

# Influence of Major Ionic Content of Water on Uncertainties in Calculating Salinity and Density by Standard Methods in the Suboxic Layer of the Black Sea

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## Abstract

Biogeochemical processes change the chemical composition of seawater, which causes a change in its physical properties, such as density, salinity, temperature, etc. This can lead to uncertainties when using a CTD probe to measure the hydrophysical parameters of seawater in field studies and other traditional methods. These uncertainties (errors) can be more significant in inland seas than in the open ocean. This paper is devoted to assessing such uncertainties using the example of the deep waters of the suboxic layer (SOL) of the Black Sea, which is characterized by a layer with increased concentrations of suspended matter and complex biogeochemical processes. An analysis comparing the results of field measurements by a CTD probe in situ with laboratory measurements from six expeditions conducted in 2021 – 2024 is presented. The study shows that ionic variations lead to errors in determining salinity by electrical conductivity of up to 3%, and in density of up to 1%. Underestimation of the salinity calculated by chlorinity ( $S_{Cl}$ ) is 0.2 ‰ (1%) and significant increase in the sulfate-chloride ratio can lead to a decrease in  $S_{Cl}$  by 0.4 g/kg (~2%). It was found that in the studied area, there were often two of the density gradient, causing two maxima suspended matter layers (SM). Significant changes in the major ionic composition of Black Sea's waters depended not only on the concentration of SM in SOL but also on the seasonal variability of SM composition.

**Key words** The Black Sea, Suboxic layer, Water density, Water salinity, CTD sounding, Major ionic composition.

## Introduction

The salinity and density of seawater are of fundamental importance in oceanography and are an integral part of various scientific studies. They are taken into account when determining the quality of seawater, monitoring its biogeochemical parameters, studying the thermohaline circulation of waters, modeling their dynamics, and developing hydrophysical equipment (Yang et al. 2024; Le Menn et al. 2019). The vertical distribution of water density controls the mixing of waters in the ocean (Millero 2013). The

methods for determining salinity and density have been constantly improved, from the calorimetric method for determining salinity to calculations based on chlorinity and electrical conductivity using the practical salinity scale (PSS-78), as well as the development of the equation of state of seawater EOS-80 to calculate its density and later the thermodynamic equation of state (TEOS-10) (Millero 2010; Jenkins & Williams 2025). However, the issue of accurately determining the salinity of seawater in situ, especially in marine areas and inland seas, remains relevant to this day.

The most common method for calculating seawater salinity is based on electrical conductivity, which has been widely used for over 40 years. However, the electrical conductivity of water is limited to measuring the conductivity of certain dissolved elements, and non-conductive dissolved substances (inorganic compounds and organic substances) are not taken into account when calculating salinity, even though they affect its density. The accuracy of calculating salinity from electrical conductivity depends on the constancy of the relative abundance of these ions in the water (Millero et al. 2008; McDougall et al. 2012; Pawlowicz 2013). Underestimation of salinity and density values calculated from electrical conductivity is caused by variations in the concentration ratios of silicates, nitrates, total alkalinity, dissolved inorganic carbon (Millero et al. 2010; Pawlowicz et al. 2011), and many other elements that have not yet been sufficiently studied (Le Menn et al. 2019), along with macroelements. It follows from the above that practical salinity in seawater can differ significantly from the absolute salinity of seawater. The same problem arises when using the TEOS-10 equation for waters of this type if appropriate corrections are not made to account for their specific characteristics (Pawlowicz 2010). The largest anomalies in ion composition are observed in coastal ocean areas, inland seas, and estuaries, where the influence of river salts and solids from river runoff leads to changes in the chemical composition of the waters (Woosley et al. 2014; Savenko & Savenko 2022). The absolute deviation of salinity can be up to 0.03 g/kg in the open ocean, up to 0.1 g/kg in some coastal regions (Pawlowicz 2013), and much more in inland seas.

This work is devoted to determining the salinity and density of waters in the Black Sea, namely in its most complex section from the point of view of biogeochemical processes — the suboxic layer (SOL). The discovery of this layer at the time became a new feature in the study of the chemical structure of the Black Sea (Konovalov & Murray 2001). On the one hand, the Black Sea is an inland sea of the Atlantic Ocean basin; on the other hand, it is the largest meromictic water body on the planet, with a depth of up to 2212 m. The sea is filled with oxygen-free waters containing hydrogen sulfide starting from a depth of approximately 150–170 m. The volume of these waters is almost 80% (Deuser 1974). The salinity of Black Sea waters is almost two times lower than oceanic salinity (Skopintsev, 1975; Sorokin, 1982). The hydrographic regime of the sea is characterized by slightly saline surface waters (17–19 ‰) mixed with river runoff, and more saline deep waters (20–22 ‰) coming from the Mediterranean Sea. As a result, the chemical composition of the Black Sea waters differs from the waters of the World Ocean in a higher content of carbonate ions, higher alkalinity (Sorokin, 1982), and a higher content of sulfates (Kremling 1974) even in the open part of the sea. In coastal waters and in the SOL, there is also an increased content of hydrocarbonates and calcium (Andrulionis et al. 2022; Andrulionis et al. 2024). The  $\text{SO}_4^{2-}/\text{Cl}^-$  ratio is an important characteristic of seawater due to the predominance of sulfates and chlorides in it. The accuracy of determining the physical properties of seawater largely depends on this ratio. The relative content of  $\text{SO}_4^{2-}$  in Black Sea's water is slightly higher than in ocean water and in standard seawater. The presence of sulfates in Black Sea's water is due to their entry from various sources: from the Mediterranean Sea, polluted air, and from continental runoff polluted as a result of economic activity. In addition, the physicochemical composition of the Black Sea has great spatiotemporal variability (Konovalov et al. 2018).

In the water column of the Black Sea at depths of 100–200 m lies the main pycnocline, which is the cause of anaerobic conditions below, as it blocks vertical mixing and the oxygen supply to the deep layers (Vidnichuk & Konovalov 2021). Changes in the freshwater balance and intense eutrophication, together with features of the formation of vertical and horizontal oxygen fluxes and vertical fluxes of suspended matter (SM) and hydrogen sulfide, determine the existence of the SOL between the oxic and hydrogen sulfide layers (Murray et al. 1989). The suboxic zone is the site of an intense oxidation-reduction cycle of sulfur, nitrogen, manganese, and iron species. Similar reactions occur in hemipelagic marine sediments (Murray et al. 1995). The lower boundary of the SOL is taken to be the horizon of the appearance of deep hydrogen sulfide, and the upper boundary is the horizon of the disappearance of oxygen. The position of the upper boundary and the thickness of the SOL are

controlled by the biological processes of the upper layer (Oguz 2005). At the lower boundary of the SOL, oxidation processes of reduced forms of  $\text{H}_2\text{S}$ ,  $\text{NH}_4^+$ ,  $\text{Mn}^{2+}$ ,  $\text{Fe}^{2+}$ , and  $\text{CH}_4$  coming from deep waters occur (Volkov et al 1997). The presence of a large density gradient ( $\nabla\rho$ ) here prevents rapid settling and dispersion of suspended particles in the water column, forming a layer of increased turbidity (SM concentration) (Brewer & Bradshaw 2009; Vinogradov & Nalbandov 1990). A complex chain of oxidation-reduction transformations in the SOL, with the participation of bacteria in the processes of denitrification, anammox, and sulfate reduction, contributes to changes in the ionic composition and leads to an underestimation of salinity values by up to 0.4 ‰, as well as the associated density by up to  $0.2 \text{ kg/m}^3$ . Changes in the ionic composition of water are the main cause of errors in salinity calculations (Andrulionis & Podymov 2024).

Meanwhile, SM is an important component of the marine ecosystem and is formed mainly in the upper water column as a result of the complex impact of substances from different geospheres: the lithogenic component arrives with river runoff, while the biogenic component forms from organic matter, amorphous silica, and suspended calcium carbonate, which are part of plankton and its detritus (Emelyanov 1962; Lisitsyn 1977; Klyuvitkin et al. 2018). The SM flux shows seasonal variability and is determined mainly by biological processes involving plankton. SM delivered with continental runoff and coastal abrasion is distributed under the action of submesoscale eddies, which promote water exchange between the shelf and deep waters of the Black Sea (Ginzburg et al. 2002; Zatsepin et al. 2007; Kubryakov et al. 2018). The contribution of the organic component can reach 83% of the total mass of matter (Mukoseev & Orekhova 2024). An increase in SM in the deep waters of the Black Sea may be preceded by increased eutrophication. Over the last 40 years, eutrophication of the Black Sea's basin has doubled under the influence of anthropogenic load and climate change, increasing the flow of organic SM from the photosynthesis zone to deeper layers. The intensity of oxygen consumption for oxidation of the organic matter flow has increased in all layers of the aerobic zone, and its content in the water has decreased. As a result, the upper boundary of the SOL in the 1970s rose from  $\sigma_t = 15.9$  to  $\sigma_t = 15.4 \text{ kg/m}^3$ , but then partially decreased to  $\sigma_t \sim 15.6 \text{ kg/m}^3$  in the period from 1986 to the mid-1990s due to a decrease in agricultural activity during this period, and has remained in this range since then (Konovalov & Murray 2001; Yunev et al. 2019; Vidnichuk & Konovalov 2021). Since increased eutrophication changes the ionic composition of waters through oxidation-reduction and ion-exchange processes, this affects the physical properties of the reservoir. A brief description of the suboxic zone is as follows: the SOL is located between the lower boundary of the oxycline ( $\sigma_t \sim 15.6\text{--}15.9 \text{ kg/m}^3$ ) and the upper boundary of the hydrogen sulfide zone ( $\sigma_t \sim 16.2 \text{ kg/m}^3$ ). The cold intermediate layer (CIL) is in the range of  $\sigma_t \sim 14.2\text{--}14.8 \text{ kg/m}^3$ , with a core at  $\sigma_t \sim 14.5\text{--}14.6 \text{ kg/m}^3$ . In the range of  $\sigma_t \sim 14.5\text{--}16.5 \text{ kg/m}^3$ , the main pycnocline is usually located, which is divided into upper and lower parts at  $\sigma_t \sim 15.5 \text{ kg/m}^3$ . Between the CIL core and the upper boundary of the SOL, between  $\sigma_t \sim 14.4\text{--}14.6$  and  $\sigma_t \sim 15.6\text{--}15.9 \text{ kg/m}^3$ , there is an oxycline (Konovalov & Murray 2001).

Traditional CTD probe data collection has been used in the Black Sea (Podymov et al. 2023). In addition, “the calculation of practical salinity is based on the practical salinity scale (PSS-78), which corresponds to the most accurate measurements made on standard seawater within the experimental uncertainty and there is no reason to revise it” (Millero 2008). Oceanologists have been using chlorinity to calculate the salinity of Black Sea's waters for over 50 years, applying the formula  $S_{\text{Cl}} = 1.813\text{Cl} \text{ ‰}$  (Kremling, 1974). This ratio correlates quite well with the practical salinity in the open waters of the Black Sea, although in coastal waters and water areas, the difference between  $S_{\text{Cl}}$  and the sum of ions can be up to 2‰ (Andrulionis et al. 2022). According to a number of scientific papers, the physical and biogeochemical properties of the Black Sea's water column change over time due to natural and anthropogenic causes. Air and sea surface temperatures, as well as water salinity, have increased over the past 40 years, while its ice cover and the freshwater part of its water balance have decreased (Ginzburg et al. 2021). It was found that, on average, salinity in the upper 200-meter layer of the sea has increased by about 0.05–0.06 PSU annually over the past decade, which changes water density and can affect the position of the upper boundary of the oxygen-containing layer (Podymov et al. 2021). The biochemical parameters of seawater are also subject to temporal changes. Seasonal fluctuations in the SM flux are associated with biological productivity in the surface layer (Izdar 1984). For example, the increase in phosphate concentration in surface water observed over the past 40 years is accompanied by an increase in the proportion of coccolithophores in phytoplankton communities (Mikaelyan et al. 2011), which are the main carbonate-concentrating biogenic particles in the SM flow of the Black Sea.  $\text{CaCO}_3$

is the second most important lithogenic substance in the SM flow after amorphous silica (Klyuvitkin et al. 2018). Its dissolution changes the carbonate system of the reservoir and affects the ratio of the major ions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ) in the water. In waters with added  $\text{CaCO}_3$ ,  $\text{CO}_2$ , and biogenic substances from the oxidation of plant material, the greatest corrections are required when calculating salinity (Millero 2008; Uchida et al. 2020). From the above, it follows that controlling the standard physical characteristics of seawater by alternative methods is becoming increasingly relevant under changing environmental conditions. Assessing possible uncertainties in determining hydrophysical parameters and analyzing their causes will expand our understanding of the processes of interaction of fine structures in the marine environment.

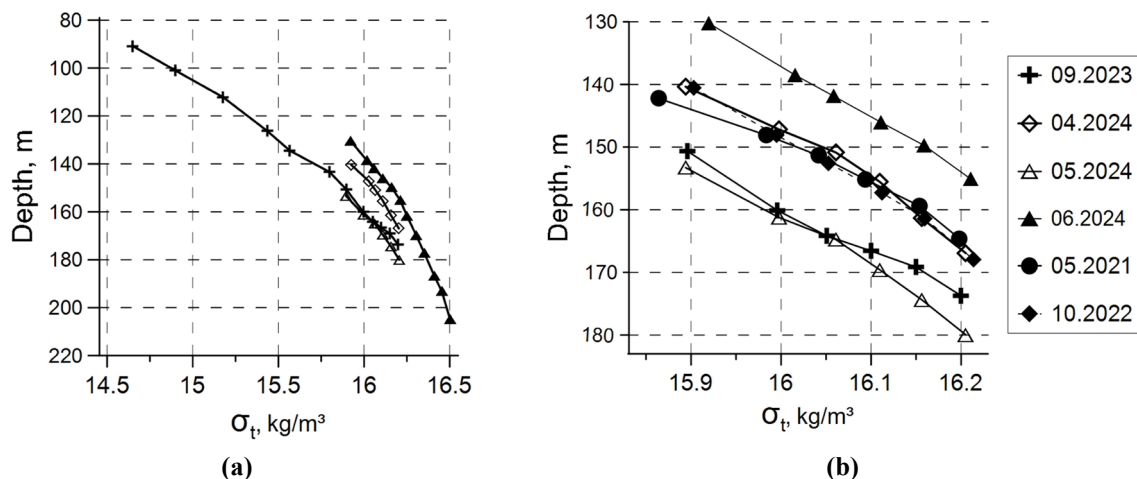
The aim of this study is to assess uncertainties in water salinity and density calculations based on CTD data obtained between 2021 and 2024 and analyze the underlying causes. According to our data, despite the extensive study of the Black Sea, similar studies in its deepwater regions had not been conducted prior to 2021.

## Data and Methods

Water sampling and measurements were carried out in the northeastern Black Sea near the town of Gelendzhik, five nautical miles from the Black Sea's coast in the area of the continental slope with a maximum depth of 1300 m.

The studies were conducted on May, 2021, September, 2022, September, 2023 as well as on April, May and June, 2024 (Tab.1). Sampling was performed using six 4-liter plastic Niskin bottles installed on a rosette equipped with an SBE 19plus CTD probe. A total of 36 water samples were obtained during four expeditions. The characteristics of the sampling stations are presented in Table 1.

A total of 36 water samples were obtained during four expeditions. The samples were collected based on the corresponding isopycnals, the depths of which are shown in Fig. 1. The analysis of the vertical distribution of physicochemical parameters of Black Sea's water with reference to the scale of conditional density began with the works of Vinogradov and Nalbandov (1990), Codispoti et al. (1991), and Tugrul et al. (1992). The use of the scale of conditional density makes it possible to compare the distribution of water properties obtained in different areas and at different times, to obtain quantitative estimates of changes in the marine environment, and to study the processes that determine the evolution of the biogeochemical structure of Black Sea's waters (Kononov & Murray 2001; Ereemeev & Kononov 2006).



**Figure 1.** Density distribution by depth in May 2021 and October 2022 (Andrulonis & Podymov 2024), new results obtained in September 2023, as well as in April, May, and June 2024. (a) – all data, (b) – only in SOL at  $\sigma_t = 15.9 - 16.2 \text{ kg/m}^3$ .  $\sigma_t$  and the depth were calculated based on CTD probe data.

**Table 1.** Characteristics of stations and dates of water sampling.

| Date of sampling | Depth, m | Station coordinates |          |
|------------------|----------|---------------------|----------|
|                  |          | °N                  | °E       |
| 26.05.2021       | 142.23   | 44.51667            | 37.90967 |
|                  | 148.11   |                     |          |
|                  | 151.34   |                     |          |
|                  | 155.14   |                     |          |
|                  | 159.39   |                     |          |
|                  | 164.59   |                     |          |
| 13.09.2022       | 140.56   | 44.51775            | 37.90723 |
|                  | 148.11   |                     |          |
|                  | 152.56   |                     |          |
|                  | 157.32   |                     |          |
|                  | 161.38   |                     |          |
|                  | 167.95   |                     |          |
| 14.09.2023       | 90.85    | 44.52243            | 37.90842 |
|                  | 101.09   |                     |          |
|                  | 112.17   |                     |          |
|                  | 126.20   |                     |          |
|                  | 134.49   |                     |          |
|                  | 143.45   |                     |          |
|                  | 150.63   |                     |          |
|                  | 160.08   |                     |          |
|                  | 164.17   |                     |          |
|                  | 166.58   |                     |          |
|                  | 169.16   |                     |          |
|                  | 173.72   |                     |          |
| 23.04.2024       | 140.353  | 44.51790            | 37.90533 |
|                  | 14.159   |                     |          |
|                  | 150.865  |                     |          |
|                  | 155.518  |                     |          |
|                  | 161.338  |                     |          |
|                  | 166.936  |                     |          |
| 21.05.2024       | 140.35   | 44.51910            | 37.90698 |
|                  | 147.16   |                     |          |
|                  | 150.87   |                     |          |
|                  | 155.52   |                     |          |
|                  | 161.34   |                     |          |
|                  | 166.94   |                     |          |
| 18.06.2024       | 130.24   | 44.51763            | 37.91373 |
|                  | 138.49   |                     |          |
|                  | 141.85   |                     |          |
|                  | 146.03   |                     |          |
|                  | 149.74   |                     |          |
|                  | 155.06   |                     |          |
|                  | 161.76   |                     |          |
|                  | 169.84   |                     |          |
|                  | 177.10   |                     |          |
|                  | 186.68   |                     |          |
|                  | 193.06   |                     |          |
|                  | 204.68   |                     |          |

This paper mainly analyzes the SOL layer in the range of  $\sigma_t = 15.9\text{--}16.2\text{ kg/m}^3$ , but to compare the studied characteristics, an additional 6 samples were taken during two expeditions: in September 2023 above the SOL at  $\sigma_t = 14.65\text{--}15.8\text{ kg/m}^3$ , and in June 2024 below its lower boundary at  $\sigma_t = 16.25\text{--}16.5\text{ kg/m}^3$  (Fig. 1ab). In this study, the paper also used results obtained earlier in 2021 and 2022 and published in Andrulionis & Podymov (2024).

The Figure 1 ab shows that the position of the SOL in the range  $\sigma_t = 15.9\text{--}16.2\text{ kg/m}^3$  changed in the water column by more than 20 m in the range 2021–2024: the upper boundary of the layer rose by 23 m, the lower one dropped by 25 m. The highest position of  $\sigma_t = 15.9\text{ kg/m}^3$  was observed in June 2024, located at a depth of 130 m, while the lower boundary of the SOL ( $\sigma_t = 16.2\text{ kg/m}^3$ ) was at a depth of 155 m. The lowest position of the SOL was recorded in May 2024. In this case,  $\sigma_t = 15.9\text{ kg/m}^3$  was located at a depth of 153 m, and the lower boundary of the SOL ( $\sigma_t = 16.2\text{ kg/m}^3$ ) was at a depth of 180 m. It was previously discovered that intense fluctuations in the position of the permanent pycnocline and the upper boundary of the hydrogen sulfide zone can have a characteristic period of about 5–10 days, and the thickness of the oxygen-containing layer can vary by 20–40 m (Zatsepin et al. 2013). Figure 1 shows that not only the thickness of the SOL, but also its position as a whole changes vertically in the water column. Water samples from the bathometers were placed in sealed 1-liter containers and delivered to the laboratory for subsequent analysis. The samples were pre-filtered through a GF/F Whatman membrane filter of  $0.7\text{ }\mu\text{m}$  pore size in 2021–2022 and  $0.45\text{ }\mu\text{m}$  pore size in 2023–2024 to remove SM and placed in 300-mL borosilicate glass containers. The density and salinity of the water were determined in two ways. First, during sampling, the electrical conductivity of seawater was measured using the SeaBird 19plus CTD probe, while the practical salinity and density were calculated using standard methods: PSS-78 and EOS-80, respectively. Second, the salinity and density of the water were determined in the laboratory.

**Salinity study.** The salinity of the deep waters of the Black Sea was determined in three ways. The practical salinity of the studied samples (SP) was calculated in the standard way by electrical conductivity. To calculate the sum of ions (SS), the concentrations of the main ions in the water composition were determined. When calculating salinity using the chlorine coefficient ( $S_{\text{Cl}}$ ), the ratio from Kremling (1974) was used:

$$S = 1.813 \text{ Cl } \text{‰}.$$

To obtain the sum of ions, their concentrations in the studied samples of the Black Sea were determined according to the methods described in detail by Andrulionis & Zavialov (2019), taking into account the mineralization of Black Sea's water. The ions  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  were determined by potentiometric titration,  $\text{K}^+$  was determined gravimetrically, and the concentration of  $\text{Na}^+$  was calculated as the difference between the sum of anions and cations in moles of equivalents, with subsequent recalculation in g/kg.

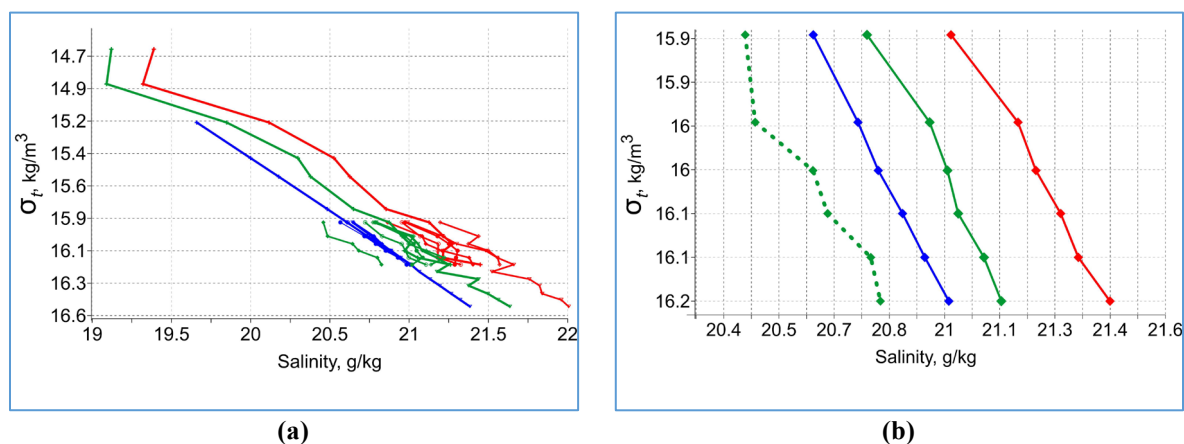
Deionized water (conductivity  $\sim 0.17\text{ }\mu\text{S/cm}$ ) was used to prepare reagent solutions and to dilute samples. The mass of the analyzed sample was measured by weighing on an OHAUS laboratory analytical balance of the first accuracy class with an error of 0.001 g. The accuracy of salinity calculation depends on the error of the equipment and methods. It is estimated at  $\pm 0.1 \times 10^{-2}\text{ }\mu\text{S/cm}$  when calculating by electrical conductivity,  $\pm 0.2 \times 10^{-2}\text{ g/kg}$  by chlorinity, and  $\pm 0.1 \times 10^{-1}\text{ g/kg}$  by the sum of ions (Millero 2013). To assess the differences between the obtained salinity values, the deviations of these values SP from SS and  $S_{\text{Cl}}$  from SS were calculated, both in ‰ and as the element share relative to the total mass of components in ‰.

**Density study.** Density was calculated in two ways: from CTD probe data using the EOS-80 equation (UNESCO, 1981) and also determined in the laboratory using an Anton Paar DMA 5000M precision density meter (Austria) applying the U-tube oscillation method, calculated by accurately determining the characteristic frequency and mathematical transformation from the ratio of the U-tube oscillation period to the reference oscillator (Instruction Manual DMA 2010). The density of water samples from the Black Sea's SOL was determined at in situ temperature and atmospheric pressure and expressed as  $\sigma_t$  in  $\text{kg/m}^3$ .

**Turbidity determination.** During sampling, turbidity or SM concentration was measured in nephelometric turbidity units (NTU) using a Seapoint turbidimeter. Turbidity measurements allow analysis of SM concentration and its distribution in the water column.

## Results

**Salinity study.** Fig. 2ab shows the results of the salinity study in the Black Sea's water column using three methods. The profiles of practical salinity (SP), sum of ions (SS), and salinity calculated from chlorinity ( $S_{Cl}$ ) are presented for the SOL, as well as for the overlying and underlying deep layers of the water column in the range of  $\sigma_t = 14.7\text{--}16.5\text{ kg/m}^3$ . The lower part of the figure shows the averaged salinity profiles only for the SOL at  $\sigma_t = 15.9\text{--}16.2\text{ kg/m}^3$ .



**Figure 2.** Salinity profiles for six expeditions: four in 2023 and 2024 and two in 2021 and 2022 (from Andrulionis & Podymov 2024) in the suboxic layer of the Black Sea. (a) – all salinity profiles obtained for six studies in the range of  $\sigma_t = 14.65\text{--}16.50\text{ kg/m}^3$  (according to CTD data), (b) – the averaged profiles for six studies, only in the range of  $\sigma_t = 15.9\text{--}16.2\text{ kg/m}^3$  (directly in the SOL). Salinity is calculated in different ways: by electrical conductivity (blue icons and lines), by the sum of major ions (red icons and lines), and by chlorinity (green icons and lines). The dotted green line shows the salinity profile obtained by chlorinity in April 2024.

Fig. 2ab shows that the salinity values calculated in different ways were mainly arranged in the following order in 2021–2024:  $SS > S_{Cl} > SP$ , with the difference between SS and SP ( $\Delta S$ ) being  $0.27\text{--}0.65\text{ ‰}$  ( $1.3\text{--}3.1\%$ , averaged  $2\%$ ), between SS and  $S_{Cl}$  ( $\Delta S_{Cl}$ )  $0.16\text{--}0.43\text{ ‰}$  ( $1\text{--}2\%$ , averaged  $1.5\%$ ), and between  $S_{Cl}$  and SP ( $\Delta S_{ClP}$ )  $0.03\text{--}0.29\text{ ‰}$  ( $0.2\text{--}1.4\%$ , averaged  $1\%$ ), except for the study in April 2024. For the samples obtained in April 2024, the principle of increasing salinity was different:  $SS > SP > S_{Cl}$ , while  $S_{Cl}$  (green dotted line on Fig 2b) was lower by  $0.14\text{--}0.29\text{ ‰}$  ( $0.7\text{--}1.4\%$ , averaged  $1\%$ ) compared to SP. That is, in April  $\Delta S_{ClP}$  has a negative value and positive the rest of the time. At the same time,  $\Delta S$  in April was at  $0.22\text{--}0.40\text{ ‰}$  ( $1\text{--}2.1\%$ , averaged  $1.6\%$ ), and  $\Delta S_{Cl}$  was  $0.41\text{--}0.60\text{ ‰}$  ( $1.9\text{--}2.9\%$ , average  $2.5\%$ ). The reason for the underestimation of  $S_{Cl}$  was a significant decrease in the relative content of chlorides and increase in sulfates in the water composition, caused by changed conditions of the aquatic environment in the spring. It is known that, unlike other months, in March and April, a high content of lithogenic (terrigenous) material in suspension enters with continental runoff and coastal abrasion, accompanied by high phytoplankton concentration (Rusanov et al. 2018). This phenomenon could affect the ion ratio in the study area.

However, good convergence of SP values between expeditions was observed despite seasonal variability in SM concentration and composition. The standard deviation (SD) between these values averaged  $0.02\text{ ‰}$  ( $\sim 0.1\%$ ). On the contrary, significant discrepancies were observed between SS values and between  $S_{Cl}$  values, with SD being  $\sim 0.15$  and  $\sim 0.13\text{ ‰}$  ( $0.7$  and  $0.6\%$ ), respectively. It is natural that waters with the same conductivity have the same practical salinity, but the relative ionic composition and chlorinity may differ (Millero et al. 2008). Conservatism and uniformity of practical salinity distribution are important in studying global processes of seawater circulation propagation and are convenient for modeling these processes, but they do not fully reflect the picture of fine-structure

interactions in the Black Sea's water column. As shown in Fig. 2, the calculation of  $S_{Cl}$  values from the relationship given by Kremling (1974) will also not give reliable results in the Black Sea SOL, since oxidation-reduction reactions and sorption-desorption processes significantly change the chemical composition of these waters.

**Density study.** Determining the density values of the Black Sea's suboxic zone water obtained using two methods made it possible to calculate the density gradient with an average step of 5 m, as well as to compare the results and analyze the differences between them. Fig. 3abcdef shows the results of calculating the density gradient ( $\nabla\rho$ ) based on CTD probe data (CTD — blue profile line) and the densitometer (DMA — red profile line) at in situ temperature and atmospheric pressure, as well as a comparison of these profiles with the turbidity profile (green line). Fig. 3ef show two distinct gradients calculated from the densitometer data. They are marked with dots and red numbers 1 and 2. In Fig. 3ef they are most noticeable due to the larger depth range. Fig. 3d shows one density gradient corresponding to the second turbidity peak (the layer of increased SM concentration) — it is quite possible that the first gradient was located above the studied zone. All figures show that the maximum value of gradient 1 was generally in the range of  $\sigma_t = 15.9\text{--}16.1\text{ kg/m}^3$ , and gradient 2 in the range of  $\sigma_t = 16.16\text{--}16.2\text{ kg/m}^3$ . The density gradient maxima are located in the regions of the turbidity peaks (green numbers 1 and 2). This means that the pycnocline prevents rapid settling of SM, causing its accumulation in regions of increased gradients. The density gradient calculated from CTD probe data was most often  $\sim 0.01\text{ kg/m}^3$  per 5 m, but sometimes reached 0.02 or 0.03  $\text{kg/m}^3$  per 5 m (Fig. 3ed). The gradient calculated from the densitometer data ranged from 0.03 to 0.04  $\text{kg/m}^3$  per 5 m, averaging 0.02  $\text{kg/m}^3$  per 5 m. Thus, the maximum density gradient according to DMA was on average about two times greater than according to the CTD probe data.

In Fig. 3cdef, two distinct maxima (peaks) are mainly visible, while in Fig. 3ab there is only one. The first peak of SM was in the range of  $\sigma_t \sim 16.04\text{--}16.10\text{ kg/m}^3$ , and the second one was at  $\sigma_t \sim 16.13\text{--}16.26\text{ kg/m}^3$ . As was established earlier, two turbidity peaks are observed more often than one in the SOL. Their appearance was associated with changes in the concentrations of oxygen and other elements. It is known that the main component of the upper turbidity maximum is insoluble manganese oxide ( $\text{MnO}_2$ ) (Volkov et al. 1997). It is known that in the presence of  $\text{MnO}_2$ , the  $\text{O}_2$  concentration decreases in the upper peak and increases again below it. In the lower peak, the  $\text{O}_2$  concentration decreases again and is no longer observed below it (Stunzhas & Yakushev 2006). The lower turbidity peak is located at the lower boundary of the SOL at  $\sigma_t \sim 16.2\text{ kg/m}^3$ , where the upper boundary of the hydrogen sulfide zone begins. Redox reactions and other complex biochemical processes occurring at these peaks alter the MIC in the surrounding waters. This causes errors in salinity determinations during in situ measurements.

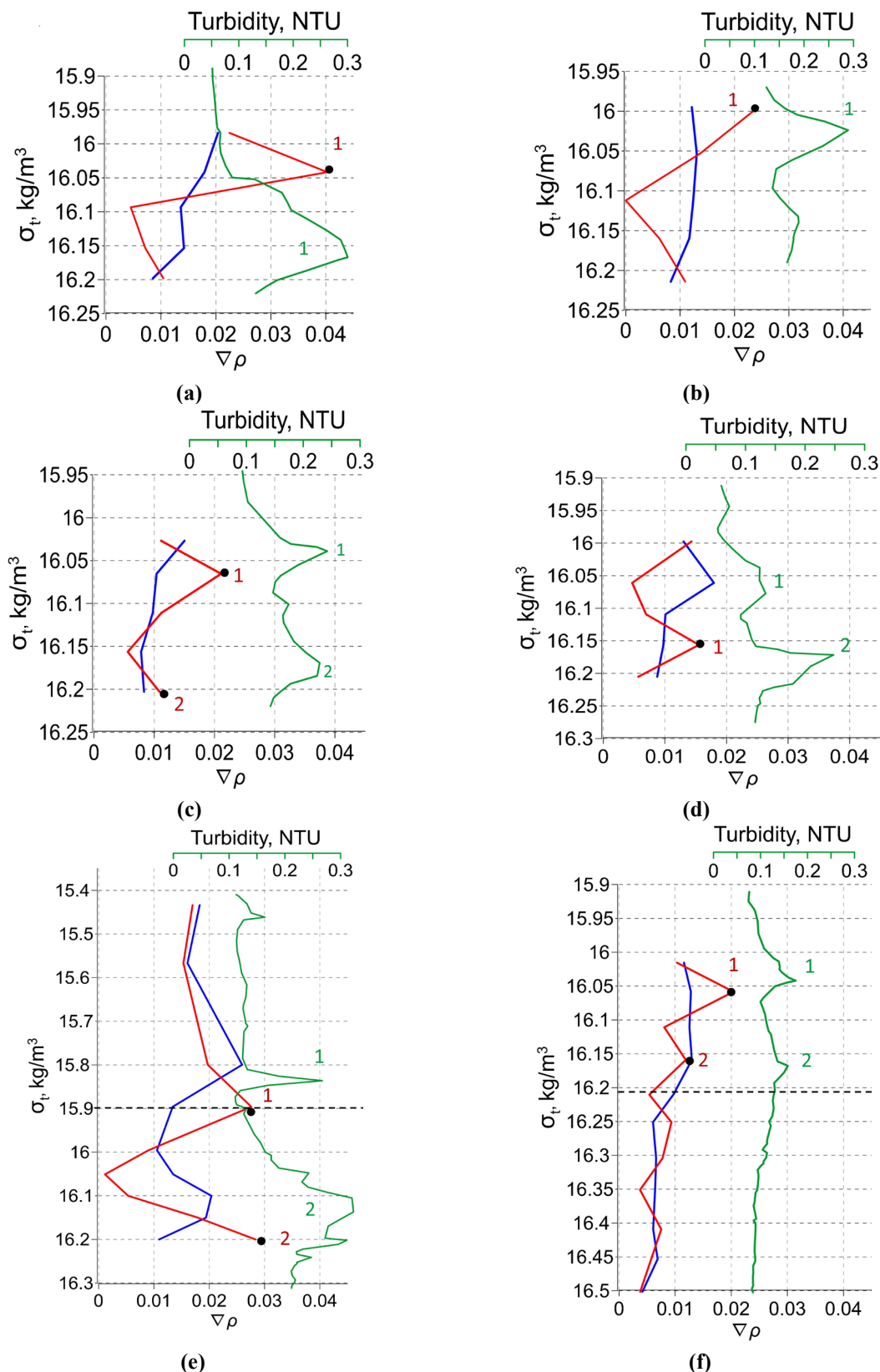
The observed correlation between turbidity peaks and density gradients indicates a relationship between these parameters, meaning that the formation of turbidity peaks is caused by the presence of significant density gradients in the SOL.

Although significant density gradients are primarily recorded only by laboratory methods, they can sometimes be visible in probe data. For example, Figure 3E shows density gradients recorded not only by a densitometer but also by CTD data. The correlation between the peaks in the red and blue profiles is clearly visible, although somewhat offset relative to each other. The best confirmation of the formation of high-SM layers is their location on density gradients.

### Effect of ion composition on salinity and density calculation uncertainties based on CTD probe data.

The results of the study examining the influence of MIC and SM on the uncertainties in calculating salinity and density in the SOL are presented in Figure 4abcdef. The figure shows the distribution of  $\Delta\sigma_t$ ,  $\Delta S$ , and  $\text{SO}_4^{2-}/\text{Cl}^-$  profiles in the water column, the relative abundance of calcium and magnesium ions, and the turbidity profile in the SOL layer, as well as in the overlying and underlying layers of Black Sea water, during the period 2021–2024.

A comparison of water salinity and density determined using different methods revealed that the laboratory values exceeded those calculated using CTD probe data. The absolute deviations between the laboratory and in situ results were: for density ( $\Delta\sigma_t$ ) 0.0–0.2  $\text{kg/m}^3$  (0–1.2%, averaging 0.6%), for salinity:  $\Delta S = 1.5\text{--}3\%$ ,  $\Delta S_{Cl} = 1\text{--}2\%$  and  $\Delta S_{ClP} \sim 1\%$ .



**Figure 3.** Density gradient (along the x-axis:  $\nabla\rho$ , kg/m<sup>3</sup> per 5 m) in the waters of the suboxic layer of the Black Sea and its relationship with the layer of increased SM concentration. The gradient calculated from CTD probe data — blue lines; measured by the densitometer — red lines. Figs. 3 (a, b) show the results obtained in May 2021 and October 2022 (Andrulionis & Podymov 2024); Figs. 3 (e, c, d, and f) — new results obtained in September 2023, as well as in April, May, and June 2024, respectively. The dotted line in Fig. 3 (e) separates the layer located above the usually studied region of the suboxic layer at  $\sigma_t = 15.9$ –16.2 kg/m<sup>3</sup>, and in Fig. 3 (f) — below the suboxic layer (hydrogen sulfide zone).

Largest and smallest deviations  $\Delta\sigma_t$  and  $\Delta S$  were observed in the areas of turbidity peaks, indicating a relationship between them. Sharp changes in the  $\text{SO}_4^{2-}/\text{Cl}^-$  profile are most pronounced in the layer of maximum turbidity, where biochemical processes are particularly active. This indicates a change in the ratio of the major ions in this water layer.

Analysis of the obtained results revealed a relationship between the layer of increased SM concentration and changes in the ionic composition, and uncertainties in calculating salinity and density from electrical conductivity. For example, the well-defined correlation of the studied parameters in Fig. 4C suggests that they in measuring the physical parameters in situ were largely related to the  $\text{SO}_4^{2-}/\text{Cl}^-$  ratio, as well as to the relative content of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions in the water and their variations. For example, an increase in  $\Delta\sigma_t$  mainly was accompanied by an increase in  $\text{SO}_4^{2-}/\text{Cl}^-$  and in the relative content of calcium ions, while the content of magnesium ions decreased. However, in April 2024 (Fig. 4d), this dependence was not observed, although  $\text{SO}_4^{2-}/\text{Cl}^-$  and  $\text{Ca}^{2+}$  content were at their highest values.

The elevated content of hydrocarbonates, sulfates, and calcium ions in the Black Sea waters compared to the ocean and the SSW contributes to an underestimation of the  $S_{\text{Cl}}$  values by 3% or more (Androulioni et al., 2022; Androulioni et al., 2024).

In this work, it was noted  $\text{SO}_4^{2-}/\text{Cl}^-$  ratio was found to be generally in the range of 0.139–0.156 with an average of 0.145, and the  $\text{Ca}^{2+}$  content was in the range of 1.15–1.48% with an average of 1.32% in the Black Sea SOL, except for samples obtained in April 2024. This circumstance typically results in  $S_{\text{Cl}}$  being 1% higher than the SP. In April 2024, the  $\text{SO}_4^{2-}/\text{Cl}^-$  ratio was in the range of 0.156–0.167 with an average of 0.163, and the  $\text{Ca}^{2+}$  content was in the range of 1.41–1.49% with an average of 1.45%. A 1% decrease in  $S_{\text{Cl}}$  relative to the SP was observed. It follows that the determination of salinity by the chlorine coefficient may significantly (to 2%) underestimate salinity due to seasonal changes in MIC.

Overall, the MIC in all water samples from SOL from 2021–2024 differed from the MIC in ocean waters and SSW from Millero et al. (2008). These differences, on average, for the SOL were 0.6% and 0.3% lower for  $\text{Cl}^-$  and  $\text{Na}^+$ , respectively, and 0.13%, 0.5%, and 0.15% higher for  $\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$ , and  $\text{Ca}^{2+}$ , respectively, than in SSW. The concentrations of  $\text{K}^+$  and  $\text{Mg}^{2+}$  ions were almost equivalent to their concentrations in SSW.

The relative content of chlorides was on average 54.47% (SD 0.12),  $\text{SO}_4^{2-}$  was 7.95% (SD 0.23) in all samples 2021–2024, except for April 2024, where  $\text{Cl}^-$  was 53.79% (SD 0.17), and  $\text{SO}_4^{2-}$  was 8.76% (SD 0.20).  $\text{Na}^+$  and  $\text{K}^+$  content ions was in all water samples averaged 30.52% (SD 0.15) and 1.26% (SD 0.07) respectively and experienced minor fluctuations with depth.  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  content was 1.34% (SD 0.08) and 3.53% (0.07) respectively.  $\text{HCO}_3^-$  content was almost constant on average 0.92% (SD 0.03) and did not change with depth in the study area.

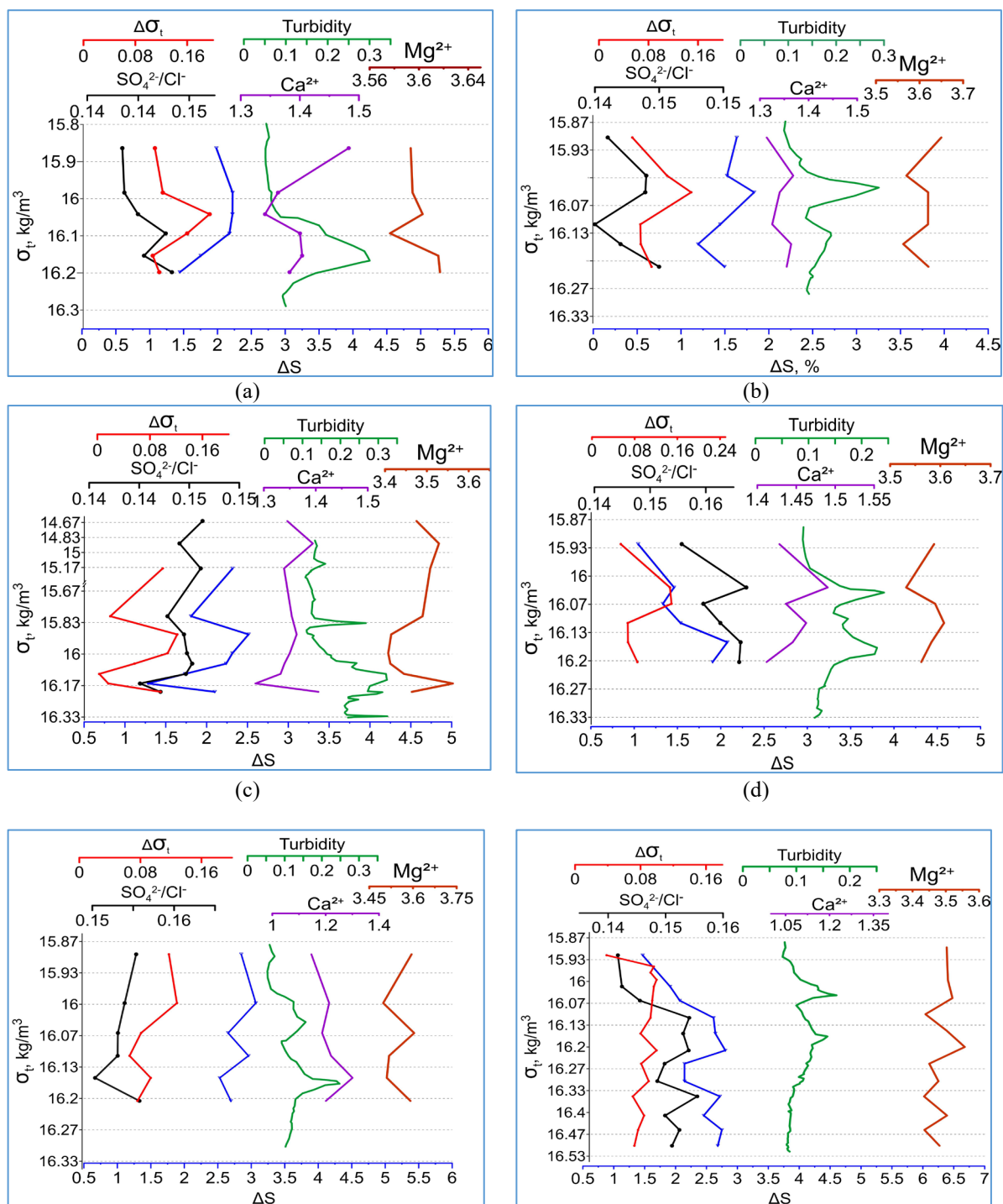
In the suboxic zone, processes similar to those occurring in marine sediments occur. Terrigenous, volcanic, and biogenic materials are mixed, decomposed, and dissolved under intense redox reactions. During the decomposition process, chemical elements are released into the aquatic environment or, conversely, absorbed from it. This shifts the ionic equilibrium in the chemical composition of the water, altering the ratio of MIC ions, which affects its electrical conductivity and, consequently, the accuracy of the STD probe's physical properties determination.

## Conclusion

The study of the major ionic composition Black Sea's deep waters showed that relative content of the major ions in the suboxic layer differed from the SSW increased content of sulfate, bicarbonate and calcium ions and decreased content of chlorides and sodium ions, with variations in their relative abundances. This causes uncertainties in calculating physical parameters using STD data.

Obtained that the magnitude of uncertainties in calculating salinity and density from electrical conductivity depends on the relative content of the MIC mainly on increased  $\text{HCO}_3^-$  content, the shift in the  $\text{SO}_4^{2-}/\text{Cl}^-$  ratio, as well as on variations in  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  content in the Black Sea's SOL. The change in ion composition is due to the high concentration of suspended matter, but also to the SM composition.

It has been found that underestimation of the salinity calculated by chlorinity ( $S_{\text{Cl}}$ ) is 1%. A study conducted in April 2024 found that significant increase in the sulfate-chloride ratio can lead to a decrease in salinity by the chlorinity by 0.4 ‰ (~2%) in the Black Sea's SOL.



**Figure 4.** Distribution of  $\Delta\sigma_t$  ( $\text{kg/m}^3$ ),  $\Delta S$  ( $\text{g/kg}$ ), as well as  $\text{SO}_4^{2-}/\text{Cl}^-$  and SM (Turbidity, NTU) in the suboxic layer of the Black Sea at different times: in May 2021 (a) and October 2022 (b) from (Andrulionis & Podymov, 2024), in September 2023 (c), as well as in April (d), May (e), and June (c) 2024.

It was found that salinity calculated from CTD probe data was underestimated by 0.27–0.65‰ (1–3%), and density by up to 0.13  $\text{kg/m}^3$  (1%). Besides, the density gradient measured by the densitometer was almost twice that obtained from CTD probe data. Furthermore, two density gradient maxima are often present, which are not accounted for by the CTD probe. These gradients cause two SM peaks to appear in the salinity layer in the SOL.

The authors propose laboratory methods for determining salinity (using the MIC or the sum of ions (SS)) and density (using a densitometer) that improve the accuracy of the results. The methods

makes it possible to determine the causes of salinity changes due to variations in the content of certain ions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) in deep waters of the meromictic water body.

The uncertainties must be taken into account when calculating salinity in the Black Sea and in similar water bodies. If the observed trend toward increasing climatic and anthropogenic impact on the Black Sea's ecosystem continues, changes in the chemical properties of its waters are inevitable, which means that the magnitude of errors in determining their physical properties by standard methods may also increase. Therefore, regular monitoring of the physicochemical parameters of the deep waters of the Black Sea is necessary to identify possible changes in the marine environment.

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