Original article

Sea-Air CO_2 Flux in the Northeastern Part of the Black Sea

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Abstract

Carbon dioxide is one of the green gases and its entry into the atmosphere and further redistribution in the waters of the World Ocean not only plays a significant role in the climate on the Earth, but also affects the characteristics of waters. The research of inland seas, e.g. the Black Sea, makes it possible to study the influence of atmospheric CO_2 on the characteristics of waters and to assess the contribution of regional ecosystems to the total budget of the CO_2 flux of the World Ocean. The paper presents numerical estimates of the sea–air CO_2 flux, analyzes its direction and identifies factors that determine the values of the CO_2 flux in the northeastern part of the Black Sea during a cold period. For the analysis, the data obtained during the cruise of R/V *Professor Vodyanitsky* in December 2022 were used. The values of the sea–air flux of carbon dioxide were calculated taking into account the wind speed and pCO_2 gradient between the sea surface and the near sea surface atmosphere. According to the direct measurements of pCO_2, the value of the CO_2 flux in December 2022 varied widely from –0.05 to –8.74 $mmol \cdot m^{-2} \cdot day^{-1}$, the average value being –2.11 ± 1.79 $mmol \cdot m^{-2} \cdot day^{-1}$. It was established that during the cold season, the CO_2 flux was directed from the atmosphere to the sea surface. Thus, the waters of the Crimean coast serve as a stock of atmospheric CO_2. Local minima of flux values were observed in the southeastern regions of the Crimean coast. When analyzing the correlation of the CO2 flux with temperature, wind speed and ΔpCO_2, the strongest relationship was found with wind speed (–0.93), while the weakest one was with ΔpCO_2 (0.22). Therefore, the intensity of the sea–air CO_2 flux was determined by wind speed, while the direction of the flux was determined by ΔpCO_2. The temperature contribution manifested as change in the concentration of CO_2 in the water column.

Keywords: CO_2 flux, Black Sea, carbon dioxide, partial pressure of carbon dioxide, carbon cycle

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Поток CO_2 на границе с атмосферой в северо-восточной части Черного моря

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Аннотация

Углекислый газ является одним из климатообразующих веществ, его поступление в атмосферу и дальнейшее перераспределение в водах Мирового океана играют значительную роль в формировании климата на Земле и влияют на характеристики вод. Изучение внутренних морей, таких как Черное море, позволяет исследовать влияние атмосферного CO_2 на характеристики вод и оценить вклад региональных экосистем в общий бюджет CO_2 вод Мирового океана. В работе приведены количественные оценки потока CO_2 на границе с атмосферой, проанализирована его направленность, выделены факторы, определяющие величину потока CO_2 в северо-восточной части Черного моря в холодный период. Для анализа использованы данные, полученные в ходе экспедиционных исследований на НИС «Профессор Водяницкий» в декабре 2022 г. Величина потока углекислого газа на границе вода – атмосфера рассчитывалась с учетом скорости ветра и градиента pCO_2 между поверхностью моря и приводным слоем атмосферы. По данным прямого определения pCO_2, значения потока CO_2 в декабре 2022 г. изменялись в широких пределах от –0.05 до –8.74 ммоль·м$^{-2}$·сут$^{-1}$, среднее значение соответствовало –2.11 ± 1.79 ммоль·м$^{-2}$·сут$^{-1}$. Установлено, что в холодный период года поток CO_2 был направлен из атмосферы в поверхностный слой вод. Таким образом, воды Крымского побережья служат стоком атмосферного CO_2. Локальные минимумы потока наблюдались в юго-восточной части Крымского побережья. При анализе корреляционной связи потока CO_2 с температурой, скоростью ветра и ΔpCO_2 наиболее сильная связь выявлена со скоростью ветра (–0.93), слабая – с ΔpCO_2 (0.22). Следовательно, интенсивность потока CO_2 на границе с атмосферой определялась скоростью ветра. Однако направление потока зависело от ΔpCO_2. Вклад температуры проявлялся в изменении концентрации CO_2 в водной толще.

Ключевые слова: поток CO_2, Черное море, углекислый газ, парциальное давление углекислого газа, цикл углерода

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Introduction

The global cycle of natural substances includes their transport among various biogeochemical reservoirs and regulating balance and budget of substances in the atmo-, litho- and hydrosphere. One of such natural cycles is the carbon cycle, the most important component of which is carbon dioxide (CO_2) [1)] [1–5].

[1)] Raven, J., Caldeira, K., Elderfield, H., Hoegh-Guldberg, O., Liss, P., Riebesell, U., Shepherd, J., Turley, C. and Watson, A., 2005. *Ocean Acidification due to Increasing Atmospheric Carbon Dioxide*. London: The Royal Society, 57 p.

CO_2 is one of the green gases [1–6] and its entry into the atmosphere and further redistribution in the waters of the World Ocean not only plays a significant role in the formation of the climate on the Earth [1], but also affects the characteristics of waters [1, 6, 7].

The waters of the World Ocean are still its natural stock despite the continuous increase in the level of atmospheric CO_2 (about 0.4% per year) and to date its content achieves more than 420 µatm (https://gml.noaa.gov/ccgg/trends/mlo.html). They absorb up to 25% atmospheric CO_2 from anthropogenic emission, thereby support to reduce CO_2 concentrations in the atmosphere [7]. However, its accumulation in the water column leads to negative consequences for the ecosystems of the World Ocean which is revealed in the disruption of natural balances, in particular carbonate ones, decrease in pH and oxygen concentration and emergence of oxygen deficiency zones. The Ocean's ability to absorb carbon dioxide from the atmosphere decreases over time [8–10] and waters can even become a source of CO_2 for the atmosphere in some extreme cases [7].

The primary factor determining the influence of CO_2 on the state of marine systems is its flux from the atmosphere which depends, other things being equal, on the ratio of the partial pressure of CO_2 in the near sea surface atmosphere and the equilibrium partial pressure of CO_2 in the sea surface. This ratio determines the direction and values of the CO_2 flux.

An important aspect of the research of the sea–air CO_2 flux and the pCO_2 value in the sea surface is the study of the nature of changes on time scales from seasonal to interannual which is associated with significant spatial and temporal variability of biological and physical processes affecting these characteristics.

Inland seas are characterized by more intense physical and biogeochemical processes compared to open areas of the World Ocean. As a result, their ecosystem is more dynamic on a temporal and spatial scale and any external influence manifests itself more quickly. First of all, such manifestations include changes in the characteristics of the system: oxygen and CO_2 concentrations, pH values, as well as speed and direction of production and destruction processes [10]. Moreover, these ecosystems are characterized by a more pronounced response to changes in CO_2 concentration in the atmosphere which manifests itself primarily in a shift in the carbonate system equilibrium, as well as changes in redox conditions [1)] [5–7, 10].

The research of inland seas makes it possible to study the influence of atmospheric CO_2 on the characteristics of waters and to assess the contribution of regional ecosystems to the total budget of the CO_2 flux of the World Ocean.

The Black Sea is one of such inland seas. The shelf water characteristics of the northern part of the sea are largely determined by freshwater river runoff and atmospheric contribution, of the northeastern part – by the Azov Sea waters, of the deep-water part – by the Rim Current [11]. This sea is characterized by a wide range of changes in salinity and temperature [11], high intensity and seasonal changes in primary production processes [12], high values of alkalinity and total inorganic carbon content [13–15]. All this largely determines the state of the carbonate system of sea waters, the CO_2 content in the sea surface and the formation of the sea–air CO_2 flux.

The factors listed above are influenced by seasonal variability. Accordingly, both CO_2 concentration and CO_2 flux also show intra-annual variability.

It can be assumed that during a cold period, the CO_2 concentration should be primarily determined by an abiotic factor – temperature and vertical transport of CO_2 by deep waters, as well as sea–air metabolic processes. In summer, the predominant factor should be biotic due to the occurrence of biogeochemical processes involving organic matter.

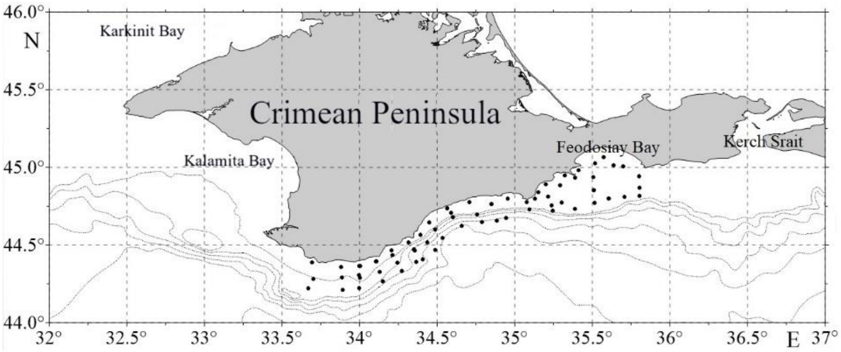
The purpose of this work was to obtain numerical estimates of the sea–air CO_2 flux and to identify its direction and the factors that determine the values of the CO_2 flux in the area of the Crimean coast of the Black Sea during the cold period when the contribution of the abiotic factor predominates.

Previously, the CO_2 flux estimates for this Black Sea ecosystem were carried out based on calculated data [13] or for a local area [14].

Materials and methods

The data obtained during the cruise of R/V *Professor Vodyanitsky* in December 2022 (the 125th cruise, 02–27.12.2022) were used in this work. According to [11], this period refers to late autumn.

Fig. 1 shows the area under study and sampling map. The studied area includes a 12-mile zone of the Crimean coast in the northern part of the Black Sea.



F i g . 1 . Sampling map

Samples from the near sea surface atmosphere were taken at a height of 10 m above sea level. The air intake tube was located in such a way as to avoid the CO_2 influx from the working mechanisms of the vessel, if possible. An LI-7000 by LI-COR infrared analyzer with a working range of CO_2 concentration of 0–3000 ppm and water vapor of 0–60 mmol/mol was used to determine the volumetric concentration and partial pressure of CO_2 directly. In this case, the measurement error is less than 1% of the measured values [15].

Water samples were taken from the sea surface (1–3 m) using a continuous seawater supply system. Next, the water was transported at a constant speed to an equilibrator with the help of which equilibrium was established with a certain volume of atmospheric air at the temperature of sea water according to the method described in [15]. Air from the equilibrator was pumped at a constant speed through the cell of the LI-7000 by LI-COR infrared analyzer in which the concentration of CO_2 and water vapor was determined at the cell temperature. The temperature of the cell is determined by a temperature sensor installed inside it and is in equilibrium with the temperature of the atmosphere surrounding the equilibrator. Next, the carbon dioxide concentration was converted to the partial pressure of carbon dioxide:

$$pCO_2 = x\,(CO_2)\cdot p_{ATM},$$

where $p(CO_2)$ is partial pressure of carbon dioxide, µatm; $x\,(CO_2)$ is carbon dioxide concentration, µmol/mol; p_{ATM} is atmospheric pressure, atm.

The temperature and salinity of the sea surface were measured with an IDRONAUT OCEAN SEVEN 320PlusM WOCE-CTD multiparameter probe, and at shallow water stations (less than 50 m) – with the GAP AK-16 hydrological CTD probe.

Meteorological parameters were measured with recording equipment of the hydrometeorological data collection complex [16]. A sensor for measuring wind speed and direction was installed on a side boom 1.5 m long in the direction of the port side on the foremast, with the north direction chosen according to the vessel's heading. The sensor is installed at a height of about 8 m from sea level. The data passed quality control with the rejection of unreliable fragments and were reduced to a standard observation height (10 m) [17]. According to the recommendations of the World Meteorological Organization, the measured parameters were averaged over 10 minutes, and further analysis was carried out for the averaged values. Wind gusts are given as instantaneous wind speed values over 5 s [17].

The values of the sea–air flux of carbon dioxide were calculated using the equations and assumptions described in [18] taking into account the wind speed and pCO_2 gradient between the sea surface and the near sea surface atmosphere:

$$F_{CO_2} = k \cdot K_0 \cdot \Delta pCO_2, \quad (1)$$

where F_{CO_2} is sea–air flux of carbon dioxide, mmol·m^{-2}·day^{-1}; K_0 is CO_2 solubility, mol·m^{-3}·atm^{-1}; ΔpCO_2 is gradient between partial pressure of carbon dioxide in the sea surface and the near sea surface atmosphere, atm; k is gas transport rate, $m \cdot day^{-1}$, parameterized as a wind speed function:

$$k = 0.251 \cdot U^2 \cdot (Sc/660)^{-0.5},$$

where U is wind speed, $m \cdot s^{-1}$; Sc is Schmidt number; ratio 0.251 is empirically de-rived parameter, $cm \cdot h^{-1} \cdot (m \cdot s^{-1})^{-2}$ [19].

In [18], it has been established that the intensity of the carbon dioxide flux is determined by the state of the sea surface (bubbles, roughness) at wind speeds of more than 15 $m \cdot s^{-1}$. Wind speeds of more than 15 $m \cdot s^{-1}$ were not recorded during the 125th cruise. Thus, only wind speed and pCO_2 gradient were taken into account when assessing fluxes.

Results

In December 2022, the average wind speed was 4.2 ± 3.8 $m \cdot s^{-1}$ with its minimum of 0.7 $m \cdot s^{-1}$ and maximum of 8.2 $m \cdot s^{-1}$. The sea surface temperature varied within 9.6–14.1 °C with its average value of 13.04 ± 1.06 °C.

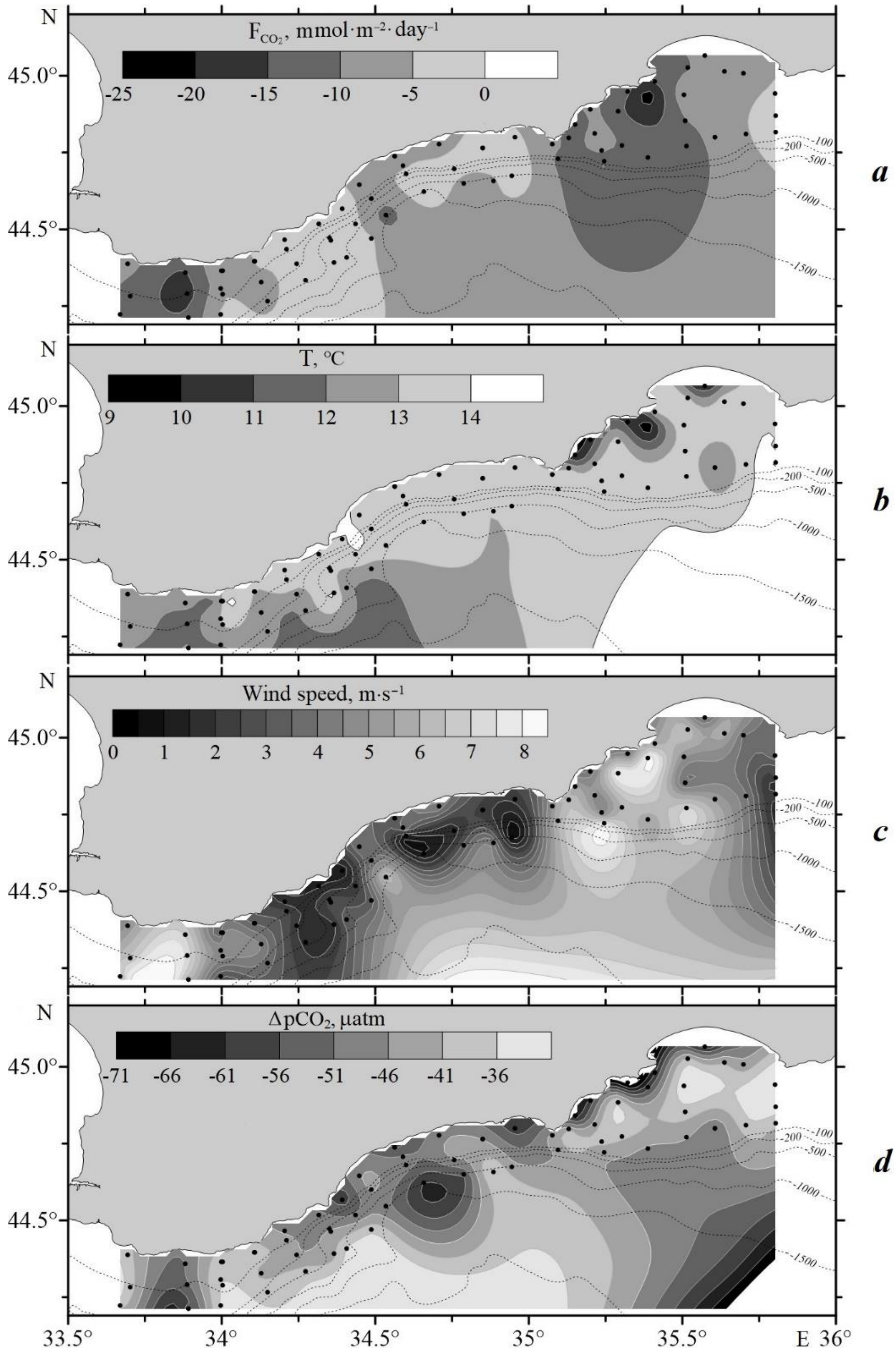
The average pCO_2 value of the sea surface was 388 ± 9 μatm while pCO_2 of the near sea surface atmosphere varied within a narrower range and the average value was 434 ± 4 μatm. Thus, the pCO_2 gradient between the sea surface and near sea surface atmosphere (ΔpCO_2) was predominantly determined by the pCO_2 variability in the sea surface. The values of ΔpCO_2 varied from –32.7 to –70.90 μatm with its average of –45.64 ± 8.56 μatm. It can be noted that the sea surface was undersaturated with carbon dioxide relative to the atmosphere during the period under study.

Based on the data obtained from equation (1), the CO_2 flux values were calculated.

The CO_2 flux intensity varied over a wide range from –0.04 to –8.74 $mmol \cdot m^{-2} \cdot day^{-1}$, the average value being –2.11 ± 1.79 $mmol \cdot m^{-2} \cdot day^{-1}$. Negative flux values indicate that the Black Sea waters absorb CO_2 from the atmosphere serving as its stock during the period under study. The calculated flux values are consistent with previously obtained data concerning the waters of the Crimean coast [14] and of the European shelf northwestern part [5].

Spatial variability of CO_2 flux values was characterized by heterogeneity (Fig. 2, *a*). Local minimum values and maximum flux intensity were observed in the area of the eastern coast of Crimea, as well as in its southern part (Fig. 2, *a*).

In terms of quality, the CO_2 flux spatial variability coincides with the distribution of temperature, wind speed and ΔpCO_2 in the sea surface (Fig. 2). Minima of temperature and ΔpCO_2 of the sea surface, as well as maximum wind speed were observed in zones of maximum intensity and minimum flux (Fig. 2).



F i g . 2 . Spatial variability of the sea-air CO_2 flux (*a*), temperature (*b*), wind speed (*c*) and gradient of pCO_2 (*d*) by data of the 125th cruise of R/V *Professor Vodyanitsky*

Discussion of results

It is known that the CO_2 flux value depends on wind speed and ΔpCO_2 to the greatest extent [18, 19].

Analysis of our data showed that the CO_2 flux was determined primarily by wind speed in December 2022 (Fig. 3). The correlation ratio (–0.93, it is statistically significant with probability belief $p = 0.99$) indicates a strong linear relationship. The relationship is inverse in itself. Flux direction determines ΔpCO_2 between the sea surface and the near sea surface atmosphere. In turn, ΔpCO_2 is determined by the ratio of the partial pressure of CO_2 in the atmosphere and the equilibrium partial pressure of CO_2 in the sea surface.

The pCO_2 value of the sea surface is proportional to the concentration of CO_2 in water. The concentration of CO_2 depends on the biogeochemical factor when the production or removal of CO_2 occur due to the transformation of organic matter and the formation of carbonates, proceeding according to the following equations:

$$6CO_2 + 6H_2O \leftrightarrow 6H^+ + 6HCO_3^- \leftrightarrow C_6H_{12}O_6 + 6O_2,$$

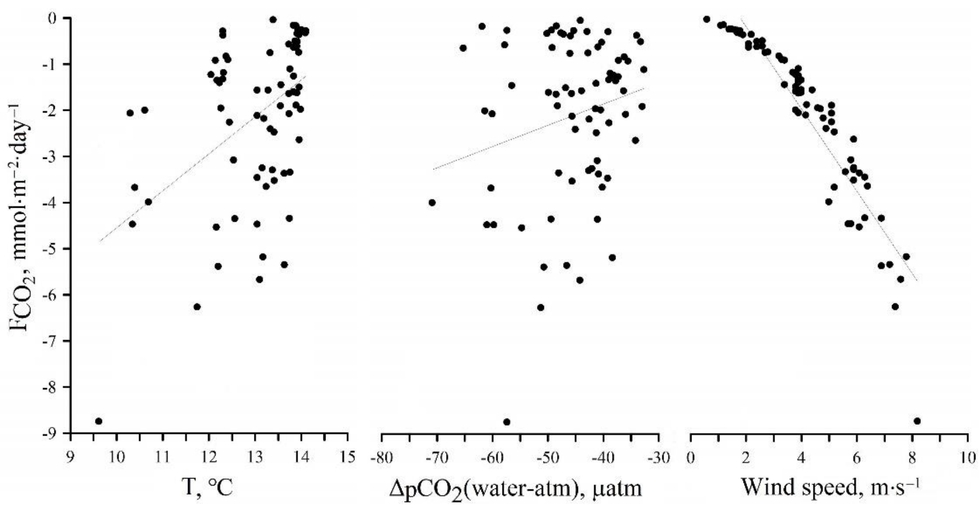
$$Ca^{2+} + 2HCO_3^- \leftrightarrow CaCO_3 + CO_2 + H_2O\,.$$

In addition, the CO_2 content in the sea surface depends on temperature which affects not only the CO_2 solubility, but also the intensity of biological processes, as well as the shift in chemical equilibria in the carbonate system [19]:

$$CO_{2(g)} \leftrightarrow CO_{2(aq)} \leftrightarrow CO_{2(aq)} + H_2O \leftrightarrow H^+ + HCO_3^- \leftrightarrow 2H^+ + CO_3^{2-}.$$

Changes in CO_2 concentration can also be caused by water dynamics, in particular by the CO_2 influx with waters from underlying layers [20].

Therein, the weak correlation of the CO_2 flux with ΔpCO_2 was unexpected (correlation ratio 0.22, it is statistically significant with probability belief $p = 0.95$).



F i g . 3 . Dependence of CO_2 flux (F_{CO_2}) on temperature, ΔpCO_2 and wind speed

A decrease in ΔpCO_2 is characterized by a decrease in the flux (Fig. 3). In turn, a decrease in ΔpCO_2 indicates a decrease in the difference between pCO_2 of the sea surface and the near sea surface atmosphere. As pCO_2 of the near sea surface atmosphere showed almost no changes during the period under study (fluctuation range ±1 %, average pCO_2 = 434 µatm), the decrease in the difference is due to an increase in pCO_2 and, accordingly, in the CO_2 concentration in the sea surface.

An increase in the CO_2 concentration in the sea surface at its low temperatures (about 13 °C) can be caused either by an increase in the CO_2 solubility with a decrease in temperature, or by the dynamics of water ensuring the CO_2 influx from underlying water layers, as well as by the decomposition of organic matter formed during the autumn blooming [12, 21].

The correlation of the CO_2 flux with the sea surface temperature was moderate enough (correlation ratio 0.47, it is statistically significant with probability belief $p = 0.99$). The sea–air intensity of the CO_2 flux decreased with increasing temperature (Fig. 3). However, since the intensity of the flux is also affected by ΔpCO_2 in addition to the wind speed, in this case it is advisable to consider the absolute values (to modulo) of the flux which determine its intensity. Thus, it should be noted that an increase in temperature leads to a decrease in ΔpCO_2 and, accordingly, a decrease in CO_2 flux during the cold season.

Therefore, we can conclude that in December 2022, the predominant contribution to the intensity of the flux is made by the wind speed while the direction of the CO_2 flux is determined by the difference in pCO_2 between the sea surface and the near sea surface atmosphere.

Conclusions

The waters of the northeastern part of the Black Sea serve as a stock of atmospheric CO_2 during the cold season.

According to the direct measurements of pCO_2 in the sea surface and in the near sea surface atmosphere, the values of the CO_2 flux in December 2022 varied widely from –0.048 to –8.74 $mmol \cdot m^{-2} \cdot day^{-1}$, the average value being –2.11 ± 1.79 $mmol \cdot m^{-2} \cdot day^{-1}$. At the same time, no pronounced features of spatial variability were identified. Local minima of flux values were observed in the eastern and southern regions of the Crimean peninsula.

In terms of quality, the CO_2 flux spatial variability coincided with the distribution of temperature, wind speed and ΔpCO_2.

When analyzing the correlation of the CO_2 flux with temperature, wind speed and ΔpCO_2, the strongest relationship was found with wind speed (–0.93), while the weakest one was with ΔpCO_2 (0.22). When the wind speed increases, an increase in the intensity of the CO_2 flux is observed, while the direction of the CO_2 flux is determined by ΔpCO_2 and, accordingly, by the value of pCO_2 and the CO_2 concentration in the sea surface.

The measurements were carried out at the Center for Collective Use R/V *Professor Vodyanitsky* of A.O. Kovalevsky Institute of Biology of the Southern Seas of RAS.

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