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DOI

[10.14710/ik.ijms.28.1.113-124](https://doi.org/10.14710/ik.ijms.28.1.113-124)

Publication date

2023

Document Version

Final published version

Published in

Ilmu Kelautan: Indonesian Journal of Marine Sciences

Citation (APA)

Muskananfolo, M. R., Latifah, N., Hartoko, A., Febrianto, S., & Winterwerp, J. C. (2023). The Partial Pressure of CO₂ and Fluxes of CO₂ in Semarang Bay, Indonesia. *Ilmu Kelautan: Indonesian Journal of Marine Sciences*, 28(1), 113-124. <https://doi.org/10.14710/ik.ijms.28.1.113-124>

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The Partial Pressure of CO₂ and Fluxes of CO₂ in Semarang Bay, Indonesia

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Abstract

The CO₂ characteristics of the Semarang Bay system on the Northern coast of Java were investigated to understand the roles in carbon dioxide flux. The flux of carbon dioxide at the sea surface between seawater and atmosphere is essential for mitigating atmospheric CO₂. The aims of this study were to assess the variation of CO₂ partial pressure seawater (pCO_{2sea}) and CO₂ flux and to analyse the water quality variable which has the larger influence on CO₂ flux in the waters of Semarang Bay. The variables in this study were temperature, pH, salinity, and DO. Different methods were adopted in the analysis of water samples: spectrophotometric to measure chlorophyll-a and phosphate and titration method to measure Total Alkalinity (TA) and Dissolved Inorganic Carbon (DIC). A CO₂ meter was used to measure the CO_{2atm}. The lowest CO₂ flux value was (1.86 mmol CO₂ m⁻² day⁻¹) during the morning period while the pCO_{2sea} reached 461.04 μatm and CO₂ flux 83.79 mmol CO₂ m⁻² day⁻¹ at night. At noon time, with increased temperature, pCO_{2sea} was 461.04 μatm, and CO₂ flux was 83.79 mmol CO₂ m⁻² day⁻¹. The high concentration of chlorophyll-a ranged between 3.55–4.11 mg.L⁻¹. This chlorophyll-a concentration has a negative correlation with CO₂ flux, and it was found that TA and DIC concentrations have no relationship with CO₂ flux. Based on PCA analysis, it was found that the variability of CO₂ flux in Semarang Bay is influenced by the variability of pCO_{2sea}, sea surface temperature and k_{wa} (affected by wind speed).

Keywords: CO₂ flux, pCO₂ seawater, Dissolved Inorganic Carbon, Semarang Bay

Introduction

The oceans have an essential role as the most significant anthropogenic CO₂ sink, larger than land-based sinks. CO₂ diffusing through the sea surface changes form becoming into dissolved carbon dioxide, carbonic acid, bicarbonate, and carbonate, which is then utilised by phytoplankton and other autotrophic organisms and converted into organic carbon during the process of photosynthesis. This mechanism is known as the biological pump. Marine and coastal ecosystems play an important role in the global CO₂ cycle by capturing and storing carbon and redistributing it (Nehren and Wicaksono, 2018). Through the biological pump, the oceans sequester large amounts of particulate organic carbon (POC) while sinking to the bottom of the waters (Cavan *et al.*, 2019). At sea level, CO₂ changes into *free aqueous* CO₂, carbonic acid (H₂CO₃), bicarbonate (HCO₃⁻) and carbonate (CO₃²⁻) (Borges *et al.*, 2014; Akhand *et al.*, 2021). The concentration of free aqueous CO₂ in seawater is proportional to the partial pressure of CO₂ in seawater (pCO_{2sea}). The difference between the partial pressure of CO₂ in seawater and in the atmosphere determines the direction of CO₂. The partial pressure of CO₂ shows more significant

ocean variations than the atmosphere. Thus the movement of CO₂ flux is controlled primarily by changes in ocean pCO₂ (Zeebe, 2012; Delaigue *et al.*, 2020). The partial pressure of CO₂ in seawater is determined by sea surface temperature and biological processes in the ocean, such as photosynthesis. Thus, these two parameters determine the sink (uptake) and source (release) of CO₂ by the ocean (Feely *et al.*, 2001; Cavan *et al.*, 2019). Various researches have been carried out to determine the effects of sea surface temperature (Takahashi *et al.*, 2002; Zhai *et al.*, 2007; Zhu *et al.*, 2009; Chierici *et al.*, 2009) and of biological processes (Takahashi *et al.*, 2002; Hülse *et al.*, 2017; Cavan *et al.*, 2019) on seawater pCO₂ and sea-atmosphere CO₂ flux distribution (Takahashi *et al.*, 1997; Takahashi *et al.*, 2002; Zhai *et al.*, 2007; Chierici *et al.*, 2009; Kartadikaria *et al.*, 2015; Li *et al.*, 2018; Brady *et al.*, 2019; Friedlingstein *et al.*, 2020).

Roy-Barman and Jeandel (2016), states that at low latitudes/equator the warm waters can reduce the solubility of CO₂ causing CO₂ to be released back into the atmosphere (source CO₂). At high latitudes with cold waters, the solubility of CO₂ is high which can increase the dissolution of gas into

seawater causing CO₂ absorption (sink CO₂) to occur. This is supported by other studies which conclude that high-latitude seas/oceans with four seasons and relatively cold temperatures function as CO₂ absorbers (Takahashi *et al.*, 1997; Takahashi *et al.*, 2002; Cai *et al.*, 2006; Yasunaka *et al.*, 2016; Arnone *et al.*, 2017; Ito *et al.*, 2018; Tian *et al.*, 2020) while low latitudes seas function as a source of CO₂ due to warm waters (Fitranti *et al.*, 2013; Kartadikaria *et al.*, 2015; Afdal, 2016; Yan *et al.*, 2018; Latifah *et al.*, 2020). Several studies in Indonesia, however, show that the sea can function as sinks of CO₂ (Fachri *et al.*, 2015; Latifah *et al.*, 2019). These studies focused on waters with seagrass and coral reef ecosystems. Studies of CO₂ flux in waters which do not have seagrass ecosystems and coral reefs surrounded by urban settlements and industries are still lacking. The waters of Semarang Bay do not have seagrass ecosystems, or coral reefs and are surrounded by many industries and settlements which necessitates a comprehensive study of the CO₂ flux. Seagrass meadows and coral reefs are essential for carbon storage representing a vital ecosystem service (Guerra-Vargas *et al.*, 2020). In coastal waters, tropical seagrass meadows are connected to coral reefs, and since the reef barrier disperses waves, which accelerates sediment accretion and avoids erosion. Therefore, coral reefs might enhance the seagrass meadows' capacity as a blue carbon sink. The first study to investigate the spatiotemporal distribution of CO₂ flux in the Java Sea (Wirasatrya *et al.*, 2020) which also the first study to examine the key parameters at the sea surface (i.e., SST, Chl_a, sea surface salinity, and surface wind) to obtain the

most influencing parameter for CO₂ flux in the Java Sea. This indicates a limited number of studies on CO₂ flux in the Java sea. Moreover, the once dense mangrove forests around the bay have largely been lost, and with them an important CO₂ sink. Therefore, a comprehensive study on the CO₂ flux in the bay is required.

The aims of the present study are 1) to determine the variation of CO₂ partial pressure (pCO₂) and CO₂ flux in the waters of Semarang Bay; 2) to analyze the water quality variables that have the most influence on CO₂ flux.

Materials and Methods

The research was conducted in August 2021 during the southeast monsoon, when the wind blows from the Australian to the Asian Continent (Muskananfolo *et al.*, 2021b). In Indonesia, including Semarang Bay, this is the dry season (Muskananfolo *et al.*, 2021a). The monsoon winds blow through the Maritime Continent or the Indonesian seas, mainly over the seas along the routes formed by the coastline of the large islands and the chains of the small islands (Alifdini *et al.*, 2021). In the Indonesian seas, the monsoon wind is a key factor affecting the regional climate and oceanographic conditions (Susanto *et al.*, 2006).

Field data collection was conducted on 21 August 2021 (3 stations), 22 August 2021 (4 stations), and 23 August 2021 (3 stations) (Figure 1.). Station selection was based on their representativeness of the entire Semarang Bay area.

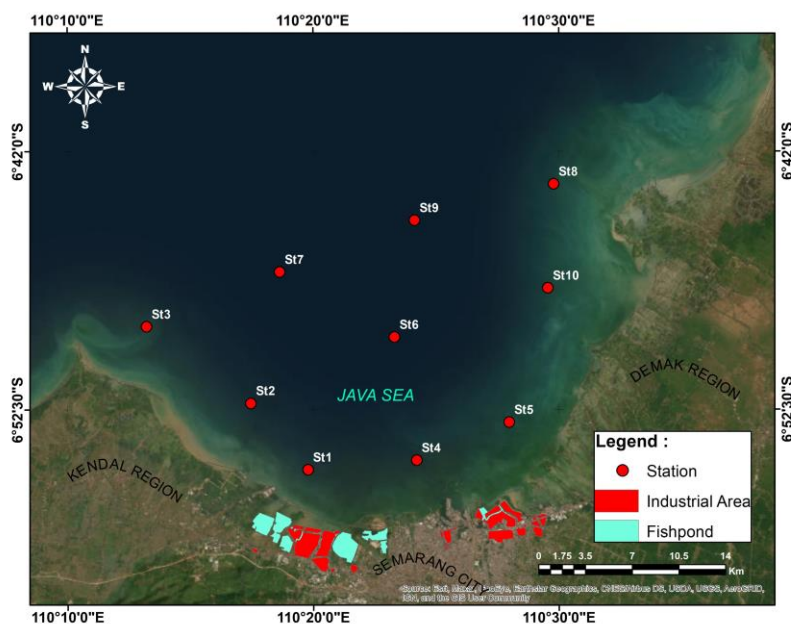


Figure 1. Study sites at Semarang Bay.

Sample collection

Using a Nansen Water Sampler, samples were collected near the sea water surface and put into several bottles: 1). 250 ml glass bottles (Dissolved Inorganic Carbon (DIC) and Total Alkalinity (TA) analysis using the titrimetric method); 2). 1,500 ml (chl-a analysis using the spectrophotometric method); and 3). 600 ml (nitrate and phosphate using the spectrophotometric method). Measurements of temperature, pH, and salinity were done using a DO meter and pH meter, measurement of Atmospheric CO₂ using a CO₂ meter, and water CO₂ using the titrimetric method.

Determination of the partial pressure of CO₂

Seawater CO₂ partial pressure was calculated using the following formula (Dickson et al., 2007):

$$fCO_2 = [CO_2] / K_o$$

$$pCO_2 \text{ sea } (\mu\text{atm}) = fCO_2 (\mu\text{atm}) * [1.00436 - 4.669 * 10^{-5} * SST(^{\circ}C)]$$

where [CO₂] is the seawater CO₂ concentration; K_o is the CO₂ solubility of seawater

The partial pressure of atmospheric CO₂ was calculated using the following formula (Dickson et al., 2007):

$$pCO_{2atm} = xCO_{2atm} (pb - pH_2O)$$

Where pb = air pressure (atm); xCO_{2atm} (ppm) is the molar fraction of CO₂ air in the troposphere obtained from https://disc.gsfc.nasa.gov/datasets/AIRX3C2_M_V005/summary, pH_{2O} is the water vapour pressure at the air-sea boundary obtained from Weiss et al. (1980):

$$pH_2O = \exp (24.4543 - 67.4509 \times (100/Tk)) - 4.8489 \times \ln (Tk / 100) - 0.000544 \times S$$

where: T is sea surface temperature (Kelvin); S is sea surface salinity (‰).

Estimation of the sea-air CO₂ Flux

Calculation of CO₂ flux or CO₂ gas flow exchange is calculated using a formula (Takahashi et al., 2002; Du et al., 2015; Fachri et al., 2015; Takahashi and Sutherland, 2017):

$$\text{Flux } CO_2 = K_h \times k_{wa} \times (\Delta pCO_2)$$

$$\Delta pCO_2 = pCO_{2sea} - pCO_{2atm}$$

Where Flux CO₂ is a carbon dioxide flux (mmol.m⁻².day⁻¹); K_h= solubility of CO₂ in mol L⁻¹ atm⁻¹ is derived from Weiss (1974); k_{wa} = CO₂ gas transfer velocity in m s⁻¹ using the RC01 and B04 formula (Raymond and Cole, 2001; Borges et al., 2004); ΔpCO₂= the partial pressure difference of CO₂ in the seawater and the atmosphere.

Results and Discussion

Partial pressure of CO₂

The partial pressure of carbon dioxide (pCO₂) is influenced by water temperature. The results of this study show that the pCO_{2sea} value in Semarang Bay in August 2021 ranges from 378–438 μatm with a temperature range of 28–35 °C (Figure 2.). The partial pressure of ocean carbon dioxide (pCO_{2sea}) increases with increasing water temperatures, with a correlation value of 0.88 (Figure 2.). Lakshmi (2000), stated that warm ocean waters , generally at low latitudes, reduced the solubility of CO₂, increasing the partial pressure of sea carbon dioxide (pCO_{2sea}), which is in agreement with the results of Figure 2. Temperature and coastal ecosystems (estuaries, mangroves, coral reefs and seagrasses) are the main factors that cause variations in pCO_{2sea} values and CO₂ flux (Afdal et al., 2020). Other studies (Arnone et al., 2017; Ito et al., 2018) found that relatively cold water temperatures reduced seawater pCO₂ and the sea acted as a CO₂ sink. The partial pressure of carbon dioxide (pCO₂) and primary ocean productivity are important keys in the biological pump mechanism.

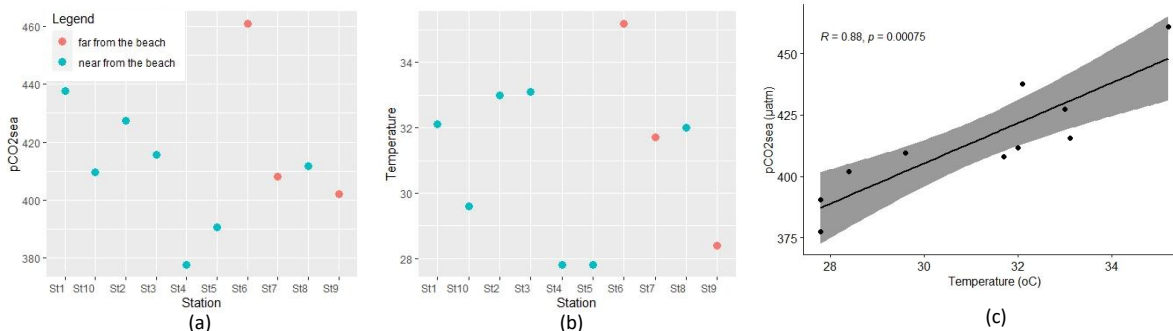
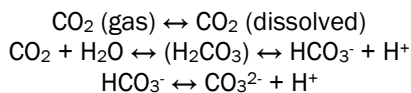


Figure 2. Distribution and relationship of pCO₂ sea and sea surface temperature

Variations in $pCO_{2\text{sea}}$ are caused not only by the sea surface temperature but also by the concentration of DIC (Dissolved Inorganic Carbon) and Total Alkalinity (TA). The present study, however, indicates that $pCO_{2\text{sea}}$ has no significant relationship with DIC or TA (Figures 3 and 4.). DIC in surface water is affected by freshwater input, biological transformations, the CO_2 exchange with the atmosphere and precipitation of calcium carbonate likewise with TA is mainly altered by the formation and dissolution of calcium carbonate (Honkanen et al., 2021). The precipitation and dissolution of calcium carbonate $CaCO_3$ affect both DIC and TA. Calcium carbonate is formed in a slow precipitation process by specific calcifying organisms such as coral reef (Zhang et al., 2023), certain types of phytoplankton (Honkanen et al., 2021), macroalgae in seagrass meadow (Macleadie et al., 2017; Howard et al., 2018). So, in this study in Semarang Bay, specific calcification organisms were not found. Therefore, maybe that's why DIC and TA have no significant effect on $pCO_{2\text{sea}}$. These results were also found in a study (Honkanen et al., 2021) in the Baltic Sea that DIC and TA do not change with related $pCO_{2\text{sea}}$ change by the hourly temperature change.



The concentration of DIC in surface waters ranges from $2,000 \mu\text{mol kg}^{-1}$, while the solubility of CO_2 does not exceed $23 \mu\text{mol kg}^{-1}$. In this study, DIC concentrations are less than $2,000 \mu\text{mol kg}^{-1}$ ranging from $1700\text{--}2000 \mu\text{mol kg}^{-1}$, with a solubility of CO_2 ranging $6\text{--}13 \mu\text{mol kg}^{-1}$. Most of the DIC is in the form of bicarbonate (87 %), and others in the form of carbonate (10 %) and carbon dioxide (0 %) (Figure 5.). In seawater, the composition of dissolved CO_2 is only 1% of the total DIC, while the most abundant are bicarbonate ions (HCO_3^-) at 89% and carbonate ions (CO_3^{2-}) at 10% (Roy-Barman and Jeandel 2016). According to Williams and Follows (2011), most of the carbon dioxide in the ocean was in the form of 90% bicarbonate ions, 9% carbonate ions and the remaining 1% as dissolved carbon dioxide. DIC (dissolved inorganic carbon) is more than 99% composed of bicarbonate and carbonate ions, which can increase the capacity of the oceans as sinks/reservoirs/storage of carbon more than 100 times to $38,000 \text{ Pg C}$.

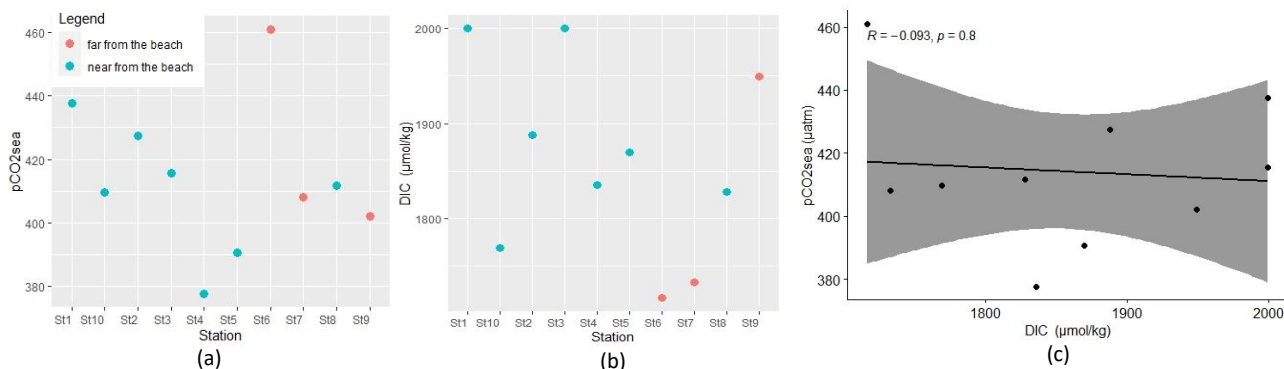


Figure 3. Distribution and relationship of $pCO_{2\text{sea}}$ and DIC (Dissolved Inorganic Carbon)

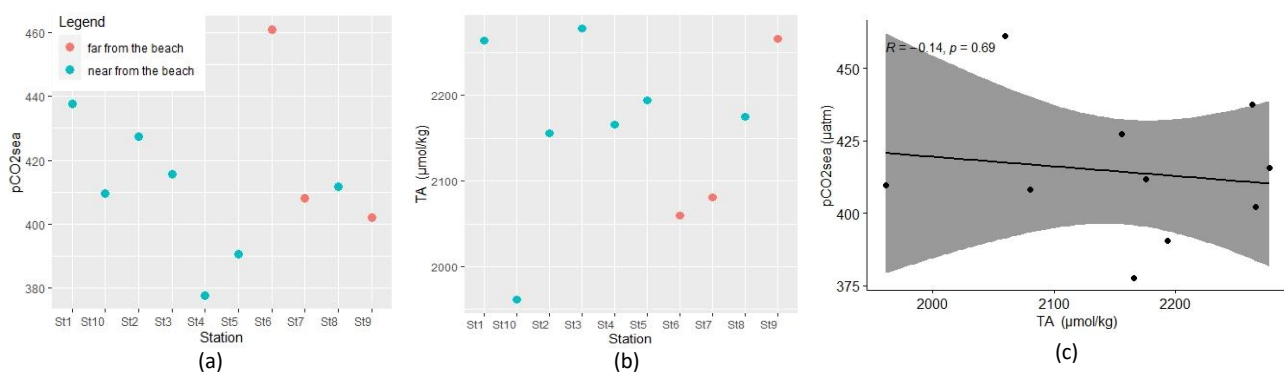


Figure 4. Distribution and relationship of $pCO_{2\text{sea}}$ and TA (Total Alkalinity)

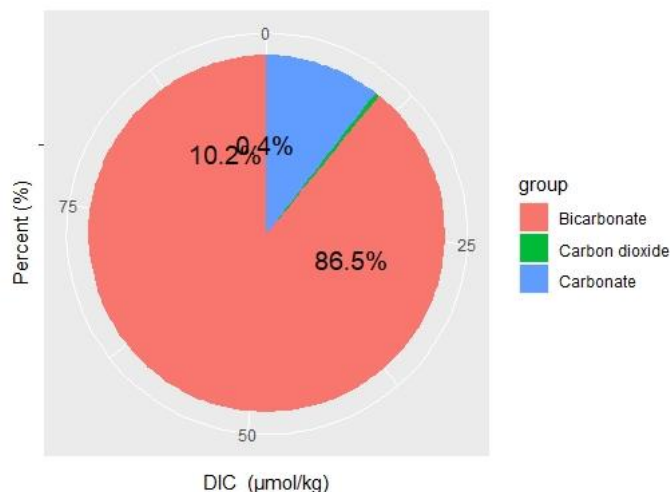


Figure 5. Average composition of the DIC distribution

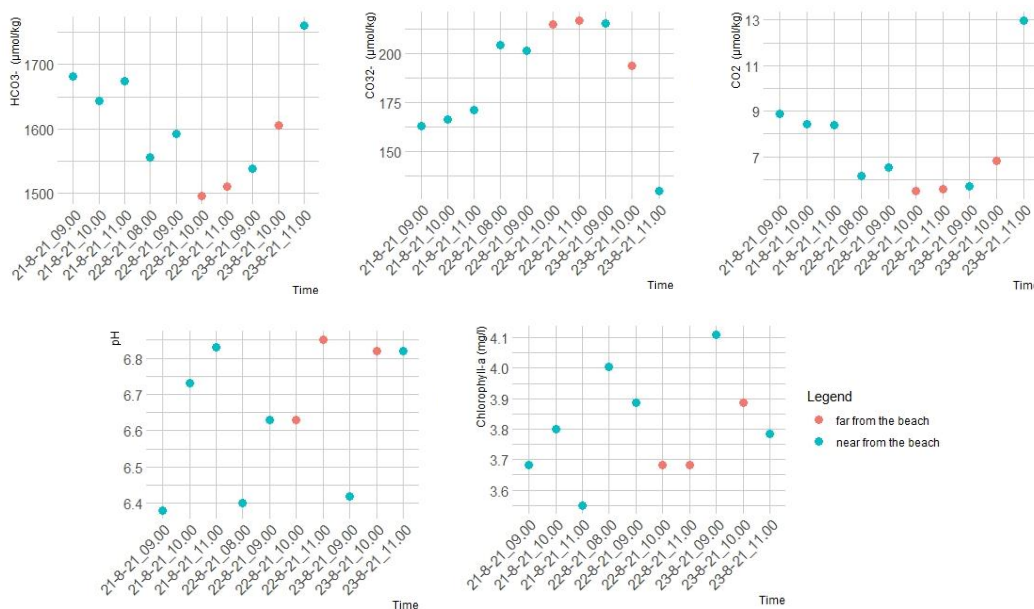


Figure 6. The concentration of bicarbonate, carbonate, carbon dioxide, pH and chlorophyll-a

Figure 6 shows that the smaller the HCO_3^- the higher the pH and vice versa. This is due to changes in the composition of phytoplankton in waters that utilise bicarbonate (HCO_3^-). The high chlorophyll-a content in the waters represents the abundance of phytoplankton in this case. The high chlorophyll-a concentration indirectly indicates the quantity of phytoplankton, causing more consumption of HCO_3^- bicarbonate (for the photosynthesis process). So, smaller HCO_3^- in the waters will reduce the number of H^+ ions and increase the pH. According to Williams and Follows (2011) and Millero (2010), a decrease in pH which is caused by an increase in consumption of HCO_3^- will reduce the number of H^+ ions and increase the pH.

Alkalinity is the ability of water to neutralize acids (Acid-Neutralizing Capacity/ANC) due to the presence of anions in water that can neutralize hydrogen cations. Alkalinity is also called buffer capacity because of its ability to buffer changes in the pH of the seas. According to Effendi (2003), the constituents of water alkalinity consist of hydroxide anions (OH^-), carbonates (CO_3^{2-}), silicates (HSiO_3^-), bicarbonates (HCO_3^-), sulfides (HS^-), borates (H_2BO_3^-), and phosphates (HPO_4^{2-} and H_2PO_4^-). In natural waters, the high constituent anion of alkalinity is bicarbonate which is the result of the ionization of carbonic acid, especially in waters with a high concentration of carbon dioxide. TA concentrations in this study ranged from 1961 – 2266 $\mu\text{mol kg}^{-1}$. TA

is form dissolution of calcium carbonate, biological and mixing processes. According to Honkanen *et al.*, (2021), TA is not affected by the sea–air CO₂ exchange, however calcifiers may have an effect on the carbon cycle in the benthic zone.

Fluxes of CO₂

CO₂ flux is the rate at which CO₂ dissolves into the waters or evaporates back into the atmosphere. At equilibrium, the net flux will be zero (Latifah *et al.*, 2021). The results of CO₂ flux calculations suggest that Semarang Bay waters form source of CO₂ releasing CO₂ back into the atmosphere after being absorbed by the oceans through a diffusion process and is used for photosynthesis. The CO₂ flux values in Semarang Bay (August 2021) range from 2–84 mmol CO₂ m⁻² day⁻¹ (Figure 7.). This flux is higher than in other waters such as the Java Sea. Measurements in October 2015 showed a maximum CO₂ flux value of 5 mmol CO₂ m² day⁻¹ (source CO₂) in the western part of the Java Sea, and in August 2015 a maximum weight of 17 mmol CO₂ m⁻² day⁻¹ (source CO₂) in its eastern part (Wirasatriya *et al.*, 2020). The Bintan Sea in April functioned as an CO₂ source (0.46±0.28 mmol m⁻² day⁻¹) and as a CO₂ sink in August (-0.27±0.10 mmol m⁻² day⁻¹) (Afdal *et al.*, 2020); The inner waters of Sekotong Bay had a range of CO₂ flux values of CO₂ 0.07–0.52 mmol m⁻² day⁻¹, and the seas acted as a source (Afdal, 2016). Another study (Macklin *et al.*, 2019) in Gilimanuk Bay showed that the average CO₂ flux in mangroves, seagrasses, and corals ecosystems was 9.8±6.0 mmol m⁻² day⁻¹ (source CO₂) in average. In mangrove ecosystems, the CO₂ flux is greater (substantial source CO₂) than in seagrass ecosystems (weak source CO₂). Figure 7 shows that the CO₂ flux distribution at stations 4 and 5 (22 August 2021

at 08.00 and 09.00 WIB) have the lowest values (1.89 and 12.19 mmol CO₂ m⁻² day⁻¹) because the sea surface temperature was measured 27.8°C. (Figure 8.) When it rained at night the pCO_{2sea} was low (377.58 dan 390.64 µatm). Whereas on the same day, the surface temperature of the waters began to rise (35.2 °C), and the wind speed was high (7.38 m s⁻¹) so that the pCO_{2sea} was high at 461.04 µatm.

The high value of CO₂ flux (1.89–46.73 mmol CO₂ m⁻² day⁻¹) in the waters of Semarang Bay may result from the high value of pCO_{2sea} (377.58–461.04 µatm) due to the high concentration of k_{wa} (21.89–38.69) caused by the wind speed (5.87 – 7.38 m s⁻¹). The wind speed variability on the calculated gas transfer rates and possible chemical enhancement of CO₂ exchange has significant effects at low wind speeds over the ocean (Wanninkhof, 1992). The high flux of CO₂ in Semarang Bay is also caused by the variations in sea surface temperature (Figure 8.). At stations 4 and 5 (22 August 2021 at 08.00 and 09.00 WIB), low surface temperatures (27.8°C) and high chlorophyll-a (3.88–4.00 mg L⁻¹) resulted in low pCO_{2sea} (377.58 and 390.64 µatm) and low CO₂ flux (1.88 and 12.19 mmol CO₂ m⁻² day⁻¹). The study by Wirasatriya *et al.* (2020) stated that wind speed played a vital role in determining the variability of CO₂ flux in the Java Sea. More substantial wind speeds (0–40 m s⁻¹) and lower temperatures (higher chlorophyll-a) during El Nino (in August 2015) increased k_{wa} so that pCO_{2sea} and CO₂ flux increased (Wirasatriya *et al.*, 2020). According to Kartadikaria *et al.* (2015), overall, pCO_{2sea} in the Indonesian seas is supersaturated relative to the atmosphere of 15.9 ± 8.6 µatm to act as a source of CO₂ / releases CO₂ into the atmosphere.

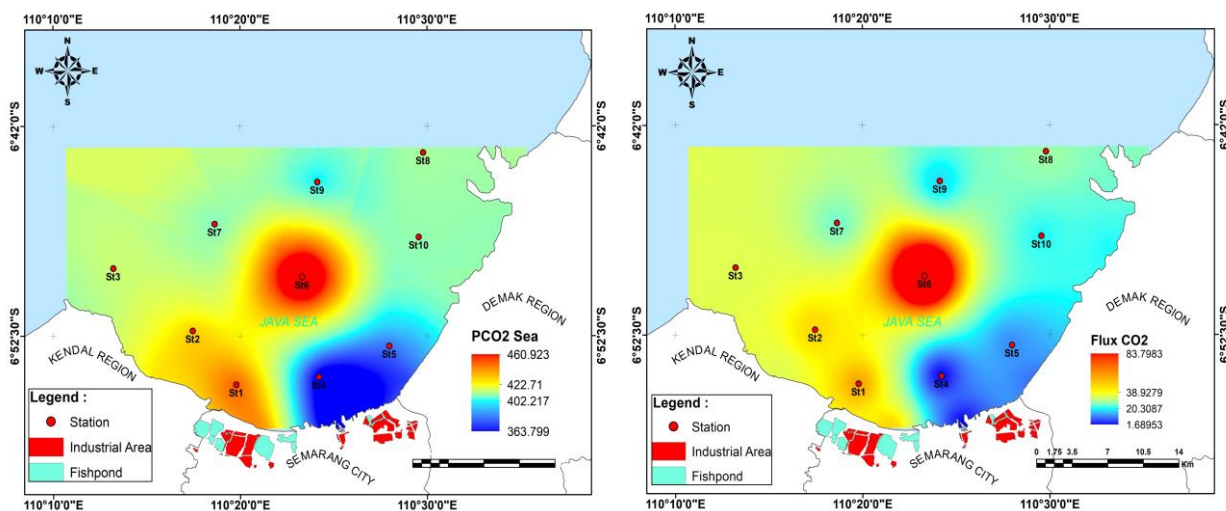


Figure 7. Distribution of pCO_{2sea} and CO₂ flux in August 2021 in Semarang Bay

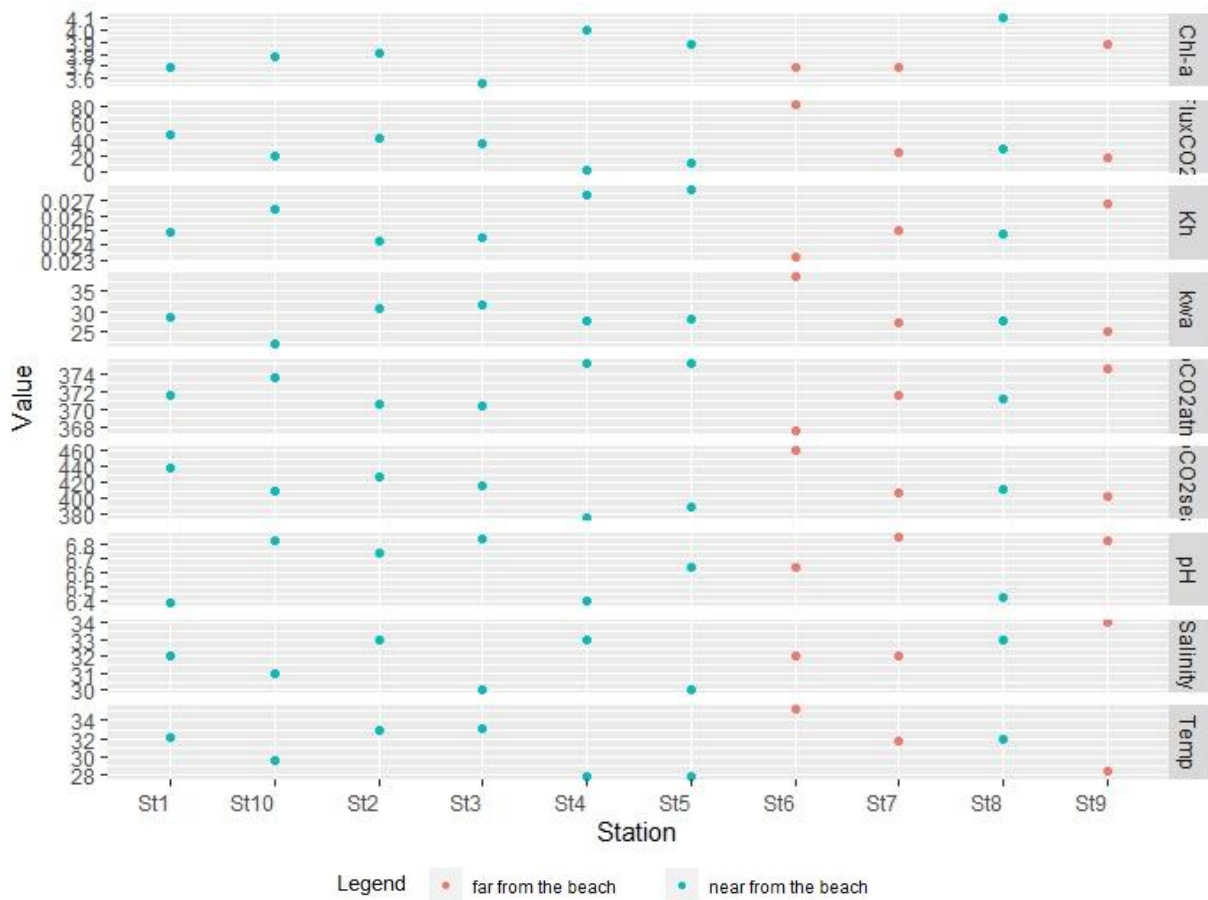


Figure 8. Fluxes of CO₂ and other variables

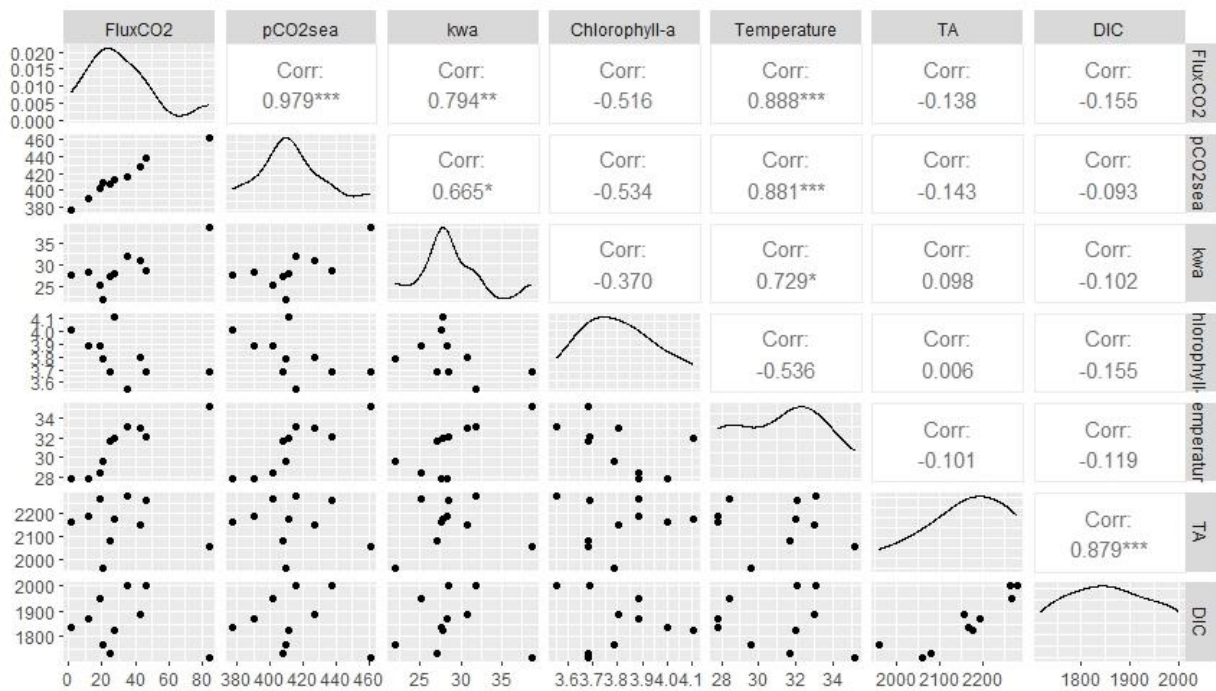


Figure 9. Spearman correlation of CO₂ flux with other variables

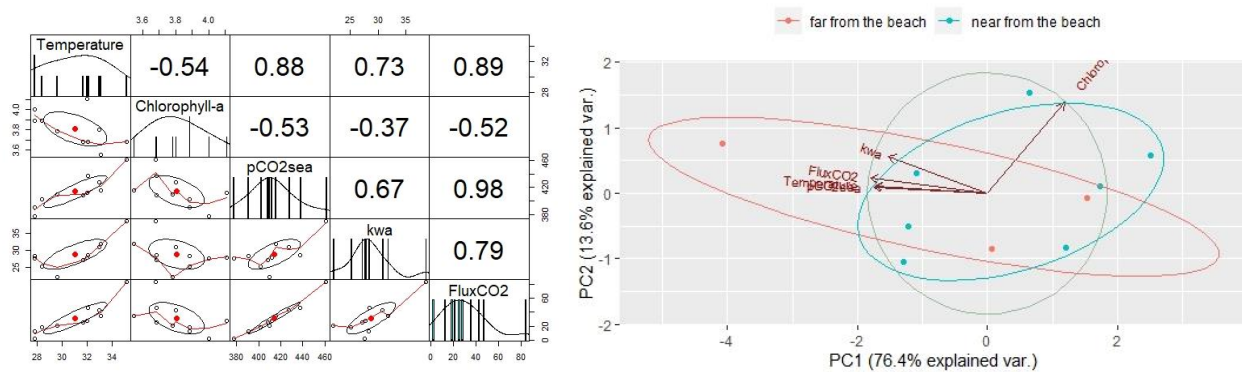


Figure 10. The results of PCA (Principal Component Analysis)

Water quality variables that affect CO₂ Flux

Previous studies (Takahashi et al., 1997; Takahashi et al., 2002; Cai et al., 2006; Takahashi et al., 2009; Otero et al., 2013; Yasunaka et al., 2016; Arnone et al., 2017; Ito et al., 2018; Tian et al., 2020) showed that in waters in middle and high latitudes with low temperatures, the oceans acted as CO₂ sinks. While studies conducted at low latitudes with high (warm) temperatures (Fitranti et al., 2013; Kartadikaria et al., 2015; Afdal, 2016; Yan et al., 2018; Latifah et al., 2020) showed that the oceans served as a source of CO₂. Warm surface water causes CO₂ to be returned to the atmosphere by the oceans (outgassing of CO₂). If the cold surface water causes CO₂ to dissolve easily, CO₂ transfer from the atmosphere to the surface occurs (uptake of CO₂).

Several variables, including pCO₂sea, pCO₂atm, k_{wa}, k_H, sea surface temperature, chlorophyll-a, salinity, wind, pH, DIC, TA influence the role of these waters as a CO₂ sink or CO₂ source. The determination of these variables is based on the CO₂ calculation formula, which requires the values of the variables pCO₂sea, pCO₂atm, seawater solubility (k_H) calculated as a function of temperature and salinity, gas transfer rate (k_{wa}) calculated as a function of the wind speed, and the Schmidt number which is affected by sea surface temperature (Weiss, 1974; Wanninkhof, 1992; Takahashi et al., 2002; Zhu et al., 2009; Robbins et al., 2010; Kartadikaria et al., 2015; Du et al., 2015; Takahashi and Sutherland, 2017; Yan et al., 2018; Wirasatriya et al., 2020). Based on the results of Spearman's correlation (Figure 9.) it shows that CO₂ flux has a strong positive significant relationship with pCO₂sea (r= 0.98), temperature (r= 0.88) and k_{wa} (r= 0.79), while chlorophyll-a (r= -0.52), has a moderate negative correlation, while TA (r= -0.14), and DIC (r= -0.15) have a weak negative relationship with CO₂ flux. Therefore, the following analysis uses PCA

where the predictor variables used are temperature, chlorophyll-a, pCO₂sea and k_{wa} to determine the effect on the CO₂ flux variable.

Based on the results of PCA analysis (Figure 10.), among these variables, the most influencing CO₂ flux is the ocean CO₂ partial pressure (pCO₂sea), temperature and k_{wa}. Temperature, pCO₂sea, and k_{wa} have the same direction as the CO₂ flux. According to Afdal et al. (2020), the temperature in coastal ecosystems (estuaries, mangroves, seagrasses, and coral reefs) is the main factor causing variations in CO₂ flux in Bintan waters in April as a source of CO₂ and in August as a sink of CO₂, both spatially and temporally. In Figure 10, it can also be seen that the CO₂ flux at stations far from the beach is more influenced by temperature, pCO₂sea and k_{wa}. While and at the station close to the beach, the chlorophyll-a factor is more dominant.

Conclusion

With a CO₂ flux value of 1.88–83.79 mmol CO₂ m⁻² day⁻¹, Semarang Bay functions as a CO₂ source. At stations 4 and 5 during morning sampling, with locations close to the coast and surrounded by many settlements and industries, the CO₂ flux value was the lowest (1.86 mmol CO₂ m⁻² day⁻¹). The value was also affected by low water temperatures (27.8°C) due to rain at night. On the same day at noon, the temperature increased again to the highest (35.2°C) measurement. Station 6 has high pCO₂sea (461.04 µatm) and CO₂ flux (83.79 mmol CO₂ m⁻² day⁻¹). The high concentration of chlorophyll-a (3.55–4.11 mg L⁻¹) is also visible from the colour (green) of the waters of Semarang Bay during sampling indicating that there is an abundance of green microalgae. This chlorophyll-a concentration has a negative correlation with CO₂ flux. The concentrations of TA and DIC show no relationship with CO₂ change. TA concentrations range from 1,961.20 to 2,277.51 µmol kg⁻¹, and DIC

concentrations range from 1,716.68 to 1,999.70 $\mu\text{mol kg}^{-1}$. Based on PCA analysis, the variability of CO_2 flux in Semarang Bay during August 2021 is influenced by the variability of $p\text{CO}_{2\text{sea}}$, sea surface temperature and k_{wa} (affected by wind speed).

Acknowledgements

The authors would like to thank the Dean of the Faculty of Fisheries and Marine Science (FPIK), Universitas Diponegoro, Indonesia, for providing research funds through Budget Year 2021, grant Number: 61/UN7.5.10.2/PP/2021. The authors also express great gratitude to anonymous reviewers for providing suggestions to improve and enable the publication of this paper.

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