boundaries in 2015–2016 [11], as well as the causes for the rise of the hydrogen sulfide zone in certain areas of the Black Sea northwestern shelf (NWS) [12], were considered.

This work, which continues the research of [10–12], is purposed at studying the features of oxygen and hydrogen sulfide vertical distribution in the water column of the Black Sea based on the data from 11 expeditions of R/V *Professor Vodyanitsky* in 2017–2019.

Materials and methods

Fig. 1 shows the location of all hydrochemical stations in 11 cruises in 2017–2019 during which samples for oxygen and hydrogen sulfide content were taken. Deep-sea hydrochemical samples were taken at specific isopycnal surfaces by means of a cassette of 12 bathometers of the Sea-Bird 911 plus CTD Seabird-Electronics INC device. As a rule, it was a series of σ_t values equal to 16.30; 16.20; 16.10; 16.00; 15.95; 15.90; 15.80; 15.60; 15.40; 15.20; 15.00 and 14.60 kg/m^3. At 8 stations, sampling for hydrogen sulfide and oxygen was carried out by three probings at 37 horizons: in the first two probings, the samples were taken by depth, from a maximum immersion of 2000 m (for technical reasons the probe was not immersed deeper), then through each 50 m to 1800 m depth and further – throught each 100 m. In the third probing, samples were taken by the series of isopycns given above and from the surface with a plastic jar.



F i g. 1. Map of the deep-sea stations at which the samples for oxygen and hydrogen sulfide were taken in 2017–2019. Solid lines indicate sections I-III which were carried out in each cruise if possible. Circles indicate the stations where samples were taken at 37 horizons.

After constructing averaged vertical oxygen profiles obtained using the Surfer or Grafer programs, such a sampling scheme made it possible to determine the upper boundaries of the sub-oxygen layer and the hydrogen sulfide zone, as well as the thickness of the sub-oxygen zone located between them, with a resolution of up to 0.05 kg/m^3 σ_t which corresponds to an accuracy of ~ 5 m in the depth scale.

We applied an oxygen sensor in addition to sampling in order to obtain vertical oxygen profiles in six expeditions (given below). The readings of the sensor for each station were calibrated using three samples taken at the surface, in the middle of the oxycline (σ_t = 15.0 kg/m^3) and in the beginning of the sub-oxygen zone (σ_t = 15.8 kg/m^3).

The hydrogen sulfide content was found by the iodometric method according to the technique [1] with iodine consumption at isopycn σ_t = 15.8 kg/m^3 taken as zero. The error in determining hydrogen sulfide is estimated at ±3 μM. Oxygen concentration was found by the Winkler method modified in terms of sampling with low oxygen content in accordance with the method [13] which provides the results with an accuracy of ±0.01 ml/L [14]. In both cases, 200 ml volumetric flasks and narrow-necked oxygen bottles were purged with argon 15 min before sampling.

Further, the average vertical profiles of distribution of oxygen and hydrogen sulfide concentrations relative to the conventional density scale were calculated with a discreteness of 0.1 kg/m^3 in conventional density range σ_t = 13.0 – 17.3 kg/m^3 for each expedition. Averaging was carried out using the inverse distance method followed by additional smoothing using the low pass filtering method [2].

Discussion of results

Vertical profiles of oxygen and hydrogen sulfide. Based on the vertical oxygen profiles obtained by averaging field data from six expeditions (dashed lines in Fig. 2), one can get the impression that in all cases the subsurface maximum of oxygen was observed and the beginning of oxycline (i.e. a noticeable decrease in oxygen concentration with depth) rose to the occurrence depth of isopycnal surface σ_t = 14.0 kg/m^3. However, when analyzing the oxygen sensor data (solid lines in Fig. 2), it becomes clear that the subsurface maximum of oxygen was observed only in April 2017 and the beginning of oxycline was still located near isopycnal surface σ_t = 14.5 kg/m^3 and was not subject to any seasonal changes since all four surveys in 2017 (April, June, November, December) showed that it remained at the same level.

It should also be noted that all profiles coincide below isopycn σ_t = 15.0 kg/m^3, when the oxygen content becomes < 150 μM. Its concentration below the indicated isopycn was higher than in other surveys by a value exceeding the determination error, which is estimated at approximately 1% [13] within the range of 30–150 μM, only in November 2017.

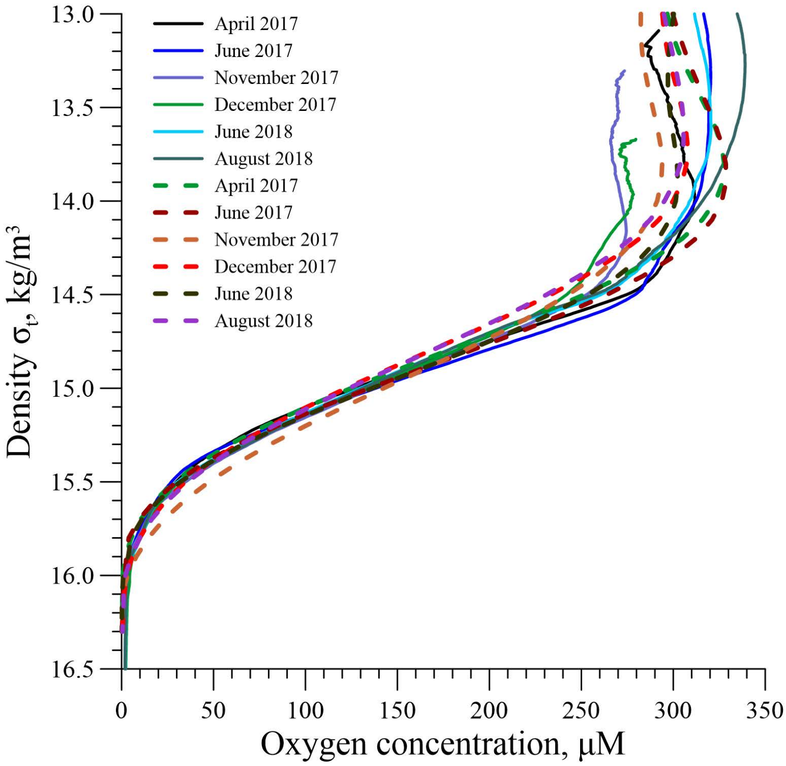
[1] Bordovsky, O.K. and Ivanenkov, V.N., eds., 1978. *Methods for Hydrochemical Studies in the Ocean*. Moscow: Nauka, 271 p. (in Russian).

[2] Masevich, A.V., 2022. *Dynamics of Oxygen in the Main Pycnocline of the Black Sea.* Thesis Cand. Geogr. Sci. Sevastopol, 151 p. (in Russian).

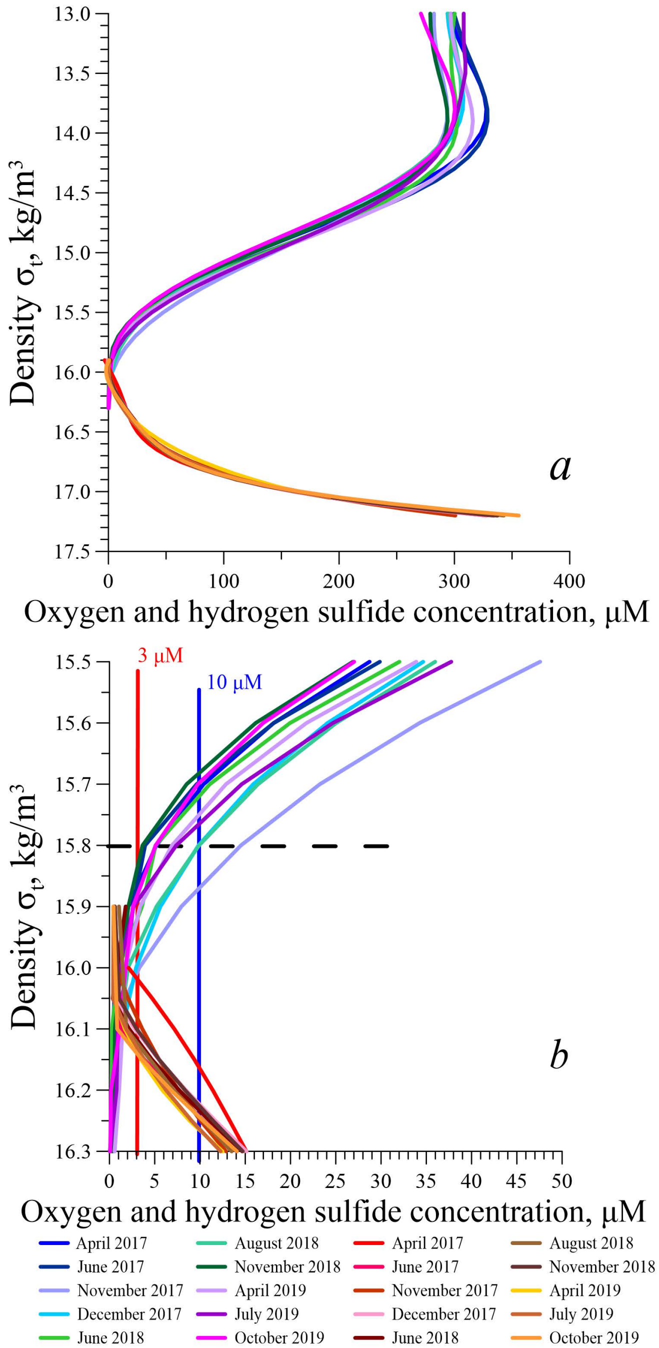
Here, we should take into account that the conditions for the 2009–2019 CIL maximum formation were observed in 2017 [15]. Therefore, the relative decrease in temperature and increase in oxygen concentration in the CIL are observed in 2017 compared to previous and subsequent years [16]. This increase could cause a short-term oxygen concentration rise below isopycn σ_t = 15.0 kg/m^3 in November 2017 (in December it disappeared).

Fig. 3 shows the density scales of oxygen and hydrogen sulfide vertical profiles averaged over field data recorded in 11 MHI expeditions.

Practical merging of vertical oxygen profiles (Fig. 3, *a*) with a more detailed analysis (Fig. 3, *b*) shows that the sub-oxygen zone upper boundary determined by an oxygen concentration of 10 μM varies over time from 15.7 kg/m^3 to 15.9 kg/m^3, which corresponds to an interval of 40 m in the depth scale. The impossibility to identify any dependence of the upper boundary position on the season is not surprising since the time for updating the characteristics in this water layer exceeds the season significantly [17, 18]. The latter is indirectly confirmed by the fact that two out of three cases when the upper boundary of the sub-oxygen zone was located below σ_t = 15.8 kg/m^3 occurred in winter months (November and December 2017), but a similar deepening was also observed in August 2018.



F i g. 2. Vertical profiles of oxygen in the density scale for six surveys obtained by the averaged Winkler method (dashed lines) and oxygen sensor data (solid lines)

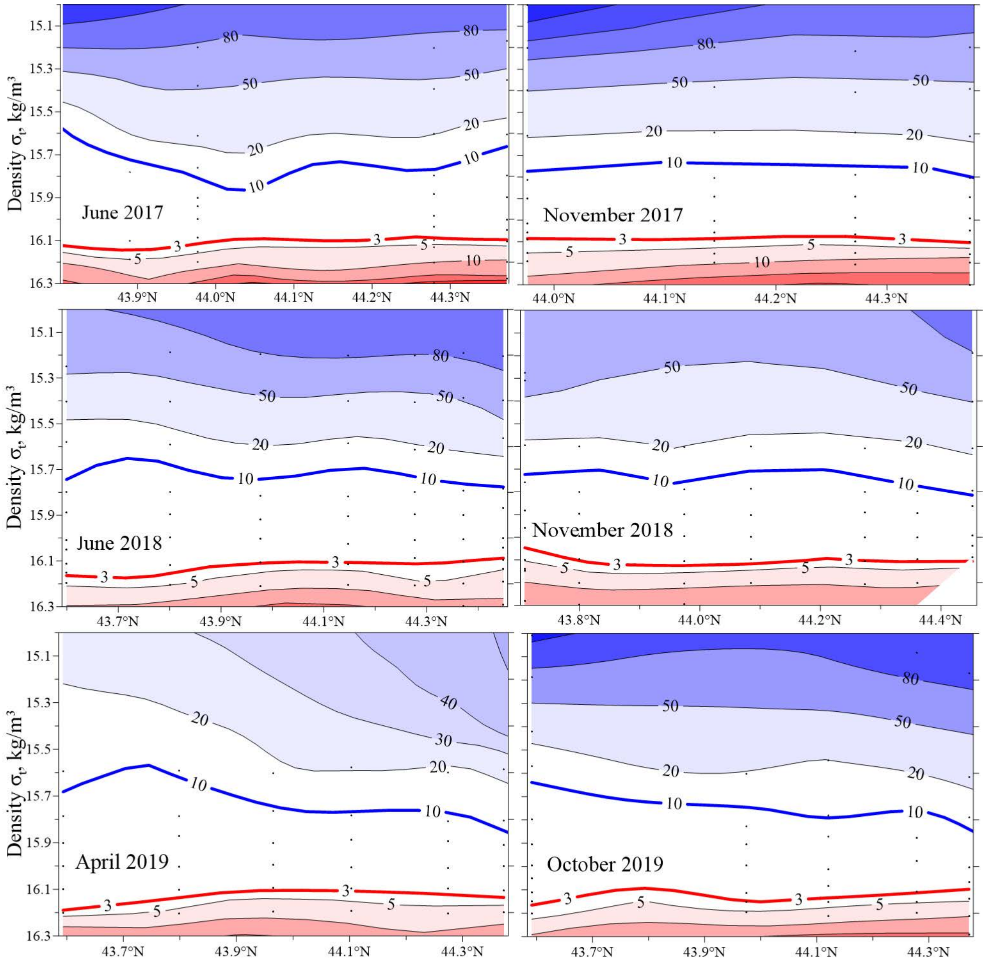


F i g. 3. Vertical profiles of oxygen and hydrogen sulfide in the density scale over the entire thickness (*a*) and in interval σ_t = 15.5–16.3 kg/m^3 (*b*) in the Black Sea in 2017–2019

The lowest position of the sub-oxygen zone upper boundary at σ_t = 15.87 kg/m^3 (Fig. 3, *b*) was observed in November 2017, the year of the maximum CIL formation over the past 10 years [13].

The vertical distribution of hydrogen sulfide turned out to be more isopycnal as isosulfide 3 μM was located in the isopycnal range of 16.10–16.15 kg/m^3 during 10 expeditions and only the survey performed in April 2017 showed its noticeable rise to almost σ_t = 16.0 kg/m^3.

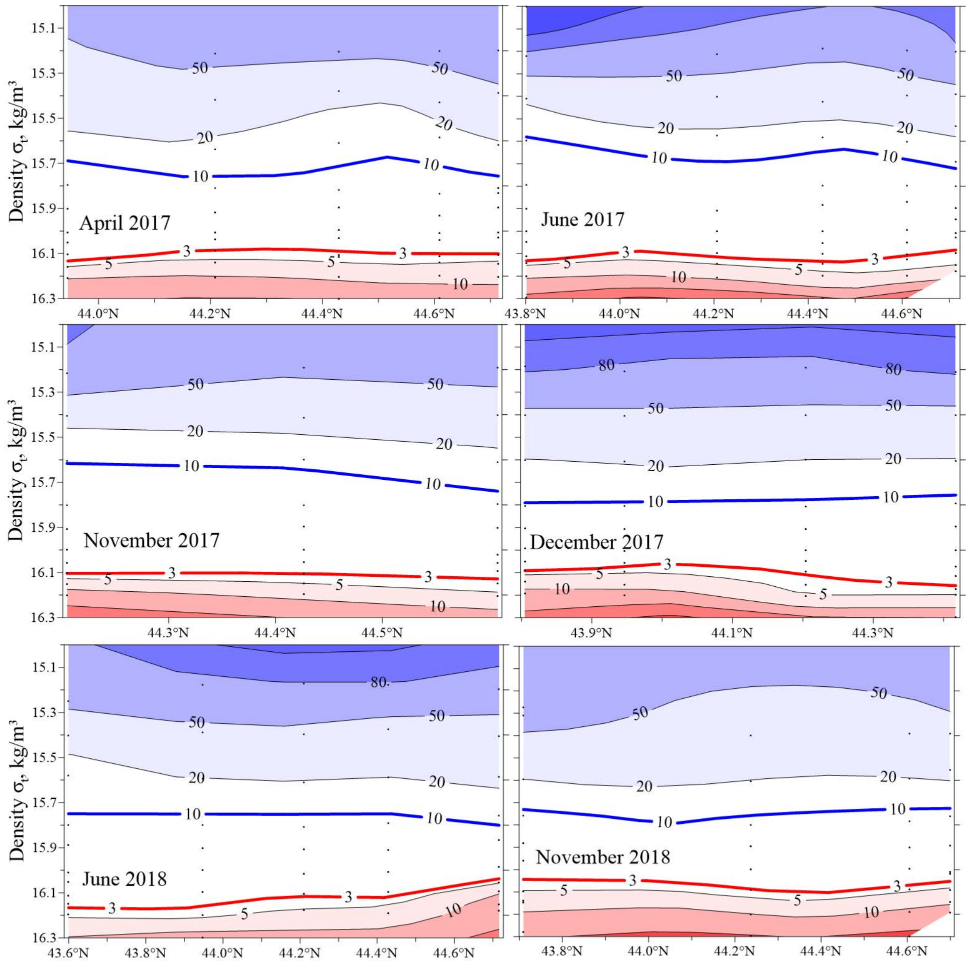
Position of the sub-oxygen zone at the sections. As a rule, all surveys conducted in 2017–2019 (except for August 2018) included “century” section II (Fig. 1) along the Cape Chersonesos – the Bosporus Strait line since it was historically regularly studied, even in 1995–2015, when expeditionary research of the Black Sea deep parts was performed extremely rarely [10].



F i g. 4. Position of sub-oxygen zone at "century" section II Cape Chersonesos – the Bosphorus Strait in 2017–2019

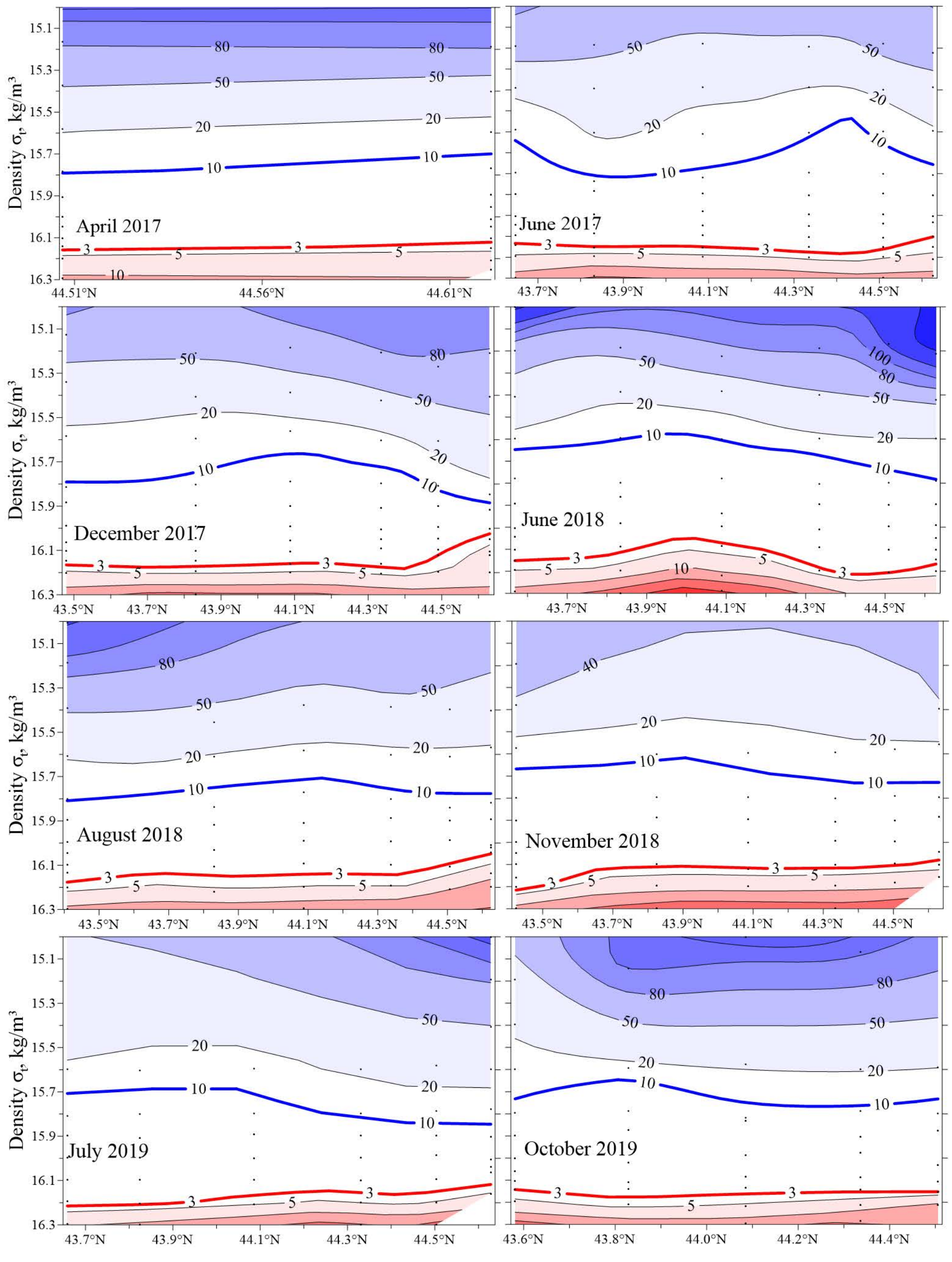
Fig. 4 shows that isosulfide 3 μM was constantly located in the isopycnal range of 16.10–16.15 kg/m^3. Against the background of this homogeneity, isooxygen 10 μM changed its position in a larger range of the conventional density scale (15.6–15.85 kg/m^3) while separate surveys revealed that the sub-oxygen zone upper boundary could either be located isopycnally (April, November 2017) or move noticeably on the density scale (June 2017, April 2019).

In addition to the "century" section, the 2017–2019 survey included "diagonal" section I (Fig. 1), which was periodically carried out perpendicular to the NWS edge, since previously a rise in the hydrogen sulfide zone was regularly discovered near it, in some cases up to depths of 85–90 m [12]. Fig. 5 shows that isooxygen 10 μM in all surveys was located almost isopycnally, but changed its position within range σ_t = 15.6–15.8 kg/m^3, and isosulfide 3 μM was invariably located near isopycnal surface σ_t = 16.1 kg/m^3.



F i g. 5. Position of sub-oxygen zone at section I in 2017–2019

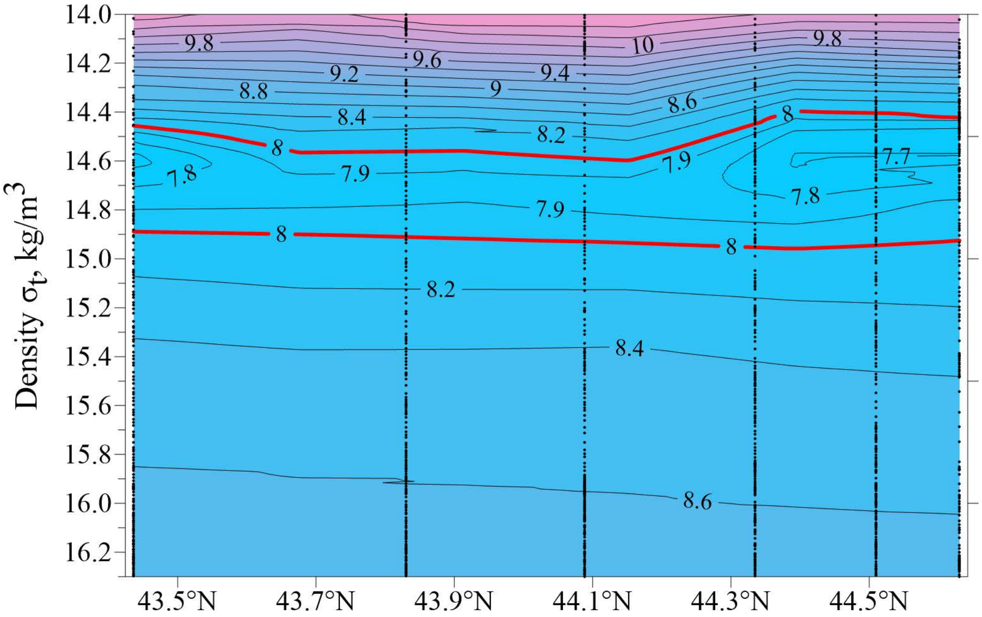
Meridional section III (Fig. 1) southwards of the Kerch Strait is of particular interest when assessing the possible rise of hydrogen sulfide zone near the shelf edge [12]. Fig. 6 shows the position of the sub-oxygen zone at this section in 2017–2019. A relatively stable position of the sub-oxygen zone upper boundary within the range of 15.65–15.8 kg/m^3 and of the hydrogen sulfide upper boundary within the range of 16.15–16.20 kg/m^3 was observed in almost all surveys in this section.



F i g. 6. Position of sub-oxygen zone at section III in 2017–2019

Against this background of relative stability, when the upper boundary of the indicated zone did not drop below σ_t = 15.8 kg/m^3 and the lower one was located in a small interval near isopycn σ_t = 16.1 kg/m^3, the survey in December 2017 is outlined. In this case, clear narrowing of the sub-oxygen zone was observed at two stations closest to the shelf both due to the lowering of isooxygen 10 μM from σ_t = 15.7–15.8 kg/m^3 to σ_t = 15.9 kg/m^3 and due to the rise of isosulfide 3 μM from σ_t = 16.15 kg/m^3 to σ_t = 16.05 kg/m^3. Thus, the sub-oxygen zone thickness was ~ 0.15 units σ_t on the density scale.

The lowering of isooxygen 10 μM can be explained by the fact that the CIL can be the only oxygen source for the sub-oxygen zone and it becomes clear when considering the temperature vertical distribution in the Kerch section in December 2017 (Fig. 7) that the CIL defined by a temperature of 8ºC was both thicker and colder than the surrounding waters in 44.4º–44.6ºN region. This was the very source of the additional amount of oxygen which was enough not only to oxidize the POM, but also to lower isooxygen 10 μM to σ_t = 15.9 kg/m^3.



F i g. 7. Temperature at section III by an STD sensor in December 2017

We explained a similar rise of hydrogen sulfide upper boundary in the NWS by the features of the bottom topography [12] which can also occur in this case.

Hydrogen sulfide maximum concentrations in the Black Sea. The hydrogen sulfide content was determined in the depth range of 1750–2000 m with a discreteness of 50 m at 8 stations depicted in Fig. 1. As is known, a uniform distribution of hydrological and hydrochemical characteristics is observed in the bottom waters of the Black Sea deep part below 1750 m due to convective mixing as a result of heat coming from the bottom surface [19]. The maximum content of hydrogen sulfide, as well as trends towards its change, are of great interest among such characteristics. Thus, a significant increase in the hydrogen sulfide bottom concentrations was observed in the past century, after which its content stabilized at level 378–387 μM [11]. As can be seen from the Table which presents the hydrogen sulfide concentrations at 8 deep-sea stations below 1750 m, almost all these values fall into the interval presented above. After combining the values for all stations into one array after statistical testing [3], we find that the hydrogen sulfide content at depths > 1750 m in the Black Sea is 383 ± 2 μM.

Hydrogen sulfide content (μM) in the Black Sea at the 1750–2000 m depths in 2017–2019

Period	Quantity of measurements	Minimum	Maximum	Average	σ
April 2017	9	378.5	383.4	381.3	1.66
June 2017	14	379.1	388.3	383.6	2.80
November 2017	3	381.7	386.1	383.5	2.27
December 2017	8	381.1	387.9	384.7	2.39
June 2018	7	379.0	386.0	382.2	2.42
August 2018	4	382.4	384.8	383.8	1.03
November 2018	4	377.8	385.6	380.5	3.48
October 2019	6	379.8	382.7	381.3	1.31

Conclusions

Based on the results of 11 expeditions of Marine Hydrophysical Institute of RAS in the Black Sea in 2017–2019, the following was revealed:

1. The decrease in oxygen content with depth (and, accordingly, the oxycline occurrence) began below the isopycnal surface σ_t = 14.5 kg/m^3 in all surveys.

2. The position of the sub-oxygen zone upper boundary determined by isooxygen 10 μM was not strictly isopycnal, but within isopycnal interval σ_t = 15.7–15.85 kg/m^3. The deepest location of the upper boundary below σ_t = 15.8 kg/m^3 was observed in November and December 2017, as well as in August 2018.

3. We assume that the decrease in isooxygen 10 μM to σ_t = 15.9 kg/m^3 in the Kerch shelf area is associated with the presence of a more voluminous and colder intermediate layer above the shelf in December 2017.

4. The position of hydrogen sulfide upper boundary determined by isosulfide 3 μM was raised almost to σ_t = 16.0 kg/m^3 in only one of 11 surveys in April 2017. It was invariably within range σ_t = 16.10 – 16.15 kg/m^3 in all other cases (including one in August 2017, i.e. six months after its rising).

5. Hydrogen sulfide concentration has remained unchanged at 1750–2000 m depths over the past 25 years and is located at level 383 ± 2 μM.

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Contribution of the co-authors:

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Anna V. Masevich – analysis of literature on the research problem, collection and systematization of data, construction of drawings and graphs, discussion of work results

The authors have read and approved the final manuscript.
The authors declare that they have no conflict of interest.