

Research Article

Evaluation of Carbon Dioxide Partial Pressure and Air-Sea Fluxes in the Sea of Marmara

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Abstract

The increasing impact of global warming has made it crucial to monitor greenhouse gases (GHG). Carbon dioxide (CO_2) is especially important due to its significant increase. Oceans and seas play a vital role in absorbing excess CO_2 from the atmosphere, but they are facing challenges such as ocean acidification and damage to coral reefs. Therefore, it's essential to thoroughly understand climate change and its human-caused effects and to take necessary precautions or improve existing measures. In this direction, in seawater, determining alkalinity is crucial for monitoring CO_2 , along with other physical and chemical parameters, in order to help set carbon budgets and reduce environmental pressures. For this purpose, in this study in the Sea of Marmara, alkalinity and CO_2 measurements were conducted for the first time at stations CG2, CG3, MD26, and MD24. The CO_2 partial pressure (p CO_2) values ranged from 95 to 165 µatm at the surface of all stations, and total alkalinity values ranged from 1.075 to 1.46 mmol kg⁻¹ at specified depths. Based on these measurements, flux values were calculated between -1.10 and -5.39 mmol m² day⁻¹, indicating that the Sea of Marmara acted as a net CO_2 sink.

Keywords: Air-sea flux, carbon dioxide, partial pressure, seawater, Sea of Marmara

Introduction

Global warming is a term used to describe the impact of human activities on the climate, specifically the burning of fossil fuels (coal, oil, and gas). These activities cause the release of significant amounts of GHG: Geenhouse gases (the most important one is CO2) into the atmosphere (Houghton, 2005). Carbon dioxide demands attention as a GHG due to its indisputable rise in concentration. To understand what controls its hence on the greenhouse effect, all processes that affect and are affected by its concentration in the atmosphere need to be taken into account. These processes involve interactions with the chemically buffered carbonate system in seawater and with vegetation, due to their vital role in photosynthesis (Keeling, 1997).

In recent years, interest in the carbonate system in the oceans has increased due to the effect of CO_2 as a GHG. Carbon can be exchanged rapidly with the atmosphere through physical processes (such as CO_2 gas dissolving into the water) and biological processes (such as the growth, death, and decay of plankton) and is transported to the deep ocean by sinking so it can be stored for a longer time (Dioxide, 1985; Lal, 2009).The regulation of seawater pH and the cycling of CO_2 between the biosphere, lithosphere, atmosphere, and ocean are both vital functions of the carbonate system and the major part of the carbon in the oceans occurs as a part of this system (Millero, 1996).

The presence of oceans is not just a factor, but a crucial determinant in shaping the behavior of climate over time. Temperature dynamics as well as CO₂ behavior are both heavily influenced by the oceans, and the chemical aspect of CO₂ behavior is particularly significant (Lindzen, 1994). Oceans play a critical role in regulating climate change and atmospheric CO2; since the Industrial Revolution began, the ocean has absorbed an amount of carbon equivalent to approximately 40% of the anthropogenic carbon dioxide emitted by fossil fuel burning, and it is currently estimated that approximately 25% of anthropogenic CO₂ is absorbed by ocean annually (Gazioğlu et al., 2015; Takeshita et al., 2018). However, it also leads to fundamental changes in the chemistry of oceans. This is because CO₂ is also an acidic gas not only a GHG (Orr, 2011).

The pH of the ocean decreases as the amount of CO_2 dissolved in it increases, and this increase in CO_2 also leads to a decrease in the availability of carbonate (CO_2^{-3}) ions and reduces the saturation state of the major carbonate minerals that form shells (Kleypas et al., 2005). This decrease in pH and changes in fundamental chemical balances are often referred to as ocean acidification, which is also known as another CO_2 problem (Doney et al., 2009; Gazioğlu and Okutan, 2016).

In the last two decades, accurately measuring seawater CO_2 has become a priority for scientists who study how much CO_2 produced by human activities enters the ocean,

its impact on ocean chemistry, and its distribution (Dickson et al., 2007). So, it is important to understand the dynamics of this system, and is also important to understand the relationship of climate change with the ocean. So measuring at least two of these four parameters, which are total alkalinity, pH, total CO₂, and partial pressure of CO_2 (Millero, 1996), the carbonate system in the oceans can be characterized (because the concentrations of the individual species cannot be directly measured in the carbon dioxide system in a solution) (Millero, 1996; Millero et al., 2002; Dickson et al., 2007). Dissolved inorganic carbon (DIC) is present in seawater in three main forms: bicarbonate ion (HCO₃⁻), carbonate ion (CO_3^{2-}) , and aqueous carbon dioxide $(CO_{2(aq)})$, which also includes carbonic acid (H₂CO₃) (Fabry et al., 2008). The alkalinity of seawater plays a pivotal role in determining the proportion of the DIC in seawater that exists as CO₂. Alkalinity is the primary control on how much DIC seawater can hold (Henson et al., 2024). The CO_{2 (aq)} concentration depends upon the chemical composition and the temperature of seawater. Also CO₂ (aq) amount is proportional to the partial pressure of CO₂ exerted by seawater (Feely et al., 2001).

One of the main methods commonly used to estimate the air-sea carbon flux is to measure the difference in CO_2 partial pressure between the atmosphere and ocean and multiply it by an estimate of the gas exchange coefficient (Wanninkhof, 2009; Takeshita et al., 2018). The air-sea gas exchange has long been of scientific interest due to its importance in the biogeochemical cycling of climate, health-related gaseous compounds, and weather (Wanninkhof et al., 2009). The oceans are considered a significant sink for CO_2 . Therefore, gaining a better understanding of the net transport flux across the air-sea interface is crucial for comprehending the fate of this greenhouse gas emitted into the Earth's atmosphere (Takeshita et al., 2018).

In particular, the Marmara Sea, located within the borders of Turkiye, is exposed to various pressures both due to the influence of the densely populated provinces in which it is located and because it is the only waterway connecting the Mediterranean to the Black Sea through the Turkish Straits System (TSS). In addition, the carbon adsorption capacity of the Marmara Sea is not yet known. In this study, four different stations were selected in order to understand how the Marmara Sea, which has a geographically important location, reacts under the effects of GHGs, especially CO₂, and at what point it is in terms of carbon sequestration capacity, and to take the necessary measures to fight against global warming and climate change. As a preliminary study, alkalinity measurements were made for the first time, and based on these, partial pressure and flux calculations of carbon dioxide were made.

Materials and Methods Study Area

The Sea of Marmara is located between the European and Asian continents, connected to the Mediterranean by the Çanakkale Strait and to the Black Sea by the Istanbul Strait (Figure 1) (Unlulata et al., 1990; Besiktepe et al., 1993; Besiktepe et al., 1994). The Sea of Marmara, which forms the TSS together with the Istanbul and Çanakkale Straits, and is located between the coordinates of $40^{\circ}00'$ and $41^{\circ}1'N$ and $26^{\circ}15'$ and $29^{\circ}55'E$ (Ozturk, 2002). The coastline of the Sea of Marmara is 972 km, with a surface area of 11,500 km² and a volume of 3,378 km³ (Besiktepe et al., 1994; Polat et al., 1997; Ozturk, 2002).

There is a two-layer water system in the Marmara Sea; while the low salinity waters of the Black Sea (approximately 18‰) come from the Istanbul Strait to the Marmara Sea and form the upper layer, the salty water of the Mediterranean (approximately 38%) come from the Çanakkale Strait and form the lower layer water (Unlulata et al., 1990; Besiktepe et al., 1994). A sharp density interface formed between these two layers at a depth of approximately 25 m separates them (Besiktepe et al., 1994). In this system, the residence time of the upper layer waters is 4-5 months, while the residence time of the lower layer waters is 6-7 years (Besiktepe et al., 1994; Algan et al., 2004). Four stations were selected as sampling stations in the study area (Figure 1, Table 1): MD24Y, MD26, CG2, and CG3. CG2 and CG3 stations are located around Balıkcı Island, which is 2 km away from Buyukada (the largest of the Prince Islands), MD26 station is located near Heybeliada, and MD24Y station is located on the coast of Yalova.

Table 1. Latitude and the longitude informations of the stations.

Statiton ID	Latitudes (°N)	Longitudes (°E)
MD26	40.84083	29.05433
CG2	40.81633	29.10983
CG3	40.8225	29.1125
MD24Y	40.70266	29.33383

Sampling

Sampling was carried out via R/V Alemdar II research vessel that belongs to Istanbul University Marine Sciences and Management Institute in September and November 2023. Seawater samples were taken at the specified stations from 3 different depths: surface, intermediate layer, and bottom, with Niskin bottles (General Oceanics Model 1010 Niskin Water Sampler, 5L) connected to the CTD + rosette system. As specially, alkalinity measurements were taken directly from the Niskin bottles with a glass apparatus that would prevent the seawater samples from coming into contact with air in 100 ml glass volumetric flasks that were previously washed with nitrogen gas.

Physicochemical Analysis

The physicochemical parameters such as pH, temperature, dissolved oxygen (DO), and salinity were determined along the water column. Temperature, salinity, and oxygen were measured with sensors connected to the CTD+Rosette system. pH measurements were performed by a BENTA210 Benchtop pH/Mv meter. DO was also measured by the Winkler method on board (modified with CTD data), and water samples were taken directly from the Niskin bottles to prevent any biological activity and contamination from the atmosphere (APHA-AWWA-WPCF, 1985).



Fig. 1. The locations of the sampling stations in the Sea of Marmara.

Alkalinity measurement and calculation of partial pressure and flux of $\ensuremath{\text{CO}}_2$

The total alkalinity is determined by titrating the sample with a standard solution of a strong mineral acid. Methods using either an indicator or a pH-meter for the titration are described. For the titration, Metrohm Dosimat 876 Plus was used. Phenolphthalein and methyl orange were used as indicators. Also, a sulphuric acid solution was used as a strong acid (Suess, 1985; Hammer and Hammer, 2013; Dhoke, 2023).

For the calculation of phenolphthalein alkalinity as;

Alkalinity
$$(mmolL^{-1}as CaCO_3) = \frac{1000*A*c}{V}$$
 (Eq.1)

For the calculation of total alkalinity (A_t) as;

$$A_t(mmolL^{-1}as \ CaCO_3) = \frac{1000*B*c}{V}$$
(Eq.2)

In the given first and second equations respectively; where A is the amount of standard acid solution (mL) to reach the phenolphthalein endpoint or pH of the solution is 8.3. B is the amount of standard acid solution (mL) to reach the endpoint of methyl orange or pH of the solution is 4.5 or the second endpoint of titration. Also, c is the concentration of acid (mol L⁻¹), and V is the volume of the sample (mL).

To describe the CO_2 system in seawater, four parameters are measured pH, the total alkalinity (A_T), total carbon dioxide (C_t), and the (pCO₂) (Millero et al., 1993).

$$\begin{split} A_T &= A_C + [B(OH)_4^-] + [OH^-] - [H^+] \\ &= [HCO_3^-] + 2[CO_3^{2-}] + [B(OH)_4^-] + [OH^-] - [H^+] \\ &\quad (\text{Eq.3}) \end{split}$$

For the calculation of carbonate alkalinity (A_c) as;

$$A_{c} = A_{t} + \{H^{+}\} - \frac{K_{w}}{\{H^{+}\}} - \frac{B_{t}}{(1 + \frac{\{H^{+}\}}{K_{B}})}$$
(Eq.4)

where H^+ is the concentration of hydrogen ion (mmol kg⁻¹); K_w is ionization constant of water (mmol kg⁻¹); B_t is borate alkanity (mmol kg⁻¹); K_B is boric acid alkalinity constant (mmol kg⁻¹) (Körtzinger, 1999; Grasshoff et al., 2009).

$$C_t = [H_2CO_3] + [CO_2] + [HCO_3^-] + [CO_3^{2-}]$$
 (Eq.5)

Total carbon dioxide (C_t) is calculated based on the following empirical formula;

$$C_t = \frac{A_c \left(\frac{(H^+)^2}{K_1 + (H^+) + K_2}\right)}{(H^+) + 2K_2}$$
(Eq.6)

where K_1 is first dissociation constant of carbonic acid in seawater (mmol kg⁻¹); K_2 is second dissociation constant of carbonic acid in seawater (mmol kg⁻¹) (Körtzinger, 1999; Grasshoff et al., 2009).

pCO₂ is calculated by the following equation;

$$p(C0_2) = \frac{A_C \{H^+\}^2}{K_0 K_1(\{H^+\}+2K_2)}$$
(Eq.7)

The air-sea CO_2 fluxes (F, mmol m⁻² d⁻¹) is calculated using the below equation;

$$F = k \alpha \,\Delta p C O_2 \tag{Eq.8}$$

where k is the velocity of gas transfer (cm h⁻¹); α is CO₂ gas solubility in seawater (mmol L⁻¹ atm⁻¹); Δ pCO₂ is the variance between surface seawater and atmospheric pCO₂ (pCO_{2a}) (Sun et al., 2020).

The dry air atmospheric pCO_2 (pCO_{2a} , atm) is calculated by the following equation (Dickson et al., 2007; Takahashi et al., 2002; Takahashi et al., 2009; Padhy et al., 2016)

$$pCO_{2a} = xCO_{2a} (p_b - pH_2O)$$
 (Eq.9)

where xCO_{2a} is troposphere mole fraction (ppm); p_b is sea surface pressure (atm); pH_2O is saturated vapor pressure of water.

For the calculation of pH₂O as (Weiss and Price, 1980);

$$lnpH_2O = 24.4543 - 67.4509 \left(\frac{100}{T}\right) - 4.8489 \ln(\frac{T}{100}) - 0.000544 \text{ S}$$
(Eq.10)

where T is temperature (Kelvin), S is the salinity (‰).

For the calculation of the velocity of gas transfer related to wind speed as (Wanninkhof, 2014);

$$k = 0.251 U_{10}^2 \left(\frac{Sc}{660}\right)^{-1/2}$$
(Eq.11)

where Sc is the Schmidt number of CO₂, U_{10} is the wind speed (m s⁻¹) at 10 m height. Wind speed data are collected from the earth nullschool data website (Earth Nullschool, 2024) Sc was calculated according to the empirical formula of Wanninkhof (2014);

 $Sc = 2116.8 - 136.25 t + 4.7353 t^2 - 0.092307 t^3 + 0.0007555t^4$ (Eq.12)

where t is the temperature (°C).

Results

Temperature, salinity, and pH are important factors that affect the partial pressure of CO_2 in seawater (Kao et al., 2023; Mathis et al., 2012). pH can affect the solubility of CO_2 because it affects the carbonate balance in seawater. When the pH value increases, CO_2 partial pressure and flux decrease (Cao et al., 2023). The increase in water temperature affects the solubility of CO_2 gas. Increasing water temperature can increase primary production and thus reduce pCO_2 (Wang et al., 2017). In this study, alkalinity was measured in seawater, and calculations were made using temperature, salinity, and pH values. The physicochemical properties of the study area are shown in Figure 2. The method application determined in accordance with the purpose of the study was optimized for the Sea of Marmara.

When the September and November salinity and temperature (T-S) profiles of four stations are examined, the double-layer structure of the Marmara Sea is seen (Unluata, 1990; Besiktepe, 1994). In the T-S profiles of the MD24Y station located at the southern exit of the Gulf of Izmit, the upper layer of water originating from the

Black Sea is seen in the first 16-18 meters in both September and November measurements. The water mass with a salinity value of 30 psu, representing the Marmara Sea interface, is at ~19 and 21 meters in September and November, respectively. It is seen that the intermediate layer thickens in September compared to November due to the effect of vertical mixing (Muftuoglu, 2008).

The minimum dissolved oxygen (DO) located just above the Mediterranean water can be associated with organic material that has precipitated and accumulated in this layer. The presence of a cold intermediate layer remaining from the winter month in September is noteworthy. The presence of a cold intermediate layer is also seen in the other three stations, although it loses its prominence. The intermediate layer observed in November and the approximately 1°C excess temperature in the water column up to a depth of 60 meters indicate that the Mediterranean-origin water entering from the Çanakkale Strait in this month settled in this layer in the entire Marmara Sea due to its relatively low density.

At station MD26, which reflects similar characteristics to the MD24Y profile, a uniform structure in terms of both salinity and temperature is noticeable starting from 70 meters. The fluctuating structure in the DO value again becomes regular after 70 meters and decreases. The relatively warm surface water at stations CG2 and CG3, located in the south of the islands, contains less DO than the water immediately below, depending on the solubility (Muftuoglu, 2008).

In September, the highest A_c and C_t were measured at station MD26 in the bottom, respectively 1.42 mmol kg⁻¹ and 1.336 mmol kg⁻¹. The lowest values were observed at the same station but on the surface, respectively 1.07 mmol kg⁻¹and 0.924 mmol kg⁻¹. The highest pCO₂ was observed at the bottom water of CG3 station, 386 µatm, and the lowest pCO₂ was observed at the surface water of MD26 station, 95 µatm (Table 2). In November, the highest A_c and C_t were measured at station MD26 in the surface, respectively 1.46 mmol kg⁻¹ and 1.266 mmol kg⁻¹. The lowest values were 1.13 mmol kg⁻¹ and 1.051 mmol kg⁻¹ at the CG3 station in the intermediate layer, respectively.

The highest pCO2 was observed at the bottom water of MD26, 279 μ atm, and the lowest pCO2 was observed at the surface water of MD26 station, 109 μ atm (Table 3). In the results obtained, although alkalinity values varied between 1.07–1.46 mmol kg⁻¹, no major differences were found. The lowest pCO₂ value was observed at MD26 station in September (95 μ atm), and the highest value was observed at CG2 station in September (165 μ atm). However, the opposite situation exists at the MD26 station. It is thought that this may be due to the change in primary production between two months. It can be said that CO₂ uptake increased due to the fact that the weather conditions were harsher in November and the flux amount became more negative (Wanninkhof and Triñanes, 2017).



Fig. 2. The graphs show the temperature (°C), salinity (PSU), and Dissolved Oxygen (mg L^{-1}) values versus depth in the stations CG2, CG3, MD24Y, and MD26. The blue line represents the September data, and the red line represents November.

Sea of Marmara.								
Station ID	Sampling Depth (m)	Sampling Time	Ac (mmol kg ⁻¹)	Ct (mmol kg ⁻¹)	pCO ₂ (µatm)	pН	Flux (mmol m ⁻² day ⁻¹)	
MD26	0.5	September	1.07	0.924	95	8.23	-3.55	
MD26	19	September	1.28	1.212	280	7.86		
MD26	88	September	1.42	1.336	316	7.86		
MD24Y	0.5	September	1.19	1.065	143	8.10	-2.34	
MD24Y	19	September	1.19	1.100	190	7.98		
MD24Y	66	September	1.17	1.099	264	7.86		
CG2	0.5	September	1.35	1.213	165	8.10	-1.10	
CG2	19	September	1.36	1.276	276	7.91		
CG2	28	September	1.35	1.266	313	7.85		
CG3	0.5	September	1.35	1.199	138	8.15	-1.21	
CG3	19	September	1.36	1.294	339	7.82		
CG3	25	September	1.37	1.309	386	7.76		

Table 2. The Ac (mmol kg⁻¹), Ct (mmol kg⁻¹), pCO₂ (µatm), pH, and Flux (mmol m⁻² day⁻¹) values in September in the

Table 3. The Ac (mmol kg⁻¹), Ct (mmol kg⁻¹), pCO₂ (µatm), pH, and Flux (mmol m⁻² day⁻¹) values in November in the Sea of Marmara.

September

Station ID	Sampling	Sampling	Ac	Ct	pCO ₂	pН	Flux
	Depth (m)	Time	(mmol kg ⁻¹)	(mmol kg ⁻¹)	(µatm)		$(\text{mmol } \text{m}^{-2} \text{ day}^{-1})$
MD26	0.5	November	1.46	1.266	109	8.26	-5.39
MD26	16	November	1.33	1.180	135	8.13	
MD26	85	November	1.23	1.164	279	7.83	
MD24Y	0.5	November	1.41	1.253	137	8.18	-1.77
MD24Y	18	November	1.16	1.090	240	7.89	
MD24Y	60	November	1.21	1.086	140	8.08	
CG2	0.5	November	1.41	1.224	116	8.24	-4.26
CG2	18	November	1.34	1.221	183	8.03	
CG2	28	November	1.21	1.136	255	7.86	
CG3	0.5	November	1.44	1.260	114	8.24	-4.32
CG3	19	November	1.13	1.051	204	7.93	
CG3	28	November	1.25	1.173	263	7.86	

High C_t concentration was associated with saltier water, and low Ct was associated with less salty water (Sun et al., 2020). In this study, an increase in Ct towards the lower layer was observed at all stations in September (Figure 3). This is because saltier Mediterranean waters dominate the lower layer of the Sea of Marmara. However, in November, Ct decreased slightly towards the bottom layer at all stations. (Figure 3). Some of the organic matter entering the aquatic environment is biologically mineralized (Capelle et al., 2020; Mann et al., 2012). Another portion of the organic matter rapidly sinks to the bottom (seabed), which may mineralize into CO₂ if sufficient O₂ is available. Organic matter that remains unmineralized is buried in sediment (Capelle et al., 2020). The relatively slight decrease in Ct values from the surface to the bottom water at the study stations in November can be explained by the entry of high organic matter into the receiving environment this month and the fact that some of this organic matter was mineralized with O2 while some remained buried in the sediment without being mineralized.

As pH value decreases, pCO₂ increases. In September and November, pH values decrease towards the lower

layers and accordingly pCO2 values increase at all stations. However, a decrease in pCO2 value was observed at station MD24Y in November. The reason is that the lower layer pH value (8.08) was higher than the intermediate layer pH value (7.89) (Table 3). In the study conducted by Sun et al. in 2010, pCO₂ values (137 - 591 µatm) in Bering Sea surface water were higher than the values determined in this study (Sun et al., 2020). Kao et al. in 2018, pCO₂ values in surface water in the Southeast China Sea (305 - 325 µatm) were higher than the results of this study (Kao et al., 2023). CO₂ fluxes were calculated based on pCO2 and daily wind speeds for different stations. Since the pCO₂ in the atmosphere is higher than the pCO2 in water, the calculated flux values gave negative results (-1.10 to -5.39 mmol m⁻² day⁻¹) (Tables 2 and 3). The negative flux meaning is the net CO_2 exchange from the atmosphere to the sea (Zhai etal., 2005). So It was concluded that there was a transfer of CO₂ from the atmosphere to the sea at the time of sampling. The flux values of determined in the studies of Sun et al. (2010) and Kao et al. (2018), which is similar to the value determined in this study (Kao et al., 2023; Sun et al., 2020).



Figure 3. Ct (mmol kg-1) and pCO2 (µatm) distributions at the surface, intermediate, and bottom depths of different stations of the Marmara Sea in September (blue line) and November (orange line).

Conclusion

pCO2 is an important parameter needed to calculate airsea flux and gain information about ocean acidification. In this study, the distribution of pCO2 and air-sea flux in the surface waters of the Sea of Marmara was investigated in September and November 2023. To determine pCO2 and air-sea flow, alkalinity was measured in seawater and calculations were made using temperature, salinity and pH values. The method application determined in accordance with the purpose of the study was optimized for the Sea of Marmara. At the selected station in the Sea of Marmara CG2, CG3, MD26, and MD24Y for the first time alkalinity measurments and calculations of pCO2 and fluxes were done. According to the pCO2 and air-sea flux results obtained, it was concluded that the CO2 sink dominated in the Sea of Marmara at the sampling time. This study is important as it is the first study conducted in this context and contains the first data. It is one of the first steps taken to find out how the Sea of Marmara behaves in terms of CO2 uptake by showing its condition at the

specified stations and at the time the sampling was carried out. In the future, more frequent and long-term studies are required to evaluate the effects of the variability of physicochemical and biological parameters and global warming on the marine environment.

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