Original article

Estimation of Carbon Dioxide Fluxes through the Surface of the Black Sea from Numerical Simulation Results

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Abstract
Based on numerical simulation, the paper studies the spatiotemporal distribution of CO_2 fluxes through the free surface of the Black Sea. The basic equation for solving this problem is the three-dimensional evolutionary transport–diffusion equation for the concentration of dissolved inorganic carbon. The simulation uses hydrodynamic fields resulting from a previous physical reanalysis as input parameters. A model of the lower level of the Black-Sea ecosystem food chain is used to describe the influence of biological factors on the dissolved carbon dioxide distribution. The concentration and equilibrium partial pressure of dissolved carbon dioxide in the surface layer of the Black Sea were calculated from the numerical simulation results. It is shown that the time dependence of these quantities is highly seasonal. The seawater temperature significantly affects the solubility of carbon dioxide and therefore its fluxes. The equilibrium partial pressure of carbon dioxide averaged over the area of the Black Sea is minimal in January–February and maximal in June–July. Accordingly, in the warm season, the flux of carbon dioxide is directed mainly from the sea to the atmosphere; in the cold season, the sea mainly absorbs carbon dioxide. Biological factors also influence the CO_2 content in the sea. Thus, at the beginning of the year, a high concentration of phytoplankton is observed almost throughout the entire Black Sea water area, which is why the absorption of carbon dioxide predominates during photosynthesis. In summer, the release of carbon dioxide predominates due to plankton respiration and oxidation of organic matter. The simulation results are in fairly good agreement with *in situ* measurements of the partial pressure of dissolved carbon dioxide obtained during scientific cruises.

Keywords: carbonate system, carbon dioxide, partial pressure of carbon dioxide, Black Sea, marine ecosystem, numerical simulation

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Оценка потоков углекислого газа через поверхность Черного моря по результатам численного моделирования

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Аннотация
На основе численного моделирования изучается пространственно-временное распределение потоков CO_2 через свободную поверхность Черного моря. Основным уравнением для решения этой задачи является трехмерное эволюционное уравнение переноса – диффузии для концентрации растворенного неорганического углерода. При моделировании в качестве входных параметров используются гидродинамические поля, являющиеся результатом проведенного ранее физического реанализа. Для описания влияния биологических факторов на распределение растворенного углекислого газа используется модель нижнего уровня пищевой цепи экосистемы Черного моря. По результатам численного моделирования были рассчитаны концентрация и равновесное парциальное давление растворенного углекислого газа в поверхностном слое Черного моря. Показано, что зависимость от времени этих величин носит выраженный сезонный характер. На растворимость углекислого газа и, следовательно, на его потоки существенно влияет температура морской воды. Осредненное по площади Черного моря равновесное парциальное давление углекислого газа минимально в январе – феврале и максимально в июне – июле. Соответственно в теплый сезон поток углекислого газа направлен преимущественно из моря в атмосферу, в холодный сезон море в основном поглощает углекислый газ. На содержание CO_2 в море влияют также биологические факторы. Так, в начале года почти по всей акватории Черного моря наблюдается высокая концентрация фитопланктона, из-за чего преобладает поглощение углекислого газа в процессе фотосинтеза. Летом преобладает выделение углекислого газа вследствие дыхания планктона и окисления органического вещества. Результаты моделирования достаточно хорошо согласуются с натурными измерениями равновесного парциального давления растворенного углекислого газа, полученными в ходе научных рейсов.

Ключевые слова: карбонатная система, углекислый газ, парциальное давление углекислого газа, Черное море, морская экосистема, численное моделирование

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Introduction

A large number of papers (e. g. [1–7]) have been devoted to the study of the Black Sea carbonate system and, in particular, the flux of carbon dioxide through the surface. These studies are mainly based on measurement data. The disadvantage of these data is that they are limited in space and time. To obtain spatial distributions and temporal evolution of carbonate system components, it is necessary to use numerical three-dimensional models of the marine environment. Recently, numerical simulation of marine ecosystems has become widespread, including the Black Sea ecosystem simulation [8–11]. As an example of a model for the entire world ocean, the PISCES model can be mentioned [12]. Numerical ecosystem models describe the evolution of biological and hydrochemical fields as well as the carbonate system elements. They advantage the possibility of obtaining a sequence of the marine environment parameters on a regular grid. Of course, simulation results cannot replace *in situ* measurements, but they can extrapolate their results over time and over wider spatial areas. This paper presents preliminary results of estimation of carbon dioxide fluxes between the Black Sea and the atmosphere based on numerical simulation.

Method of study

Quantitative estimation of carbon dioxide fluxes through the sea–atmosphere interface is one of the main tasks in the study of the carbon cycle. This paper solves this problem by numerical simulation. The basic equation is three-dimensional evolutionary transport–diffusion equation for the dissolved inorganic carbon concentration

$$\frac{\partial C}{\partial t}+\frac{\partial (uC)}{\partial x}+\frac{\partial (vC)}{\partial y}+\frac{\partial (wC)}{\partial z}=K_h\nabla^2 C+\frac{\partial}{\partial z}\left(K_v\frac{\partial C}{\partial z}\right)+R\,, \qquad (1)$$

Where u, v, w are current velocity components; K_h, K_v are coefficients of horizontal and vertical turbulent diffusion, respectively. Representing the coefficients of equation (1), these fields, are provided by the Black Sea circulation model. Summand R in the right-hand side of equation (1) has the form $R = Res - upt + Ox$, where Res describes the input of carbon dioxide due to the respiration of all plankton species; – upt describes reduction of dissolved inorganic carbon from photosynthesis during primary production and Ox – its input due to oxidation of suspended organic matter [13].

An existing three-dimensional model of the lower level of the Black Sea ecosystem food chain is used to calculate these values [14, 15]. From the mathematical point of view, the biogeochemical part of the model represents a system of fifteen (according to the number of state variables) transport-diffusion equations similar to equation (1). The summands in the right-hand sides of this system describe biogeochemical interactions among the state variables of the ecosystem model. A view of these sources for the ecosystem model is presented in [15]. Variable units are converted from nitrogen to carbon using the C:N ratios for the Black Sea taken from [16].

The connection of the circulation model with the biogeochemical part is one-way in this work. That is, current velocity fields, temperature, salinity and turbulent diffusion coefficients obtained from the hydrodynamic model in advance are then used to calculate the biogeochemical model parameters and in equation (1). The calculation domain for equation (1) and the biogeochemical part of the model coincides horizontally with the corresponding domain for the circulation model (grid steps equal to 4.8 km coincide, accordingly) and occupies vertically the upper 200 m layer of the Black Sea. At the same time, the computational horizons correspond to the circulation models. The results of a 28-year physical reanalysis of the Black Sea fields (1993–2020) were used as hydrodynamic fields in this work [17].

To obtain the initial fields, the calculation for 2017 was carried out in a cycle in which the input parameters of the ecosystem model (current velocity, temperature and salinity fields for 2017) were taken from the reanalysis. Once the biogeochemical fields reached the stationary regime, the calculation was terminated and the obtained fields were used as initial fields for the main calculation.

Dissolved CO_2 concentration was calculated from the obtained dissolved inorganic carbon fields using formula

$$[DIC] = [\mathrm{CO_2}] \times \left\{ 1 + \frac{K_1}{[\mathrm{H^+}]} + \frac{K_1 K_2}{[\mathrm{H^+}][\mathrm{H^+}]} \right\},$$

where effective dissociation constants of carbonic acid K_1, K_2 depend on seawater temperature and salinity. Then its equilibrium partial pressure is determined from the concentration of dissolved carbon dioxide using Henry coefficient K_0 by formula $[\mathrm{CO_2}] = K_0\, p\mathrm{CO_2}$ [18]. The Henry coefficient is not a constant as it depends on seawater temperature and salinity.

To calculate the concentration of dissolved carbon dioxide from the total inorganic carbon concentration in water, it is necessary to know the concentration of hydrogen ions in addition to coefficients K_1, K_2. To estimate the fluxes of carbon dioxide through the sea surface, only surface values of carbon dioxide concentration are necessary. In this work, the pH parameter was not calculated by the model, but approximated in time and space according to the data from atlas [1)] containing maps of pH distribution on the Black Sea surface for four seasons.

The partial pressure of carbon dioxide in the atmosphere surface layer was assumed to be constant and equal to 410 µatm. The carbon dioxide flux between the sea and the atmosphere was calculated by formula $F = Tr\,(\mathrm{pCO}_{2w} - \mathrm{pCO}_{2a})$, where Tr is carbon dioxide transport coefficient between the sea and the atmosphere; pCO_{2w} and pCO_{2a} are partial pressure values in water and in the atmosphere. The value

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[1)] Mitin, L.I.. ed., 2006. *Atlas of the Black Sea and Sea of Asov Nature Protection.* Saint Petersburg: GUN i O, 436 p. (in Russian).

of coefficient *Tr* was chosen to be 0.5 gC·m^{-2}·µatm^{-1}·month^{-1} (the World Ocean average according to [19]).

Results

The main calculation covered four years from 2017 to 2020. Fig. 1 shows the evolution of basin area averaged concentration of carbon dioxide at the sea surface and its partial pressure obtained from simulation results. Temporal dependence is highly seasonal. Dissolved carbon dioxide pressure values are minimal around January–February and maximal around June–July. When the partial pressure of carbon dioxide dissolved in seawater exceeds the pressure in the atmosphere, the flux is directed from the sea to the atmosphere through the free surface, and vice versa. Thus, it follows from the graph of pCO_2 change that invasion is observed on average in the Black Sea water area during cold times (approximately from October to April) because the partial pressure of CO_2 dissolved in the sea is lower than the partial pressure in the atmosphere. On the contrary, evasion is observed on average during the warm period (approximately from April to October).

The maxima and minima on the graph of temporal variability of carbon dioxide concentration in the Black Sea surface layer do not coincide with the corresponding extremes on the graph of pressure. They are shifted by about three months. This is stipulated by the fact that the Henry constant, which relates the values of CO_2 concentration in the sea and its equilibrium partial pressure, depends, among other things, on the sea water temperature, which varies considerably during the year.

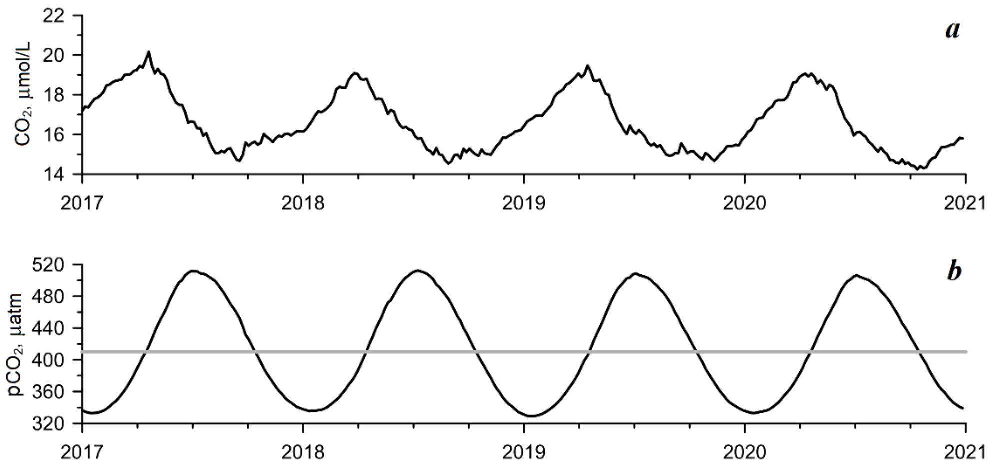
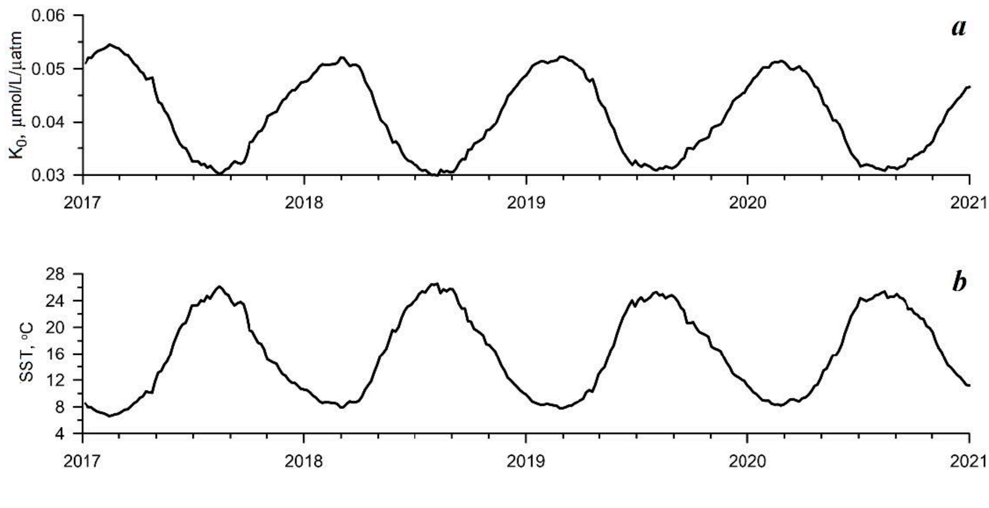


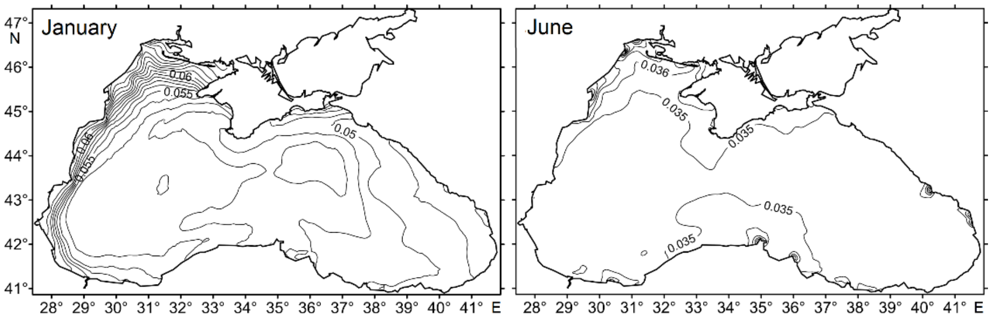
Fig. 1. Evolution of the average over the basin area concentration of carbon dioxide at the sea surface (*a*) and its partial pressure (*b*) obtained from simulation results. The straight line shows the CO_2 partial pressure in the atmospheric surface layer

At the same time, the range of fluctuations of the basin area averaged concentration of carbon dioxide (approximately 25 % of the maximum value) is significantly smaller than the range of pressure fluctuations. The graphs (Fig. 1) also show the inverse dependence of dissolved carbon dioxide concentration on partial pressure clearly: the CO_2 concentration increases in those periods of time when the CO_2 flux is directed from the atmosphere to the sea, and vice versa, the concentration decreases when the flux is directed to the atmosphere.

Figure 2, *a* shows the graph of the time dependence of the sea area averaged value of the Henry coefficient. The temperature and salinity fields obtained from the reanalysis of the Black Sea hydrodynamic fields were used to calculate the value of the coefficient at each grid point. The variability of the Henry coefficient is also highly seasonal and is almost antiphased to the change in the carbon dioxide partial pressure (see Fig. 1, *b*). Over the course of a year, the area average of the Henry coefficient changes by almost a factor of two. Figure 2, *b* shows the graph of sea surface temperature evolution. These two graphs change in antiphase and it can be concluded that the variation of the sea area averaged value of the Henry coefficient is determined mainly by the sea water temperature. That is, the main contribution to the intra-annual variability of the CO_2 partial pressure in the sea surface layer is made by hydrological factors (mainly water temperature). The partial pressure decreases as the water temperature decreases and increases as the temperature increases. Accordingly, as long as the dissolved CO_2 pressure is greater than the atmospheric one, the flux through the sea surface is directed towards the atmosphere which is accompanied by a decrease in the dissolved CO_2 concentration. Then, when the dissolved gas pressure becomes less than the atmospheric one, the flux through the surface changes direction with an increase in the dissolved carbon dioxide concentration.



F i g . 2 . Evolution of sea area averaged values of the Henry coefficient (*a*) and sea surface temperature (*b*)



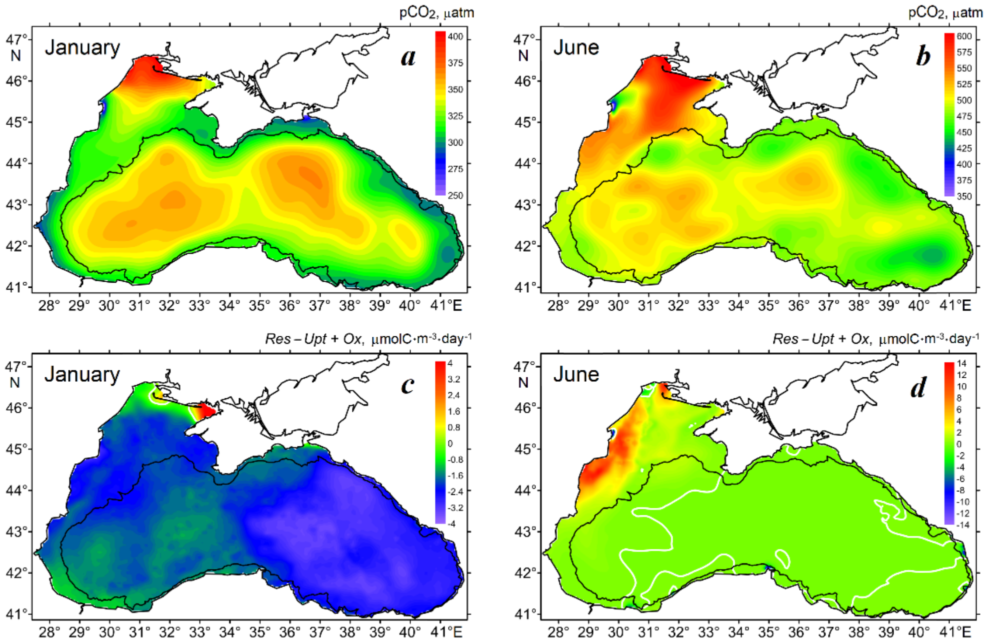
F i g . 3 . Spatial distributions of the Henry coefficient (μmol/L/μatm) for January and June 2017

Figure 3 shows the spatial distribution of the Henry coefficient. In summer, the distribution of the Henry coefficient is almost uniform over the entire water area except for the river confluences where the water is much freshened. In winter, the coefficient value is higher than in summer. In addition, an increase in the coefficient values is clearly visible in the north-western shelf (NWS) and along the western coast of the Black Sea. This is due to the fact that the NWS water is the coldest and most freshened in winter. The Black Sea Rim Current, intense in winter, carries this water along the western coast.

Figure 4, *a*, *b* provides an insight into the spatial distribution of the carbon dioxide partial pressure in the Black Sea surface layer. It shows the monthly average maps for two months, January and June 2017 (which corresponds to the minimum and maximum in Fig. 1, *b*). Spatial distributions of pCO_2 are similar for both months: the maxima are observed in the centre of the basin and in the NWS (only in its northern part in January). However, the average level varies significantly. Thus, the surface partial pressure of dissolved carbon dioxide is lower than the atmospheric pressure (410 μatm) in the entire water area in January and it is higher practically in the entire water area in June.

Biological processes also affect the dissolved carbon dioxide pressure distribution. Figure 4, *c*, *d* shows average monthly maps of the surface distribution of value $R = Res - upt + Ox$ describing the input of dissolved carbon dioxide from the plankton respiration and organic oxidation and its loss during photosynthesis for the same months.

In January, this value is negative almost over the entire water area (except for a small area in Karkinitski and Tendrovski Bays). A local maximum is identified at this location on the pCO_2 distribution map (Fig. 4, *a*). That is, the absorption of carbon dioxide prevails over its production due to biological processes in January. In June, most of the area has positive values, except for the central part of the sea, where it is close to zero.



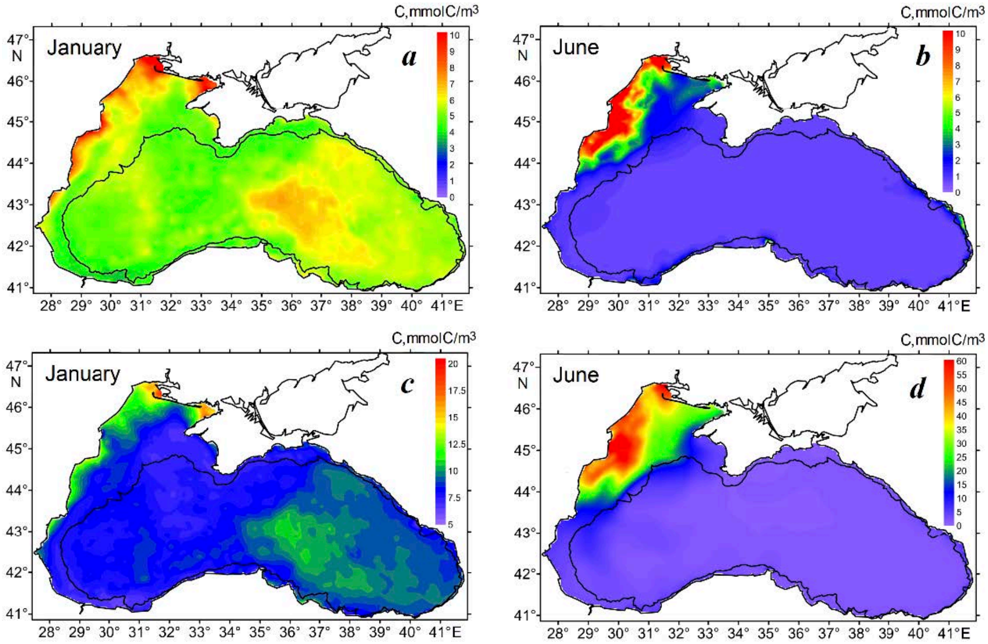
F i g . 4 . Spatial distribution of partial pressure of carbon dioxide in the surface layer of the Black Sea (*a*, *b*) and monthly average maps of the surface distribution of the *R* = *Res* – *upt* + *Ox* value (*c*, *d*) for January and June 2017 (the white line in Fig. 4, *d* indicates the zero isoline)

The predominance of carbon dioxide absorption over its production as a consequence of biological processes or vice versa is directly related to plankton concentrations in the sea upper layer. Figure 5 shows surface phytoplankton concentrations (*a*, *b*) and total plankton concentrations (*c*, *d*) for the same two months.

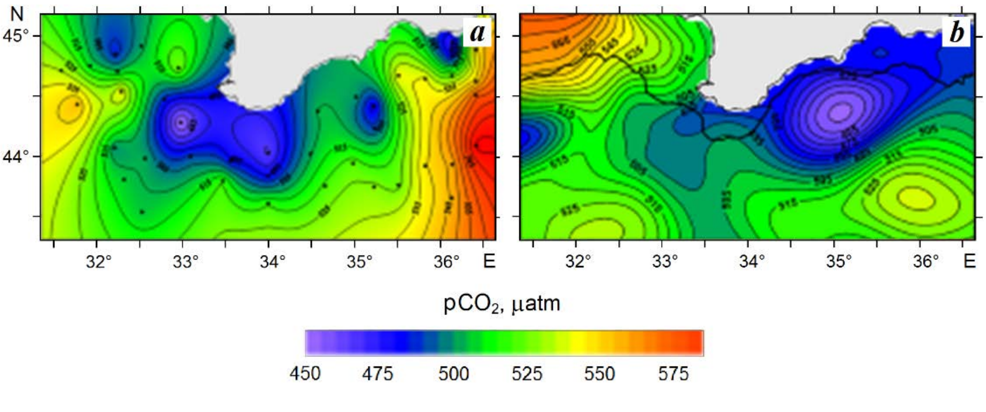
In January, the surface phytoplankton concentration is relatively high throughout the Black Sea area (Fig. 5, *a*), including its deep water. Total plankton concentration also shows high values (Fig. 5, *c*), but mainly due to phytoplankton. Accordingly, the absorption of carbon dioxide in the process of photosynthesis predominates in Fig. 5, *c*.

In June, concentrations of both phytoplankton and total plankton are low in the deep sea and high in the NWS (Fig. 5, *b, d*). Moreover, the total plankton biomass is significantly greater than the phytoplankton biomass in the NWS. Accordingly, Fig. 4, *d* shows that the CO_2 production caused by the plankton respiration dominates in the western Black Sea, especially in the NWS.

The obtained numerical simulation results were compared with observational data, unfortunately, few and local. Fig. 6 shows the pCO_2 distribution maps based



F i g . 5 . Spatial distribution of surface phytoplankton concentrations (*a*, *b*) and total plankton concentration (*c*, *d*) for January and June 2017



F i g . 6 . pCO_2 distribution maps based on observational data (*a*) and numerical simulation results (*b*) on observational data (Fig. 6, *a*) [20] and numerical simulation results (Fig. 6, *b*). The left map (Fig. 6, *a*) represents the result of processing samples taken at 132 stations during the 95th cruise of R/V *Professor Vodyanitsky* from 14 June to 4 July 2017.

In general, the values of the CO_2 equilibrium partial pressure near the sea surface obtained from the simulation results are quite close to the measurement data. The spatial distribution of pressure on two maps does not coincide, however, a characteristic decrease of pressure values near the Southern Coast of Crimea and Sevastopol is observed on both maps. It should also be noted that the map obtained from the model refers to a specific date (26 June), while the survey, on the results of which the left map is based (Fig. 6, *a*), lasted more than two weeks.

Figure 7 shows graphs of intra-annual variability of the Black Sea water area average values of equilibrium partial pressure of carbon dioxide pCO_2 obtained from measurements and simulation results averaged over four years. The left graph (Fig. 7, *a*) was kindly provided by the Marine Biogeochemistry Department of Marine Hydrophysical Institute. It is based on the processing of data obtained in 2015–2021 during the R/V *Professor Vodyanitsky* expedition studies of Marine Hydrophysical Institute. The location of stations is given in [7, p. 871].

Both graphs are relatively similar. Thus, the intra-annual variability of the Black Sea average equilibrium partial pressure of dissolved carbon dioxide in the sea surface layer is reproduced well by the model.

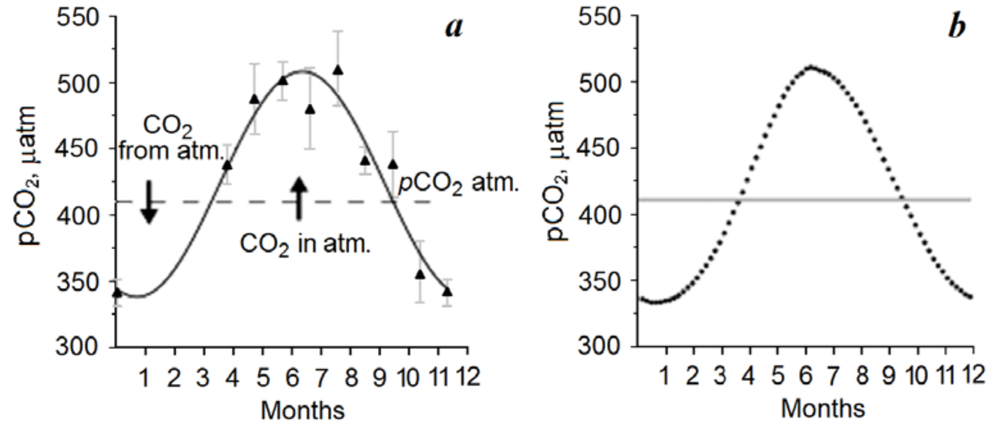


Fig. 7. Intra-annual variability of pCO_2 from observational data (*a*) and numerical simulation results (*b*)

Conclusions

Based on the results of numerical simulation, the time course and spatial distribution of such elements of the carbonate system as the concentration of dissolved carbon dioxide in the surface layer of the sea and its equilibrium partial pressure were obtained. The time course of these parameters is highly seasonal.

It has been shown that during the time period approximately from October to April, i. e. during the cold season, invasion is observed on average in the Black Sea water area because the equilibrium partial pressure of CO_2 dissolved in the sea is lower than the partial pressure in the atmosphere. Evasion occurs during the warm season, approximately from April to October, when pCO_2 in water is on average higher than in the atmosphere.

At constant partial pressure of CO_2 in the atmosphere (it changed during the year by 5 % according to the measurement data in 2017, according to atlas [1)]), the direction of carbon dioxide flux through the Black Sea surface is mainly influenced by seawater temperature. The equilibrium partial pressure of the dissolved gas decreases when the temperature decreases and increases with its increase. As long as the CO_2 pressure in the water is greater than the atmospheric one, the flux through the sea surface is directed towards the atmosphere, which is accompanied by a decrease in dissolved CO_2 concentration. When the dissolved gas pressure becomes lower than the atmospheric one, the flux through the surface reverses its direction, which is accompanied by an increase in dissolved carbon dioxide concentration.

The flux of carbon dioxide through the sea free surface is also influenced by biological processes. In winter, almost the entire Black Sea area is dominated by carbon dioxide absorption over carbon dioxide production due to high phytoplankton concentration near the Black Sea surface. In summer, however, carbon dioxide release predominates over most of the water area due to the plankton respiration.

The equilibrium partial pressure of dissolved carbon dioxide obtained as a result of simulation was compared with the data of hydrochemical surveys. The comparison showed a fairly good agreement between the results of numerical simulation and measurements.

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Viktor L. Dorofeev – general scientific supervision of the study, formulation of the goals and objectives of the study, carrying out numerical simulations, analysis of the obtained results and their interpretation

Larisa I. Sukhikh – literature review, numerical simulations, analysis of the results

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