CO₂ Sink/Source Characteristics in the Tropical Indonesian Seas

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Abstract.

Two distinct CO₂ sink/source characteristics are found in the tropical Indonesian seas from the compilation of observed data for the period 1984-2013. The western region persistently emits CO₂ to the atmosphere, whereas the eastern region is dynamic and acts either as a small source or sink of CO₂ to the atmosphere, depending on sites. The segregation is proximal to the Makassar Strait, which is located over the continental shelf and is one of the main routes of the Indonesian Throughflow (ITF). Lower salinity and higher silicate were found in the western region, suggesting a terrestrial influence in this area. Temperature has a limited influence in controlling different CO₂ sink/source characteristics in the west and east. However, an SST change of -2.0 °C during La Niña events effectively reduces the pCO₂ difference between the atmosphere and surface seawater by 50% compared to normal year conditions. During La Niña events, higher wind speeds double the CO₂ flux from the ocean to the atmosphere compared to that of a normal year. In the continental shelf area where the CO₂ sink area was found, data of over 29 years show that the seawater pCO₂ increased by 0.6-3.8 $\mu\text{atm yr}^{-1}$. Overall, the seawater pCO₂ of the Indonesian Seas is supersaturated relative to the atmosphere by $15.9 \pm 8.6 \mu\text{atm}$ and thus acts as a source of CO₂ to the atmosphere.

1. Introduction

Research on seawater CO₂ is in demand in Indonesia due to the lack of reliable and well-documented ocean CO₂ data. Global maps of sea surface partial pressure of CO₂ [Takahashi et al., 2009; Bakker et al., 2014] and inventories of anthropogenic CO₂ [Sabine et al., 2004] show Indonesian seas as a blank spot for CO₂ measurement. As a country with one of the longest coastlines, Indonesia possesses an extensive coastal ocean region, but so far little attention has been given to its carbon budget and what role this plays in global carbon cycle [e.g., Borges et al., 2005]. Many parts of the Indonesian coastal ocean receive large amounts of terrestrial organic matter and nutrient inputs [Sasai et al., 2011]. Other parts (e.g., the Makassar and Lombok Straits) are strongly influenced by throughflow of water masses from the Pacific to the Indian Ocean, known as the Indonesian Through Flow (ITF) [Gordon et al., 1999, 2008; Susanto et al., 2007; Kartadikaria et al., 2011b]. Considering this setting, complex air-water CO₂ interactions can be expected, and extensive measurements are required to determine the CO₂ sink/source characteristics of the Indonesian seas, including the role of the oceans surrounding Indonesia in terms of binding excessive atmospheric CO₂.

According to Laruelle et al., [2014], a new global CO₂ sink estimate of 0.19 ± 0.05 Pg C yr⁻¹ in continental shelf seas falls in the low end of previous estimates, i.e., 0.18-0.45 Pg C yr⁻¹, implying overestimation of the earlier studies of the continental shelf CO₂ sink process. Several studies have concluded that for certain continental shelves, the shallow zone (< 200 m) [Tsunogai et al., 1999; Frankignoulle and Borges, 2001; Thomas et al., 2004], slope-shelf zone [Walsh et al., 1985] and deeper zone, especially in the Atlantic

Ocean, all act as sinks, i.e., they take up atmospheric CO₂. In the open ocean, tropical seas are natural sources of CO₂ [Takahashi et al., 2002; Feely et al., 2006]; however, high latitude oceans have an opposing role [Sabine et al., 2004]. These reasons have motivated us to map the sea surface partial pressure of CO₂ (SSpCO₂) that spatially represent the primary Indonesian seas. Here, we present measured SSpCO₂ from tropical Indonesian seas based on the observed data and discuss a possible CO₂ sink/source mechanism, the influence of La Niña on air-sea CO₂ exchange, and annual trends of oceanic CO₂ in the region.

2. Materials and Methods

Locations and archived CO₂ data – The study area was located within 12°S-5°N, 104-130°E. Six datasets were obtained by direct measurements of SSpCO₂ over four years (2010-2013). These were mostly collected during the austral summer season (Table 1). Six additional datasets of the mole fraction of CO₂ in surface seawater (xCO₂) from the CDIAC-LDEO database V2009 (<http://cdiac3.ornl.gov/waves/underway/>) and AOML-Xuelong expedition (<http://www.aoml.noaa.gov/ocd/gcc/index.php>) were downloaded and analyzed in conjunction with the data collected as part of this study. The atmospheric xCO₂^{ATM} data were obtained from the NOAA/ESRL database (<http://ds.data.jma.go.jp/gmd/wdcgg/jp/wdcgg-j.html>). We selected the nearest location of atmospheric xCO₂^{ATM} data that were available during the listed cruises in Table 1. The atmospheric xCO₂^{ATM} data were used to calculate the pCO₂ difference between the surface ocean and atmosphere ($\delta p\text{CO}_2$). The method of calculating pCO₂ from xCO₂ is explained in the next subsection. In total, CO₂ data from 12 cruises were

used for this study. This is one of the most comprehensive CO₂ data compilations for the Indonesian archipelago (Table 1).

In situ measurements - One- to ten-minute recordings of sea surface temperatures (SST), sea surface salinity (SSS), chlorophyll-a (Chl-a), turbidity, and dissolved oxygen (DO) were obtained using Compact sensors (JFE Advantech Co. Ltd., Japan). An onboard weather station was utilized to measure air temperature, humidity, solar radiation, wind direction and wind speed, and air pressure (Onset Computer Corporation, US). Starting from the EWIN cruise in 2010, continuous measurements were collected by pumping sea surface water into a plastic container. A fast, continuous, and portable CO₂ analyzer capable of measuring the in situ xCO₂ in the water and atmosphere was utilized in our observations (C02-09, Kimoto Electric Co. Ltd., Japan). For one hour of continuous measurements of xCO₂ (equal to one cycle), we measured the xCO₂ every minute in the water for 50 minutes and then continued to measure the xCO₂ in the air for 5 minutes and xCO₂ in the CO₂-free gas (air fed through a *soda-lime* column) for 5 minutes. The CO₂ analyzer was pre-calibrated with a known concentration of CO₂ in dry air (250, 500, 797, and 1100 ppm) and was intended to measure the mole fraction of xCO₂ in gas form [Saito et al., 1995]. In water, the xCO₂ level was measured as a fraction of the CO₂ in dry air because the membrane filter (*Gore-Tex*) in the pCO₂ analyzer was able to equilibrate the CO₂ in seawater and air after removing the water vapor. With this machine, a direct pCO₂ value was not obtained; this value must be calculated from the measured xCO₂ value with the following equation:

$$pCO_2 = xCO_2 (P_{eq} - P_w) . \quad (1)$$

where P_{eq} is the total pressure of the carrier gas measured in the equilibrator chamber, and P_w is the saturation vapor pressure of water at the equilibrium temperature, T_{eq} . P_w can be calculated from the equation following the methods of Weiss and Price [1980] [Körtzinger 1999]. The accuracy of the pCO₂ analyzer was estimated at 7 μ atm [Saito et al., 1995].

Discrete oceanographic samples were also collected using a CTD and *rosette sampler*. The seawater that was collected from certain depths by the *rosette sampler* was then used for the following analyses: (1) CO₂ system parameters (total alkalinity (TA), dissolved inorganic carbon (DIC), pH, and salinity); (2) nutrients; (3) suspended solids (SS); and (4) Chl-a. For the treatment of TA, DIC, and pH, we placed the sampled water into a 250 mL *Schott Duran* bottle according to standard procedures outlined in Dickson et al., [2007], and preserved the sample with 200 μ L of a saturated HgCl₂ solution to halt any biological activity. In the treatment for nutrients, a 10-mL sample was collected in two acrylic tubes and frozen in a scientific freezer (temperature < -2 °C) for further laboratory analysis. One liter of water was filtered with a 47-mm diameter GF/F filter to collect the SS data, and 200 mL of water was filtered with a 25-mm diameter filter to collect the Chl-a data. The 47-mm GF/F filters for SS were dried and weighed in advance, stored in a freezer after the filtration, and then dried and weighed again in the laboratory. The 25-mm GF/F filters for Chl-a were immediately placed in a tube that contained 6 mL of *N-N dimethylformamide* and preserved in a freezer until analysis.

Laboratory analyses – For each 250-mL *Schott Duran* bottle sample, we conducted CO₂ and salinity analyses. For the CO₂ analysis in the laboratory, we used a carbonate system analyzer (Kimoto Electric Co., Ltd., Japan) containing a flow-through-type analyzer that

could accurately analyze TA, DIC and pH [Kimoto et al., 2001, 2002; Watanabe et al., 2004]. Each sample was continuously fed into the analyzer, which trifurcated the flow to the TA, DIC and pH lines, and each reading was determined by the appropriate detectors.

Table 2 summarizes the list of parameters measured using the instruments and their accuracies relative to Certified Reference Materials (CRMs) for both in situ and ex situ measurements. We analyzed the nutrient samples to obtain the NO₃, NO₂, NH₄, PO₄, and SiO₂ values using the QuAatro-SFA analyzer (SEAL analytical Ltd.), and we measured the discrete Chl-a concentration using a 10-AU fluorometer from Turner designs. For TA and DIC analyses, CRMs from the Scripps Institution of Oceanography were used. For all nutrients except for NH₄, reference materials from KANSO Japan were used (<http://www.kanso.co.jp/eng/index.html>). We did not have CRMs for Chl-a; therefore, we calibrated our analysis against a standard that was prepared from pure chlorophyll powder. Calculated pCO₂ values from combinations of DIC, TA, and pH inevitably have errors due to error propagation from DIC, TA, and pH themselves. DIC and TA were measured with 2-3 $\mu\text{mol kg}^{-1}$ accuracy and precision, and pH was measured in total scale with 0.002 precision and 0.005 accuracy. The calculated pCO₂ has an accuracy and precision of $\sim 10 \mu\text{atm}$ from TA-DIC and $5 \mu\text{atm}$ from TA-pH or DIC-pH (evaluated based on Chapter 8 in Grasshoff et al., 1999). The direct measurement has an accuracy of $\sim 7 \mu\text{atm}$ and precision with a similar range or better. The purpose of comparing calculated pCO₂ from TA-DIC and direct measured pCO₂ was to check the internal consistency (i.e., to evaluate whether all of the parameters were measured accurately). Furthermore, the merit of measuring TA-DIC is that these measurements can

describe more information about biogeochemical processes occurring in the system (such as photosynthesis, respiration, calcification, etc.).

Data validation and assessment – The final data were compiled at oneminute intervals based on the temporal resolution of the SS_pCO₂ data. Five minutes of weather data and ten minutes of DO data were interpolated to one-minute data. One-minute SST, SSS, Chl-*a*, and SS data can be directly used after being re-calibrated according to accurate discrete data (i.e., CTD). The assessment of underway data deletion was performed when the SST and SSS values collected by the CTD/*rosette* instrument (i.e., direct sea surface water measurement) had significant errors compared with the continuous underway SST and SSS data collected by the Compact-CT/Infinity-CT and Compact-CLW/Infinity-CLW instruments (i.e., pumping water measurement). The error usually originated from the mixture of old and new water inside the container that needed to be cleaned depending on water clarity. In addition, when the water discharge fell far below the ideal water flow rate (1 L sec⁻¹), data were then discarded.

Sink/Source (net flux) calculation - We estimated the sink/source of CO₂ from the result of Equation 1 by first subtracting the calculated pCO₂ value for seawater (pCO₂^{SW}) from the mean calculated pCO₂ value of the atmosphere (pCO₂^{ATM}) of each cruise using the following equation:

$$\delta pCO_2 = pCO_2^{SW} - \overline{pCO_2^{ATM}} \quad (2)$$

Because the pCO₂ analyzer did not measure the xCO₂ in the atmosphere concurrently with the measurements of the xCO₂ in seawater, we could not instantaneously obtain the δpCO₂ value. Therefore, we calculated the mean value of pCO₂^{ATM} from the average of

one cruise and subtracted the averaged $p\text{CO}_2^{ATM}$ data from each of the $p\text{CO}_2^{SW}$ values. This assumption was valid because the atmospheric CO₂ values did not show significant temporal variability and was spatially homogeneous and less variable, especially during the limited duration of the cruise. For example, the standard deviation of SSpCO₂ was recorded smallest at 1.2 μatm during the EWIN2010 cruise and highest at 5.0 μatm during the EWIN2013 cruise. The $\delta p\text{CO}_2$ information was further analyzed to estimate the CO₂ flux; the net air-sea CO₂ flux (F_{CO_2}) ($\text{mol m}^{-2} \text{yr}^{-1}$) can be estimated using the following equation:

$$F_{CO_2} = k_{CO_2} \times S_0 \times \delta pCO_2 \quad (3)$$

where the above equation is a function of the transfer velocity (k_{CO_2}), the solubility of CO₂ (S_0) and the sea-air pCO₂ difference ($\delta p\text{CO}_2$). The transfer velocity of CO₂ (cm hour^{-1}), which is commonly parameterized as a function of the mean wind speed (U_{10}) (m s^{-1}) and the solubility of CO₂ in seawater ($\text{mol kg}^{-1} \text{atm}^{-1}$), followed the general solubility of a non-ideal gas according to Weiss [1974]:

$$\ln S_0 = A_1 + A_2 (100/T) + A_3 \ln (T/100) + S [B_1 + B_2 (T/100) + B_3 (T/100)^2] \quad (4)$$

where the A and B constants can be obtained directly in the manuscript of Weiss [1974], T is the temperature (Kelvin), and S is the salinity (psu).

The quantification of the CO₂ flux between the ocean and atmosphere was explained by McGillis et al., [2004]. There was no concise information on the selection of kinematic transfer for the region. Here, without giving constraint to the final CO₂ flux result, we

adopted well-known variety types of kinematic transfer velocities for CO₂ gas that ranged from low to high k_{CO_2} estimations depending on the U_{10} (wind speed at 10 m altitude).

$$k_{CO_2} = \begin{cases} 0.39U_{10}^2(Sc/660)^{-0.5} & \text{(a)} \\ 0.22U_{10}^2 + 0.33U_{10} & \text{(b)} \\ (3.3 + 0.026U_{10}^3)(Sc/660)^{-0.5} & \text{(c)} \end{cases} \quad (5)$$

Equations 5a, 5b and 5c refer to Wanninkhof [1992], Nightingale et al., [2000] and McGillis et al., [2001], respectively. Sc is the Schmidt number, which describes the ratio between the kinematic viscosity and diffusivity process of CO₂ and is a function of temperature [Jähne et al., 1987; Wanninkhof, 1992]. Detailed results of the CO₂ sink/source for each cruise are explained in the following section.

3. Cruise Results

3.1. Spatial δpCO_2 and pCO_2

The complete compiled distribution of δpCO_2 in the Indonesian seas for the period 1984–2013 is depicted in Figure 1a. Here, we interpolated the underway data to 15-minute pixel grids with a radius of approximately 150 km because the largest interval between stations was 20–30 minutes during the EWIN cruise in 2010. In this study, the data spanned from 1984 to 2013. The CO₂ data not only showed high seasonal variability, but also gradually increased inter-annually for the duration of the measurements. For measurements with a wide temporal range, Takahashi et al. [2002, 2009] normalized pCO_2 data to a single reference year of 2000 by identifying the rates of pCO_2 change and the seasonal correction factor in various areas of the global ocean. Following this concept, a normalization technique has been applied to Figure 1b of this study. Our method to generalize the distribution of δpCO_2 is described in the Appendix. From

these figures, the obvious pattern from the above results indicates that the inner western region of the Indonesian seas (Java Sea) had higher $\delta p\text{CO}_2$ compared to the eastern region (Flores and Banda Seas). The Java Sea is part of the shallow Sunda continent, but the bathymetry increases toward the continental shelf. In the Makassar Strait, where the continental shelf is located, we observed CO₂ sink areas. A considerable CO₂ sink area was also found in the Banda Sea. However, the mean observed $\delta p\text{CO}_2$ from each mission indicated that the overall region act as a CO₂ source to the atmosphere; the mean value along the track during the February 2010, April 2010, April 2011, June 2011, November 2012 and May 2013 cruises were +15.6, +11.7, +20.6, +11.5, +34.3, and +17.2 μatm , respectively. Consequently, the SSpCO₂ level was generally higher with respect to the observed mean atmospheric CO₂ level. The difference of SSpCO₂ to the atmosphere is approximately 11 - 34 μatm and taking into account the average of SSpCO₂ from the other open-source datasets generate $+15.9 \pm 8.6 \mu\text{atm}$ uptake of atmospheric CO₂. On the other hand by taking a broader view, the regions were situated between appreciable CO₂ sink regions located in the Pacific (ITF entrance) and Indian Oceans (ITF outlet) (Figure 1). Therefore, the CO₂ characteristics were observed to change as the water flowed through the Indonesian seas.

Insights into the characteristic differences between the western and eastern regions of the Indonesian seas can be obtained by analyzing the data collected during the April 2010, June 2011 and June 2013 cruises. The cruise tracks covered the west and central/east regions of the Indonesian seas. In April 2010, the average of SSpCO₂ level was higher compared to the observed mean atmospheric CO₂ level of $379.2 \pm 1.2 \mu\text{atm}$ (Figure 2), but with considerable variability. The observed mean SSpCO₂ level was 387.4 ± 49.7

μatm . In this case, under similar wind conditions, the Java Sea can potentially release a larger amount of CO₂ to the atmosphere than the amount contributed by the other seas. In contrast, SSpCO₂ significantly decreased in the continental slope region around the Surabaya and Lombok Strait ($\delta\text{pCO}_2 \approx -40 \mu\text{atm}$), and the SSpCO₂ level was lower than the atmospheric CO₂ in this area. In the Flores and Banda Seas, the SSpCO₂ level steadily increased; it was slightly higher than the atmospheric CO₂ level but less than $420 \mu\text{atm}$. A maximum $\delta\text{pCO}_2 \approx 100 \mu\text{atm}$ was observed in the Java Sea, whereas in the Flores and Banda Seas, the difference in pCO₂ between the ocean and atmosphere was $40 \mu\text{atm}$.

From the end of June to the beginning of July 2011, the average SSpCO₂ level was $388.3 \pm 16.4 \mu\text{atm}$, whereas the observed mean atmospheric CO₂ level was $376.3 \pm 2.9 \mu\text{atm}$ (Figure 3a). A majority of the high positive SSpCO₂ was located in the Java Sea, and the shallow areas of the Flores Sea (close to Makassar) exhibited higher values of δpCO_2 ($\approx 30 \mu\text{atm}$) compared to the Banda Sea (Figure 3b). In this time period, the temperature in the Java Sea was higher and the salinity was lower; however, this condition gradually changed toward the Banda Sea (Figure 3c). Temperatures slightly decreased and salinity increased by 1-2 units. The DO values were relatively lower ($\approx 5 \text{ mg L}^{-1}$) than the normal value ($6\text{--}7 \text{ mg L}^{-1}$) (Figure 3d). This low DO trend was independently verified using iodometric titration (*Winkler method*) and the DO sensors in the CTD and Compact instruments. All of these measurements and calculations suggest that the DO during this cruise was relatively low compared with the normal surface DO levels. Nutrient measurements in the Java Sea indicated that the surface silica concentration was

higher in the Java Sea than in the Banda Sea (Figure 3e). One surface silica measurement on June 26th was recorded at $29.9 \mu\text{mol L}^{-1}$ (not shown in the graph).

In June 2013, the cruise mission was aimed at the Makassar Strait area next to the Java Sea. The SSpCO₂ level was generally higher with respect to the observed mean atmospheric CO₂ level ($383.3 \mu\text{atm} \pm 5.0 \mu\text{atm}$) (Figure 4). The average δpCO_2 was $+17.2 \mu\text{atm}$, which showed that the Java Sea contributed higher CO₂ emissions compared to the other seas. Temperature was lower in the center of the Makassar Strait and higher elsewhere.

3.2. Increasing pCO₂ in the Makassar Strait

The Makassar Strait area is a potentially important region for CO₂ studies in that we found a mixture of CO₂ sinks and sources advected from the Java Sea and Pacific Ocean. Using our data, we examined the possible changes in pCO₂ in this area (Figure 5). There were four cruise tracks that crossed the Makassar Strait (cruises in 1984, 2007, 2008 and 2013); however, two additional tracks (cruises in 2010 and 2011) from south of the Makassar Strait were also used to investigate the pCO₂ trend over this time period. The selected underway cruise data (denoted with a colored dashed line) were then extrapolated to a radius of up to 150 km. We performed a transect slice (denoted by the black dashed line) through the Strait and compared the pCO₂ levels from 1984 to 2013, as calculated from Equation 1. No de-seasonalization or normalization methods were applied in Figure 5.

Based on the cruise tracks that crossed the Makassar Strait, the pCO₂ level was clearly observed as increasing from $354.5 \mu\text{atm}$ in 1984 to $390.7 \mu\text{atm}$ in 2013. At a latitude of 5°S, where all data existed, the pCO₂ level in 2010 was smaller than that in 2008.

However, in general, we found an increasing trend of pCO₂ in the Makassar Strait. The linear regression for the years 1984 to 2007 was $y = 1.01x + 9.53$ ($R^2=0.75$); however, if we include data for the year 2008, the regression was $y = 0.66x - 956.84$ ($R^2 = 0.89$). Then, for the period 1984-2007/2008, the pCO₂ increased at a rate of 0.6-1.0 $\mu\text{atm yr}^{-1}$. The trend was more apparent in the period 2007-2013 in which the linear regression was $y = 3.82x - 7288.3$ ($R^2 = 0.89$); the pCO₂ level increased 3- to 6-fold. Table 3 explains in more detail the underway-averaged pCO₂ levels from the diagonal dashed black line in Figure 5. A 35.5 μatm increase was observed within the last 29 years; however, the trend has been dramatic in recent years. With a degree of confidence of $R^2 > 0.75$, the increasing trend in CO₂ was calculated as approximately $\pm 1.0 \mu\text{atm yr}^{-1}$.

3.3. CO₂ source areas

The SSpCO₂ distribution in the Java Sea indicates that this area is a major contributor of CO₂ to the atmosphere (Figure 6). From the four cruises conducted in April 2010, April 2011, June 2011, and November 2012, evidence of persistent positive δpCO_2 was found. By comparing the EWIN2010 (April 2010) and EWIN2011 (April 2011) data only for the Java Sea, it is evident that the area averaged value of δpCO_2 decreased from 58.9 μatm in 2010 to 20.6 μatm in 2011. In the June 2011 data, recorded after the La Niña event, the δpCO_2 increased again to 25.1 μatm . During the onset of the northwest monsoon in November 2012, the δpCO_2 was 34.3 μatm . All of the δpCO_2 values in the Java Sea fall in the high-end level of the mean δpCO_2 range in the Indonesian seas.

The CO₂ fluxes for all cruises were calculated according to Equation 3 and are shown as the black line in Figure 6. From our observations, the average CO₂ fluxes indicated the average CO₂ emissions from the sea to the air during the total cruise periods of the

February 2010, April 2010, April 2011, November 2012 and May 2013 cruises were +1.3, +2.2, +5.3, +5.8 and +2.9 mol m⁻² yr⁻¹, respectively. As we did not measure the wind data in June 2011, we could not determine the average CO₂ fluxes during this period. Our results suggest that during our cruises, the Java Sea acted as a main source of CO₂ to the atmosphere.

4. Discussion

The physical and biogeochemical properties of the Indonesian seas are characterized by large temporal and spatial variability. Specifically, our study showed that the spatial distribution of SSpCO₂ was different between the western and eastern regions, with proximal segregation occurring at the Makassar Strait. This transition zone coincides with the "*Wallace line*", a virtual line segregating the terrestrial and marine flora and fauna, which is also located in the continental shelf of the Makassar Strait [Barber et al., 2000]. In general, the Indonesian archipelago acts as a source of CO₂ to the atmosphere, which is consistent with the findings of Gruber N. [2015] and Laruelle et al. [2014], which indicated that low latitude coastal regions release CO₂ to the atmosphere.

4.1. Physical and biogeochemical aspects related to the SSpCO₂ distribution

Both temperature and salinity affect pCO₂. A one-degree warming increased the pCO₂ by approximately 13 μatm, whereas a 1 psu increase in salinity resulted in a pCO₂ increase of 9 μatm [Takahashi et al., 1993; Keeling et al., 2004]. Our data showed that the surface layer salinity and temperature varied greatly between the west and the east. In the western part, the surface salinity was lower, while temperature was higher compared to that in the eastern Indonesia seas. The SSS varied around two units between west and

east, whereas the SST varied slightly about one degree Celcius between west and east. The spatial change in salinity and temperature was followed by an observed change in SS_pCO₂, with more prominent change was reflected by the change in salinity.

The SS_pCO₂ source pattern (red color in Figures 2-4) not only showed a tendency to correlate negatively with salinity as explained above, but also depicted a positive correlation with silica. For instance, when the positive $\delta p\text{CO}_2$ was high, the salinity was low and silica concentration was high, and when the positive $\delta p\text{CO}_2$ was low, the salinity was relatively high and silica concentration was low. This is an indication of freshwater and nutrient input from the land. Silica concentrations were high in shallow waters, such as in the Java Sea, which could have been caused by freshwater input from adjacent lands supplying silica. The similar condition of high silica was also found during the EWIN 2013 cruise. Normalized TA (NTA) and SiO₂ were positively correlated during this cruise (not shown). This is evidence that relatively high NTA water was brought by rivers together with SiO₂. The EWIN 2013 cruise was targeted to take samples along rivers in Kalimantan Island, especially at the second largest delta in Indonesia, which is called Mahakam Delta. Therefore, larger surface NTA data were found during the EWIN 2013 cruise than was mostly observed in the Makassar Strait (Figure 7). Observing water quality of mining areas was one of the aims during the Banggai 2011 cruise, and relatively higher NTA was also found during this cruise. This evidence indicates a potentially strong terrestrial influence in the Indonesian seas. Yet, in straits connecting to the Pacific or Indian Oceans, the change in pCO₂ could be caused by ocean advection or changes in salinity and temperature of the outside waters.

Other physical and biogeochemical factors, such as temperature and DO, also showed differences between the western and eastern regions; however, they were not as significant as the differences in salinity and silica. Based on the Chl-a data, all of the measurements were collected during non-phytoplankton bloom conditions; therefore, we were not able to examine and determine the quantitative correlation of Chl-a (and DO) on the SSpCO₂ distribution during bloom conditions.

From the preceding discussion, even though differences in the salinity characteristics influenced the spatial SSpCO₂ differences, we found that salinity did not have a significant correlation with the increasing SSpCO₂ trend in the Makassar Strait for the period 1984-2013 (not shown). This was also true for the SST, as the scatter in the data for the Makassar Strait showed that the time series of SST did not present the clear increasing trend, such as that of pCO₂. However, this explanation should be further analyzed and clarified by obtaining fixed long-term time-series data for Indonesian seas.

4.2. Possible influence of terrestrial input to pCO₂

In addition to section 4.1, we carefully analyzed the influence of salinity, especially the role of nutrients input from land to the pCO₂ distribution in the Indonesian seas. Using CO2SYS software, the observed SST and SSS in April 2010 were used to reconstruct the SSpCO₂ distribution of the western and eastern parts of the Indonesian seas. We selected the dissociation constant from Mehrbach et al., [1973], which was refitted by Dickson and Millero [1987], the KSO₄ constant from Dickson [1990], and pH based on the total scale, as the main constant parameters. We calculated pCO₂ from in situ SST and SSS data obtained during the cruise and from normalized TA (NTA) and DIC (NDIC). NDIC and NTA were calculated from the following equation:

$$NTA(NDIC) = NTA_o(NDIC_o) \times 35.0/SSS \quad (6)$$

where NTA_o and $NDIC_o$ are the offshore end-member values, as determined below. Samples taken from the far-ocean sites in the eastern Indonesian seas (Flores or Banda Seas) were averaged and normalized at $S=35$, then used as NTA and $NDIC$ (2290 and 1964 $\mu\text{mol kg}^{-1}$, respectively). In the eastern regions, water was less influenced from terrestrial inputs, which can clearly be observed at higher salinities, and water can be regarded as an offshore end-member. If TA and DIC were changed solely from freshwater input/output through rainfall/evaporation, the observed $p\text{CO}_2$ should match the $p\text{CO}_2$ calculated from this method. If not, other factors influencing TA and DIC (i.e., river discharge with significant TA and DIC , calcification and photosynthesis, upwelling of deep-sea water, gas-exchange, etc.) should be considered. The calculated $p\text{CO}_2$ showed that the temperature and salinity did not fully explain the $p\text{CO}_2$ variability that was observed in the Java Sea; however, the calculation sufficiently depicted the variability at the Flores/Banda Seas (Figure 8). In the Java Sea, a deviation of more than 44.0 μatm occurred between the calculated and observed $p\text{CO}_2$, whereas in the Flores/Banda Seas, this deviation was only 1.1 μatm . The high $p\text{CO}_2$ in the Java Sea can be explained from higher terrestrial organic matter inputs in this region. Thus, the large discrepancies were attributable to greater terrestrial impacts due to shallower water depth and to more densely populated cities in the western region. This theoretical approach suggested that a small change in salinity could influence the distribution of $p\text{CO}_2$ levels. However, the discrepancy that was found in the theoretical calculation using CO2SYS software showed that the $p\text{CO}_2$ condition at the surface was complex and influenced by many factors,

including the carbonate system, water-mass characteristics, nutrients, organic matter and atmospheric conditions.

4.3. Influence of temperature during La Niña on CO₂ emissions

Although the role of temperature was not as significant as salinity and silica, in this subsection, we show the influence of the interannual variability of temperature on the CO₂ conditions in the Indonesian seas, especially in the Java Sea. We describe an interesting feature of the $\delta p\text{CO}_2$ and CO₂ flux that was observed in the Java Sea. April 2011 was the termination period for the La Niña event of 2010. We found that the $\delta p\text{CO}_2$ decreased by approximately 50% in April 2011 compared to its value in 2010. This decrease was associated with the lowest averaged SST in the region.

The SST in April 2011 was lower than that in April 2010 by 1-2 °C, which was similar to the condition that occurred in February 2010 during the peak austral monsoon. Additional information collected from the NCEP/NCAR data for the SST and wind speed during the cruise missions is listed in Figure 9. The wind was stronger in April 2011 than in 2010; during this cruise, the La Niña event occurred, and its effects were prolonged in the Pacific Ocean, where strong winds with heavy rains and a typhoon were recorded before the cruise. These occurrences suggest that the cruise was not conducted during a normal year. The weather conditions in April 2011 resulted in lower SST values, which contributed to lower SSpCO₂ values (because of the thermodynamics) compared to those from the EWIN2010 (April 2010) data obtained from the Java Sea during normal conditions. It is clear that $\delta p\text{CO}_2$ decreases during La Niña events. Thus, in the absence of a La Niña event, the April 2011 levels of $\delta p\text{CO}_2$ would have been higher than those during the austral winter

conditions (June-August) because, in general, the temperature during the austral summer is higher than during the austral winter.

Although the $\delta p\text{CO}_2$ was lower during the April 2011 cruise ($20.6 \mu\text{atm}$) as a result of lower SST distributions, the CO₂ flux during the EWIN 2011 cruise ($5.29 \text{ mol m}^{-2} \text{ yr}^{-1}$) was 1.7 times larger than that of the EWIN 2010 cruise ($3.1 \text{ mol m}^{-2} \text{ yr}^{-1}$). This phenomenon occurred because the wind speeds during the April 2011 cruise ($4.4 \pm 2.3 \text{ m s}^{-1}$) were higher than those during the April 2010 cruise ($3.4 \pm 1.3 \text{ m s}^{-1}$) (Figure 9); however, the CO₂ flux was proportional to a second- and higher-order wind speed, suggesting that during the La Niña conditions, the CO₂ flux to the atmosphere increased.

5. Summary

This study demonstrates the first region wide compilations of the regional surface oceanic pCO₂ levels in the tropical Indonesian seas. The subsequent discussions focused on the different distribution of SSpCO₂ between the shallow Java Sea and deep Flores/Banda Seas. The results of these analyses revealed evidence that the Indonesian seas act as a source of CO₂, contributing approximately $3.7 \pm 2.2 \text{ mol m}^{-2} \text{ yr}^{-1}$, especially during the austral summer season. The results also showed that the shallow Java Sea released more CO₂ to the atmosphere than the deep Flores/Banda Seas, and the high SSpCO₂ in this region was associated with higher freshwater and nutrient input from land. Along the main ITF routes, we found that in the Makassar Strait, the SSpCO₂ trend over the past 29 years showed an average unit increase per year but a more appreciable increase in recent years. Despite the strong influence of salinity on the changes in pCO₂, the impact of temperature is more significant on a temporal (inter-annual) scale rather than a spatial scale, e.g., the changes in CO₂ emissions in the shallow Java Sea during La Niña

years. An SST change of -2.0 °C could effectively impact $\delta p\text{CO}_2$ and the CO₂ flux. The analysis presented in this study supports the notion that no single factor controlled the CO₂ dynamics in this region and that each parameter had its own specific influence either spatially or temporally, though salinity, silica, and TA appeared to control the SSpCO₂ through freshwater supply from adjacent lands. The authors believe that the ITF variability may contribute to the SSpCO₂ distribution; therefore, this topic should be considered a valuable subject for future research by the scientific community.

Appendix

To eliminate biases due to sampling year and seasonal changes, we normalized the data to last years data in 2013 by assuming that pCO₂ increases are associated with SST increases. This assumption is driven by the discovery that SST best fits the seasonal pattern of oceanic SSpCO₂. The SST model output of 1993-2007 [Kartadikaria et al., 2011a] was used to obtain the weighted average factor for the austral summer and winter seasons. We considered the seasonal factor of 0.96 and 1.04 for the austral summer and austral winter, respectively. The regression function that was used to normalize pCO₂ data to a single reference year was derived from the Hawaii Ocean Time-series (HOT) surface CO₂ time series data [Dore et al., 2009] with a rate of pCO₂^{HOT} change of 0.43 μatm per season. In Figure 1b, the normalized SSpCO₂ data were de-seasonalized using the seasonal factor (w) and de-trended using the rate of pCO₂ change (m), as mentioned previously, as follows:

$$SSpCO_2|_{t_{ref}} = SSpCO_2(t) \times w(t) + [m \cdot (pCO_2^{HOT}(t) - pCO_2^{HOT}|_{t_{ref}})] \quad (7)$$

When the data were reanalyzed (normalized), the atmospheric pCO₂ in 2013, at a level of 383.3 μ atm, was selected to obtain the δ pCO₂ value.

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Table 1. Available underway SSpCO₂ data (1984-present) during the austral summer season in the Indonesian seas.

No.	Cruise	Time (ddmmmyy)		Location		Interannual condition	SSxCO ₂ data	Atmospheric xCO ₂ data (name of station)
		Start	Finish	Latitude	Longitude			
1.	Makassar Strait	09Feb84	11Feb84	9.0°S-5.0°N	115.5°-119.6°E	Normal	CDIAC-LDEO	NOAA/ESRL (Mauna Loa, Hawaii)
2.	Savu Sea	11Mar92	20Mar92	8.5°-12.0°S	120.0°-124.0°E	El Niño	CDIAC-LDEO	NOAA/ESRL (Cape Ferguson, Australia)
3.	Sunda Strait	01Nov95	03Nov95	7.0°S-1.0°N	104.0°-107.0°E	La Niña	CDIAC-LDEO	NOAA/ESRL (South China Sea)
4.	Celebes Sea	25Jan96	25Jan96	0.0°-3.0°N	115.0°-123.0°E	La Niña	CDIAC-LDEO	NOAA/ESRL (South China Sea)
5.	24th Chinare Expedition	20Nov07	23Nov07	12.0°S-5.0°N	115.0°-126.0°E	La Niña	AOML-XUELONG	AOML-XUELONG ¹
6.	24th Chinare Expedition	04Apr08	10Apr08	12.0°S-5.0°N	115.0°-126.0°E	Normal	AOML-XUELONG	AOML-XUELONG ¹
7.	Spy-Mks Exp.	14Feb10	15Feb10	4.5°-7.0°S	112.0°-120.0°E	Normal	<i>This study</i>	NOAA/ESRL (Bukit Kota Tabang)
8.	EWIN2010	22Apr10	01May10	5.0°-10.0°S	106.0°-127.0°E	Normal	<i>This study</i>	<i>This study</i> , 379.2 ± 1.2 μ atm
9.	EWIN2011	12Apr11	23Apr11	3.5°S-4.0°N	106.0°-108.0°E	La Niña	<i>This study</i>	<i>This study</i> , 385.5 ± 1.5 μ atm
10.	Banggai Expedition	24Jun11	06Jul11	5.0°S-1.0°S	106.0°-123.0°E	Normal	<i>This study</i>	<i>This study</i> , 376.3 ± 2.9 μ atm
11.	SITE2012	22Nov12	30Nov11	6.0°S-1.0°S	106.0°-108.0°E	Normal	<i>This study</i>	<i>This study</i> , 380.9 ± 4.4 μ atm
12.	EWIN2013	05Jun13	20Jun13	6.0°S-0.0°S	111.0°-120.0°E	Normal	<i>This study</i>	<i>This study</i> , 383.3 ± 5.0 μ atm

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Table 2. Underway and discrete measured parameters.

No.	Parameter	Main instruments		Validation			
		Name	Output used	Method	Name	Output used	Remark
1.	Carbonate system	portable pCO ₂ analyzer	SSpCO ₂ , atm. pCO ₂	<i>In situ</i>	pCO ₂ analyzer	DIC, TA, pH	For the details see
							Saito et al., 1995
2.	Temperature	Compact CT	SST	<i>In situ</i>	CTD	Temperature	eq. to 5-9 μ atm
3.	Salinity	Compact CT	SSS	<i>In situ</i>	CTD	Salinity	± 0.001 °C
					Salinometer	Salinity	± 0.005 psu
4.	Chlorophyll-a	Compact CLW or Infinity CLW	Surface chlorophyll-a	<i>In situ</i>	CTD	Chl-a Fluorescence	± 0.003 psu
					Fluorometer	Chlorophyll-a	$\pm 0.2\%$
5.	Dissolved Oxygen (DO)	Compact DOW or Infinity AROW	Surface DO	<i>In situ</i>	CTD	DO	1% FS
					Titration	DO	± 3 μ mol/kg
6.	Nutrient	Auto analyzer	NO ₃ , NO ₂ , NH ₄ , PO ₄ and SiO ₂	<i>Ex situ</i>	-	-	-
7.	Suspended Sediment (SS)	Compact CLW or Infinity AROW	Surface SS	<i>In situ</i>	CTD	Turbidity	
					Filtering	SS	

Note: CTD used is CTD Sea-Bird SBE 911

Table 3. Underway (snapshot) averaged level of pCO₂ in the Makassar Strait.

No.	Year (x variable)	Average level (y variable)	Regression function	mean pCO ₂ ^{ATM}
1.	1984	$354.6 \pm 7.2 \mu\text{atm}$	$y = 1.01x + 9.53$	$344.8 \mu\text{atm}$
2.	2007	$366.5 \pm 10.7 \mu\text{atm}$	$y = 3.82x - 7288.3$	$366.5 \mu\text{atm}$
3.	2008	$373.3 \pm 8.6 \mu\text{atm}$	$\downarrow$	$365.4 \mu\text{atm}$
4.	2010	$386.4 \pm 10.2 \mu\text{atm}$	$\downarrow$	$381.9 \mu\text{atm}$
5.	2011	$387.6 \pm 3.8 \mu\text{atm}$		$376.3 \mu\text{atm}$
6.	2013	$390.0 \pm 13.4 \mu\text{atm}$		$383.3 \mu\text{atm}$

Figure 1. Compilation of the available $\delta p\text{CO}_2$ data (in μatm) from 1984-2013 according to cruise tracks (upper figure). Smaller-sized dots in the upper panel were obtained from expeditions other than those in this study. In the lower figure, the interpolation uses a 30-min grid and 150-km radius. Positive $\delta p\text{CO}_2$ values indicate that the ocean has a $p\text{CO}_2$ concentration that is significantly higher than the atmospheric $p\text{CO}_2$.

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Figure 2. Time series of the surface parameters measured during the EWIN 2010 cruise. From the upper to lower panel: (a) oceanic and atmospheric pCO₂ values; (b) δ pCO₂; (c) SST and SSS values; (d) Chl-a values and DO values; and (e) NO₃, PO₄ and SiO₂ concentrations. The Surabaya and Lombok Strait areas are located between the Java Sea and Flores-Banda Sea and are indicated with a blue line. The red line in (b) separates the pCO₂ sinks (negative) and sources (positive).

Figure 3. Time series of the surface parameters measured during the Banggai cruise (June-July 2011). From the upper to lower panel: (a) oceanic and atmospheric pCO₂ values; (b) δ pCO₂; (c) SST and SSS values; (d) Chl-a values and DO values; and (e) NO₃, PO₄ and SiO₂ concentrations. The Bali and Kangean areas are located between the Java Sea and Flores-Banda Sea and are indicated with a blue line. The red line in (b) separates the pCO₂ sinks (negative) and sources (positive).

Figure 4. Time series of the surface parameters measured during the EWIN 2013 cruise (June 2013). From the upper to lower panel: (a) oceanic and atmospheric pCO₂ values; (b) δ pCO₂; (c) SST and SSS values; (d) Chl-a values and DO values; and (e) NO₃, PO₄ and SiO₂ concentrations.

Figure 5. Interpolated deseasonalized pCO₂ values for the Makassar Strait, which shows an increasing trend of pCO₂ trend of 35.5 μ atm over 29 years (Figures A-F). The pCO₂-latitude plot is extracted from the interpolated cruise tracks (denoted by colored dots) and only uses the data from along the diagonal dashed black line.

Figure 6. Time series of the $\delta p\text{CO}_2$ and CO₂ flux values (left panel) and spatial pCO₂ levels (right panel) around the Java Sea. The cruise tracks on the right panel correspond to the time series for the left panels, which originated from Jakarta port. The left figures show the time series of the $\delta p\text{CO}_2$ and CO₂ flux values for the cruises in the 2000s. At the end of the La Niña event in April 2011, the ocean emitted less CO₂ gas to the atmosphere, whereas during a normal season, the Java Sea emitted $\sim 40 \mu\text{atm CO}_2$. The black dotted line denotes $\delta p\text{CO}_2 = 50 \mu\text{atm}$.

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Figure 7. Deffeyes diagram for the normalized surface TA and DIC values from different cruises denoted with different colors. The unit is in $\mu\text{mol kg}^{-1}$. The calculated pCO₂ contour was calculated using the average SSS (=32.4 psu) and SST (=29.6 °C) of five cruises.

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Figure 8. Time series of the measured (black line) and calculated (red line) pCO₂ values during the EWIN 2010 cruise. The calculated pCO₂ time series was measured as a function of the observed SST, SSS, NTA and NDIC. The NTA was normalized to TA(SSS=35)=2290, and the NDIC was normalized to DIC(SSS=35)=1964 $\mu\text{mol kg}^{-1}$.

Figure 9. Distribution of the sea surface temperatures (shaded) and 10-m wind above the ocean surface (vector) during the northwest monsoon season in (a) February 2010, (b) April 2010, and (c) April 2011. Wind and SST data are plotted from the averaged 6-hour NCEP/NCAR data obtained during the cruises. Temperature and wind speed units are in °C and m s⁻¹, respectively.

Java Sea → Flores Sea ← Banda Sea

■ 1984
 ■ 2007
 ■ 2008
 ■ 2009
 ■ 2011
 ■ 2013

Jakarta

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- Sby-Mks (Feb 2010)
 ● EWIN 2010 (Apr-May 2010)
- EWIN 2011 (Apr 2011)
 ● Banggai 2011 (Jun-Jul 2011)
- EWIN 2013 (Jun-Jul 2013)

(a) 13-15 February 2010

(b) 22 April - 1 May 2010

(c) 12-23 April 2011