RESEARCH LETTER

10.1002/2017GL074724

Key Points:

- $f\text{CO}_2$ temporal trend increases from south to north in ocean margins and boundary current-influenced areas
- Ocean margin $f\text{CO}_2$ responds to thermal and nonthermal effects differently in boundary current-influenced areas

Supporting Information:

- Supporting Information S1
- Software S1

Correspondence to:

H. Wang,
hongjie.wang@tamucc.edu

Citation:

Wang, H., X. Hu, W.-J. Cai, and B. Sterba-Boatwright (2017), Decadal $f\text{CO}_2$ trends in global ocean margins and adjacent boundary current-influenced areas, *Geophys. Res. Lett.*, 44, 8962–8970, doi:10.1002/2017GL074724.

Received 27 JUN 2017

Accepted 10 AUG 2017

Accepted article online 15 AUG 2017

Published online 9 SEP 2017

Decadal $f\text{CO}_2$ trends in global ocean margins and adjacent boundary current-influenced areas

Hongjie Wang¹ , Xinping Hu¹ , Wei-Jun Cai² , and Blair Sterba-Boatwright³
¹Department of Physical and Environmental Sciences, Texas A&M University-Corpus Christi, Corpus Christi, Texas, USA,

²School of Marine Science and Policy, University of Delaware, Newark, Delaware, USA, ³Department of Mathematics and Statistics, Texas A&M University-Corpus Christi, Corpus Christi, Texas, USA

Abstract Determination of the rate of change of sea surface CO_2 fugacity ($f\text{CO}_2$) is important, as the $f\text{CO}_2$ gradient between the atmosphere and the ocean determines the direction of CO_2 flux and hence the fate of this greenhouse gas. Using a newly available, community-based global CO_2 database (Surface Ocean CO_2 Atlas Version 3 coastal data set) and a newly developed statistical method, we report that the global ocean margins (within 400 km offshore, 30°S–70°N) $f\text{CO}_2$ temporal trends on decadal time scales ($1.93 \pm 1.59 \mu\text{atm yr}^{-1}$) closely follow the atmospheric $f\text{CO}_2$ increase rate ($1.90 \pm 0.06 \mu\text{atm yr}^{-1}$) in the Northern Hemisphere but are lower ($1.35 \pm 0.55 \mu\text{atm yr}^{-1}$) in the Southern Hemisphere, reflecting dominant atmospheric forcing in conjunction with different warming rates in the two hemispheres. In addition to the atmospheric $f\text{CO}_2$ forcing, a direct warming effect contributes more to $f\text{CO}_2$ increase in the western boundary current-influenced areas, while intensified upwelling contributes more to $f\text{CO}_2$ increase in eastern boundary current-influenced areas.

1. Introduction

The increase of the atmospheric concentration of carbon dioxide (CO_2) due to fossil fuel burning, cement production, and land use changes is causing changes in the global climate as supported by independent observations [Pachauri *et al.*, 2014]. The world ocean is an important long-term CO_2 sink and has absorbed approximately one third of anthropogenic CO_2 at a rate of $1.6\text{--}2.6 \text{ Pg C yr}^{-1}$ since the start of industrial revolution [Le Quéré *et al.*, 2015; Takahashi *et al.*, 2009], albeit with strong decadal and regional variations [Landschützer *et al.*, 2016]. However, the continuous uptake of CO_2 has resulted in increasing ocean surface $f\text{CO}_2$ (fugacity of CO_2) levels at rates ranging $1.5\text{--}1.9 \mu\text{atm yr}^{-1}$ [Bates *et al.*, 2014; Kitidis *et al.*, 2016; Takahashi *et al.*, 2009]. Therefore, studying seawater $f\text{CO}_2$ temporal trend is crucial to understanding how the oceanic CO_2 sink evolves under climate change conditions, as the thermodynamic gradient between the atmospheric $f\text{CO}_2$ and sea surface $f\text{CO}_2$ determines the direction and strength of CO_2 flux [McKinley *et al.*, 2016].

Most studies to date have focused on the open oceans, whereas little is known about the temporal evolution of $f\text{CO}_2$ in ocean margins that are subject to land-ocean interactions and boundary current influences. Although the ocean margins have a small surface area relative to the open ocean, they play a disproportionately important role in global CO_2 fluxes and total marine productivity [Liu *et al.*, 2010]. For example, the continental shelves (<200 m depth) only account for <8% of open ocean surface area, yet CO_2 flux there is about ~17% of the CO_2 flux of the open ocean [Bauer *et al.*, 2013; Cai, 2011]. However, $f\text{CO}_2$ in ocean margins typically exhibits large fluctuations on various time scales (daily to interannual), ranging from <100 μatm to >8000 μatm [Dai *et al.*, 2009; Frankignoulle and Borges, 2001; Guo *et al.*, 2009; Huang *et al.*, 2015]. Therefore, it is both critical and challenging to address how the CO_2 source and sink characteristics change in the ocean margins.

In this study, $f\text{CO}_2$ temporal trends in global ocean margins are calculated based on the Surface Ocean CO_2 Atlas (SOCAT) Version 3 Coastal data set [Bakker *et al.*, 2016]. The $f\text{CO}_2$ trends in western boundary current (WBC)- and eastern boundary current (EBC)-influenced areas are also discussed using both the west and east coasts of North America as examples. We also propose possible mechanisms that control the observed $f\text{CO}_2$ trends in different ocean margins.

2. Data and Methodology

2.1. Data

The $f\text{CO}_2$, sea surface salinity (SSS), sea surface temperature (SST), and sampling coordinates and date were obtained from the SOCAT Version 3 Coastal databases. The latitude ranged from 30°S to 70°N [Bakker *et al.*, 2016]. SOCAT $f\text{CO}_2$ data have been quality controlled following standard protocols, but SSS, SST, coordinates, and sampling date were reported by the data originators without further quality control in this study [Bakker *et al.*, 2016]. Conventional ocean margins or continental shelves were typically defined as within 200 m isobath. This definition results in unequal widths of ocean margin areas across the globe and limits land-ocean interaction in a narrow zone. In the EBC regions, in particular, many of the land-ocean interaction processes go beyond the 200 m depths [Gruber *et al.*, 2011]. SOCAT data set defines coastal areas using within 400 km from the coastline as a criterion in order to better reflect the environmental significance of these regions as continental margins [Pfeil *et al.*, 2012]. As a result, some of the grids that we examined include water depths as much as 7000 m at deep trenches (for example, in the North Pacific) where a continental plate joins with an oceanic plate. In addition, if we filtered out deep waters using a uniform 200 m depth, as much as 79.9% of grids (here $1^\circ \times 1^\circ$) would be removed. This would make our discussions on spatial distribution of $f\text{CO}_2$ trend much more data constrained and prevent us from drawing conclusions based on large-scale oceanic features such as the influence of boundary currents and warming on $f\text{CO}_2$ trends. More importantly, a 400 km limit allowed us to examine $f\text{CO}_2$ trends in important ocean boundary currents adjacent to the margins. Therefore, we chose not to further filter the SOCAT coastal $f\text{CO}_2$ database based on water depth.

Monthly dry air CO_2 ($x\text{CO}_2$, ppm) data were downloaded from NOAA's Earth System Research Laboratory (<http://www.esrl.noaa.gov/gmd/ccgg/trends/>), and the in situ monthly air $f\text{CO}_2$ was calculated based on averaged SSS, SST, and atmospheric pressure in each $1^\circ \times 1^\circ$ grid [Wang *et al.*, 2016].

$$f\text{CO}_{2\text{air}} = x\text{CO}_2 \times [P_b - P_w] \times e^{\left(\frac{P_b(B+2\delta)}{RT}\right)} \quad (1)$$

where P_b is the monthly average barometric pressure in the study area. P_w is saturated water vapor pressure calculated using monthly average salinity and temperature in the study area [Weiss and Price, 1980]. B , the first virial coefficient of CO_2 , and the cross virial coefficient δ are both functions of temperature [Weiss, 1974].

2.2. Trend Calculations

We used the Generalized Additive Mixed Modeling (GAMM) approach to analyze the $f\text{CO}_2$ trends [Wang *et al.*, 2016]. Briefly, the GAMM method predicted $f\text{CO}_2$ mainly based on three terms: seasonal cycle, the additional $f\text{CO}_2$ variation caused by SST and SSS changes, and the long-term sea surface $f\text{CO}_2$ change. A cyclic penalized spline was used to fit the seasonal cycle with the entire time series data in each grid. Second-order polynomial models were used to fit the nonlinear relationships of $f\text{CO}_2$ variation with SST and SSS. With the best fitted seasonal cycle and environmental variability, we then assumed the residual $f\text{CO}_2$ variation ($f\text{CO}_2$ trend $\times$ year) was a linear function of sampling date, and its coefficient represented the temporal $f\text{CO}_2$ trend. We consider the $f\text{CO}_2$ as having a significant long-term trend in this grid if the p value of this linear regression is less than 0.05.

In the GAMM method, autocorrelation was accounted for due to the lack of independence of consecutive observations taken close together in time (see supporting information). The method also weighted the observations using explicit models for heteroscedasticity to account for unequal variance in sampling date, SSS, or SST [Wang *et al.*, 2016]. Because of high variability, temporal trends based on short records (such as 10 years or less) are very sensitive to the beginning and end dates [McKinley *et al.*, 2011]. Thus, we only calculated the trends in grids that had more than 10 years of data record, and we reported the slope of trends with a significant regression (p value < 0.05). Grubbs' test [Grubbs, 1969] was adopted to detect and remove the $f\text{CO}_2$ trend outliers at significance level of 0.05 in each studied area, such as Northern Hemisphere or Southern Hemisphere and eastern boundary current-influenced or west boundary current-influenced areas.

2.3. Thermal and Nonthermal Trend Calculation

In order to separate the influence of temperature from the influences of all other physical, chemical, and biological processes on $f\text{CO}_2$ trend, we adopted the empirical relationship between temperature and $f\text{CO}_2$ ($\partial \ln f\text{CO}_2 / \partial T = 0.0423^\circ\text{C}^{-1}$) [Takahashi *et al.*, 2002] to calculate the thermal $f\text{CO}_2$ ($T\text{-}f\text{CO}_2$) and nonthermal

$f\text{CO}_2$ ($\text{NT}-f\text{CO}_2$) in every grid. The two components were calculated following equations (2) and (3), in which the over bar represents the mean value of SST, SSS, or $f\text{CO}_2$ in each grid.

$$T - f\text{CO}_2 = \overline{f\text{CO}_2} \times \exp[0.0423(\text{SST} - \overline{\text{SST}})] \quad (2)$$

$$\text{NT} - f\text{CO}_2 = f\text{CO}_2 \times \exp[0.0423(\overline{\text{SST}} - \text{SST})] \quad (3)$$

Then $T-f\text{CO}_2$ and $\text{NT}-f\text{CO}_2$ trends were calculated in the same manners as the $f\text{CO}_2$ trend with the exception that SST was not used in the GAMM.

2.4. CO_2 Sink/Source Categorization

For each grid, the mean $f\text{CO}_2$ level in surface ocean ($f\text{CO}_{2\text{ocean}}$) was compared with the mean $f\text{CO}_2$ level in the atmosphere ($f\text{CO}_{2\text{air}}$) during the same period. If $f\text{CO}_{2\text{ocean}}$ is lower than $f\text{CO}_{2\text{air}}$, the ocean is an overall CO_2 sink; otherwise, the ocean is a CO_2 source for the studied period. This study assessed the CO_2 sink/source changes by only comparing the sea surface and atmospheric $f\text{CO}_2$ trends. The studied grid would become a decreasing (or increasing) CO_2 sink when the oceanic $f\text{CO}_2$ trend is higher (or lower) than atmospheric $f\text{CO}_2$ trend. In contrast, the ocean would become an increasing (or decreasing) CO_2 source when the $f\text{CO}_2$ trend in the surface ocean is higher (or lower) than in atmosphere.

3. Results and Discussion

3.1. Global Average $f\text{CO}_2$ Increase Rate and the Latitudinal Variability

The calculated surface $f\text{CO}_2$ trend in the global ocean margin is $1.91 \pm 1.63 \mu\text{atm yr}^{-1}$ (Figure 1), with the middle 50% of the trend (mean $\pm 25\%$ of variance) values ranging between $1.28 \mu\text{atm yr}^{-1}$ and $2.47 \mu\text{atm yr}^{-1}$. These results are consistent with an earlier study in both the Japanese and European margins [Wang *et al.*, 2016]. In addition, the mean $f\text{CO}_2$ increase rates in the Northern ($0-70^\circ\text{N}$) and Southern Hemispheres ($0-30^\circ\text{S}$) are $1.93 \pm 1.59 \mu\text{atm yr}^{-1}$ and $1.35 \pm 0.55 \mu\text{atm yr}^{-1}$, respectively. If we only account for the tropical areas, $f\text{CO}_2$ increase rate in the low-latitude Northern Hemisphere ($0-30^\circ\text{N}$) is $1.65 \pm 0.92 \mu\text{atm yr}^{-1}$ and is still significantly higher than that in the Southern Hemisphere ($0-30^\circ\text{S}$) (Student's t test, $p = 0.03$). On the global scale, $f\text{CO}_2$ increase rates show significant latitudinal dependence ($p < 0.05$) for both the global ocean margins (Figure 1b), ocean margins within the Northern Hemisphere (see details in supporting information), and boundary current-influenced areas. Note that the temporal spans of data examined in this study are mainly between 10 and 30 years (Figure S2). Similar to the studies in the open ocean [Fay and McKinley, 2013; McKinley *et al.*, 2011], the $f\text{CO}_2$ trend variance is related to the time scale. However, the $f\text{CO}_2$ trend is not biased by the time scales selection in the ocean margins (see supporting information and Figure S3), and our 10 year trends are comparable with the longer-term trends.

The time scales used for calculating the atmospheric $f\text{CO}_2$ trends are the same as those for the seawater $f\text{CO}_2$ trends. Since the average increase in atmospheric $f\text{CO}_2$ in global ocean margins is $1.90 \pm 0.06 \mu\text{atm yr}^{-1}$, it is clear that the $f\text{CO}_2$ increase in the margins of the Southern Hemisphere ($1.35 \pm 0.55 \mu\text{atm yr}^{-1}$) is lower than the atmospheric trend (paired t test, $p < 0.001$). Note that the SOCAT coastal data set does not include the Southern Ocean; thus, this work does not cover middle- to high-latitude regions in the Southern Hemisphere ($>30^\circ\text{S}$). For the Northern Hemisphere margins, however, the seawater and atmospheric $f\text{CO}_2$ trends are statistically indistinguishable in all studied $1^\circ \times 1^\circ$ grids as a whole (paired t test, $p = 0.74$). Based on the differences in the atmospheric and ocean margin $f\text{CO}_2$ trends, the ocean margins in the Southern Hemisphere have become either a stronger CO_2 sink or a weaker CO_2 source over the past few decades, depending on the initial sea surface and atmospheric $f\text{CO}_2$ at the beginning of each time series [Wang *et al.*, 2016]. In contrast, the overall strengths of CO_2 sinks or sources in the Northern Hemisphere margins have not changed significantly over the past few decades if we assume that the gas transfer coefficient and gas solubility remain the same (Figure S4).

Further data analysis suggests that 75% of grids in the low-latitude ($30^\circ\text{S}-30^\circ\text{N}$) margins are an overall source of CO_2 due to higher seawater $f\text{CO}_2$ than the atmosphere, but this CO_2 source has weakened in 71% of these grids. Of the other 25% of the low-latitude grids that are acting as an overall CO_2 sink, 79% have become a stronger CO_2 sink over the past few decades (Figure 2). On the contrary, the majority of grids (88%) north of 30°N are CO_2 sinks, nearly half of which (47%) have become stronger, while the other half (53%) have

Figure 1. Global distribution of $f\text{CO}_2$ trends, their thermal and nonthermal components, and their respective latitudinal distributions. (a) Global distribution of $f\text{CO}_2$ trends, (c) global distribution of thermal $f\text{CO}_2$ trends, and (e) global distribution of nonthermal $f\text{CO}_2$ trends. The inserted figures in Figures 1a, 1c, and 1e are the density plots (the area under the curve is 1) of the calculated $f\text{CO}_2$, thermal $f\text{CO}_2$ and nonthermal $f\text{CO}_2$ trends. (b, d, and f) The p value is the statistical significance of the linear regression. (Null hypothesis states that the slope is equal to zero. Unit for color bars and y-axes is $\mu\text{atm yr}^{-1}$.)

Figure 2. The summary of CO_2 sink/source change in global margins.

Table 1. A Summary of $f\text{CO}_2$, Thermal $f\text{CO}_2$, and Nonthermal $f\text{CO}_2$ Trends in the North Pacific and North Atlantic Ocean Margins^a

Area	$f\text{CO}_2$ Trend	$T\text{-}f\text{CO}_2$ Trend	NT- $f\text{CO}_2$ Trend
California Current (15°N–45°N, 114°W–128°W)	2.34 ± 1.55	-1.17 ± 2.39	3.29 ± 3.53
Kuroshio Current (22°N–45°N, 130°E–148°E)	2.30 ± 1.71	-0.19 ± 1.26	2.55 ± 1.75
Canary Current (10°N–50°N, 21°W–9°E)	1.90 ± 1.55	0.05 ± 1.82	1.94 ± 2.65
Gulf Stream (28°N–45°N, 59°W–81°W)	1.19 ± 1.75	1.10 ± 1.97	0.61 ± 2.37

^aUnit is $\mu\text{atm yr}^{-1}$.

become weaker (Figure 2). Overall, 44% of all global ocean margin grids have seawater $f\text{CO}_2$ increase rates faster than the atmospheric trend, while 56% of grids have $f\text{CO}_2$ increase rates slower than the atmospheric trend. Therefore, there are more grids in the global margins becoming a decreasing source of CO_2 to the atmosphere.

On the global scale, the average decomposed nonthermal $f\text{CO}_2$ (NT- $f\text{CO}_2$) trend is $1.89 \pm 2.63 \mu\text{atm yr}^{-1}$ in the ocean margins, much higher than the average thermal $f\text{CO}_2$ ($T\text{-}f\text{CO}_2$) trend ($-0.20 \pm 1.88 \mu\text{atm yr}^{-1}$, $p < 0.001$) (see method for $f\text{CO}_2$ decomposition). These results suggest that on the global-scale, nonthermal effects are much greater than thermal effects in controlling ocean margin $f\text{CO}_2$ changes. While NT- $f\text{CO}_2$ trends can be caused by many physical (other than temperature change), chemical, and biological processes, we suggest that on a global scale, there is no significant difference between NT- $f\text{CO}_2$ and atmospheric $f\text{CO}_2$ increase rate (paired t test, $p > 0.05$). The agreement between NT- $f\text{CO}_2$ increase rate and the atmospheric $f\text{CO}_2$ increase rate indicates that air-sea gas exchange exerts the dominant control on coastal ocean $f\text{CO}_2$ increase, similar to the observations in the open ocean [Takahashi *et al.*, 2009]. However, NT- $f\text{CO}_2$ increase rates may be either lower or higher than the atmospheric $f\text{CO}_2$ increase rates in different regions, such as the boundary current-dominated systems. These regional differences will be further discussed in the following section.

Despite the overall small thermal effect, $T\text{-}f\text{CO}_2$ trends show significant latitudinal dependence on the global scale (Figures 1c and 1d); however, there is no latitudinal dependence for the NT- $f\text{CO}_2$ trends (Figures 1e and 1f). The apparent latitudinal dependence of $T\text{-}f\text{CO}_2$ may be explained by the differential warming rates across the globe. Overall, the calculated multidecadal (1971 to 2010) [Levitus *et al.*, 2009; Pachauri *et al.*, 2014] warming rates in the upper 75 m of the global ocean range from 0.09 to $0.13^\circ\text{C decade}^{-1}$, whereas the warming is more prominent in the Northern Hemisphere (0.248°C) than in the Southern Hemisphere (0.115°C) in the surface 700 m for the 1969–2008 period [Levitus *et al.*, 2009]. Note the global average SST change from 1990 to 2006 based on the buoy data is about $0.025^\circ\text{C yr}^{-1}$ [Stocker *et al.*, 2013]. Assuming that average $f\text{CO}_2$ in the surface water is the same as average $f\text{CO}_2$ in the atmosphere from 1990 to 2006 ($358 \mu\text{atm}$), the SST change could increase the $T\text{-}f\text{CO}_2$ trend by $358 \mu\text{atm} \times 0.025^\circ\text{C yr}^{-1} \times 4.23\% ^\circ\text{C}^{-1} = 0.38 \mu\text{atm yr}^{-1}$ in Northern Hemisphere and $0.19 \mu\text{atm yr}^{-1}$ in Southern Hemisphere based on the empirical relationship between temperature and $f\text{CO}_2$ change [Takahashi *et al.*, 2002]. The absolute $T\text{-}f\text{CO}_2$ trend caused by the SST change in the marginal seas should be different from the open ocean. Nevertheless, it appears that the magnitude of $f\text{CO}_2$ trend resulted from latitudinal gradient in SST can only partially explain the increasing $f\text{CO}_2$ trends from the Southern to the Northern Hemisphere (Figures 1c and 1d and supporting information). Therefore, more studies are needed to explain this latitudinal dependence of $f\text{CO}_2$ trends.

3.2. Boundary Current Effect on $f\text{CO}_2$ Increase Rate Variability

Most global ocean margins are influenced by boundary currents. Overall, the Pacific Ocean margins have significantly higher $f\text{CO}_2$ increase rates than the margins in the Atlantic Ocean (Figure 1 and Table 1). However, the trends of $T\text{-}f\text{CO}_2$ and NT- $f\text{CO}_2$ are drastically different between the EBC- and WBC-dominated margins within each ocean basin. For example, in the North Pacific margins (influenced by the Kuroshio Current versus California Currents), our calculations show that the $T\text{-}f\text{CO}_2$ trend is higher in the WBC than EBC (-0.19 ± 1.26 versus $-1.17 \pm 2.39 \mu\text{atm yr}^{-1}$), yet NT- $f\text{CO}_2$ trend is lower in WBC than its EBC counterpart (2.55 ± 1.75 versus $3.29 \pm 3.53 \mu\text{atm yr}^{-1}$, Table 1). The situation is similar in the North Atlantic margins (the Gulf Stream versus the Canary Current), where the $T\text{-}f\text{CO}_2$ trend is higher in WBC than EBC (1.10 ± 1.97 versus

Figure 3. The $f\text{CO}_2$ trend and its thermal and nonthermal components along the North American margins and a comparison in the two boundary current-dominated areas. (a) The $f\text{CO}_2$ trend, (c) thermal $f\text{CO}_2$ trend, and (e) nonthermal $f\text{CO}_2$ trend. The comparison of (b) $f\text{CO}_2$ trend, (d) thermal $f\text{CO}_2$ trend, and (f) nonthermal $f\text{CO}_2$ trend between EBC (15°N–45°N) and WBC (28°N–45°N) along the North American margins. Note that EBC- and WBC-dominated margins were noted in Figure 3a. The inserted figures in Figures 3a, 3c, and 3e are the density plots (the area under the curve is 1) of the calculated $f\text{CO}_2$, thermal $f\text{CO}_2$, and nonthermal $f\text{CO}_2$ trends. Unit for color bars and y-axes is $\mu\text{atm yr}^{-1}$.

$0.05 \pm 1.82 \mu\text{atm yr}^{-1}$), while the NT- $f\text{CO}_2$ trend is lower in WBC than its EBC counterpart (0.61 ± 2.37 versus $1.94 \pm 2.65 \mu\text{atm yr}^{-1}$, Table 1)

Due to the greater spatial and temporal $f\text{CO}_2$ data coverage, we limit the following discussion to the Northern Hemisphere and consider only the North American east and west coast margins (Figure 3) as examples to

interpret the contributions of different forcings (thermal and nonthermal) on the regional $f\text{CO}_2$ trends in WBCs and EBCs.

The rate of increase of seawater $f\text{CO}_2$ in surface water is significantly lower in the WBC (the Gulf Stream, $1.19 \pm 1.75 \mu\text{atm yr}^{-1}$) than the EBC (the California Current System, $2.34 \pm 1.55 \mu\text{atm yr}^{-1}$) (Figures 3a and 3b and Table 1). Additionally, the rate of increase of seawater $f\text{CO}_2$ is significantly lower than the atmospheric trend in the WBC (paired t test, $p = 0.008$) but significantly higher than the atmospheric trend in the EBC (paired t test, $p = 0.01$). Previous studies have shown that the margins of both the Gulf Stream and the California Current System are a net CO_2 sink [Hales *et al.*, 2012; Laruelle *et al.*, 2014; Signorini *et al.*, 2013], and we add that the sink capacity has likely changed differently in these regions, with increasing sink capacity in the Gulf Stream margins and decreasing sink capacity in the California Current System margins (Figure S5).

The difference in $T\text{-}f\text{CO}_2$ trends between the Gulf Stream and the California Current System in North American margins is caused by the different temperature changes in these areas. The WBCs are intensifying and shifting toward the poles as a long-term effect of global warming, which results in the surface ocean warming rates over the paths of WBCs ($0.1\text{--}0.3^\circ\text{C decade}^{-1}$) outpacing other regions [Wu *et al.*, 2012; Yang *et al.*, 2016]. For example, the warming rate of the Gulf Stream is as much as $\sim 0.9^\circ\text{C decade}^{-1}$ over the period of 2005–2014 [Robson *et al.*, 2016] even though some studies argue that warming in the Gulf Stream is slowing down from 2003 to 2012 based on coastal sea level observation [Ezer *et al.*, 2013]. Meanwhile, it is likely that global warming leads to increased upwelling intensity in EBCs [Bakun, 1990; Bakun *et al.*, 2010; Wang *et al.*, 2015]. The enhanced upwelling could compensate for the regional warming effect or even cause decreased SST in EBCs including the California Current System [Jacox *et al.*, 2014; Patti *et al.*, 2010], the Humboldt Current System, the Benguela Current System, and the northwest coast of Africa [Gutiérrez *et al.*, 2011; McGregor *et al.*, 2007; Santos *et al.*, 2012]. Therefore, the higher warming rates in the WBCs than EBCs cause the higher $T\text{-}f\text{CO}_2$ trend in the Gulf Stream and negative $T\text{-}f\text{CO}_2$ trend in the California Current System.

However, the higher warming rate does not necessarily lead to a higher $f\text{CO}_2$ trend (Table 1) because of the low $\text{NT-}f\text{CO}_2$ trend in the Gulf Stream, where the $\text{NT-}f\text{CO}_2$ trend is significantly lower than the atmospheric $f\text{CO}_2$ trend (paired t test, $p < 0.05$). It is likely that there are some other processes, in addition to atmospheric forcing, suppress the $\text{NT-}f\text{CO}_2$ trends in the Gulf of Stream. One possible process that leads to this low $\text{NT-}f\text{CO}_2$ trend in the Gulf Stream is the increasing primary productivity due to anthropogenic nutrient input, which is supported by the shallow-water ocean carbonate model [Andersson and Mackenzie, 2004] and the increasing annual chlorophyll concentrations from 1998 to 2014 [O'Brien, 2015]. Another likely contributor to the low $\text{NT-}f\text{CO}_2$ trend is the shorter average residence time in WBCs (0.31 ± 0.01 month) compared to that of EBCs (1.52 ± 0.22 month) [Bourgeois *et al.*, 2016]. The accumulation of anthropogenic CO_2 in the Gulf Stream would be limited by the short residence time and rapid cross-shelf water exchange, causing the sea surface $f\text{CO}_2$ trend to lag behind atmospheric $f\text{CO}_2$ change. Nevertheless, more studies are needed to determine the exact causes.

We note that the Kuroshio Current-influenced area has a substantially larger $f\text{CO}_2$ trend than the Gulf Stream system (Table 1). We suggest that the Kuroshio transverses 400 km of active margin with many trenches and thus encompasses more deep waters and much stronger upwelling than the Gulf Stream system where upwelling is limited to the shelf-break mixing [Liu *et al.*, 2010]. We also note that data are not available in the margin areas off the midlatitude eastern Asia. Thus, a strict comparison between this region and the Gulf Stream-influenced region is not appropriate.

While SST changes have a direct effect on $T\text{-}f\text{CO}_2$, $\text{NT-}f\text{CO}_2$ is influenced by upwelling, biological production, and increasing atmospheric $f\text{CO}_2$. Generally speaking, upwelling in ocean margin areas can enhance primary production by bringing up deep, cold, and nutrient-rich water to the surface [Demarcq, 2009; Gutiérrez *et al.*, 2011; Lachkar and Gruber, 2011]. Therefore, the increasing primary production in the California Current System due to upwelled nutrients should depress the $f\text{CO}_2$ trend. However, the intensified upwelling in this region also delivers CO_2 -enriched and low carbonate saturation state subsurface water to the surface [Feely *et al.*, 2008]. Both the $f\text{CO}_2$ and $\text{NT-}f\text{CO}_2$ trends are significantly higher than the atmospheric $f\text{CO}_2$ trend (paired t test, $p < 0.05$). It is likely that there are some other processes, in addition to atmospheric forcing, that can accelerate the $\text{NT-}f\text{CO}_2$ trend in the California Current System-dominated margin. The relatively high $f\text{CO}_2$

and NT- $f\text{CO}_2$ trends also show that the CO_2 uptake caused by enhanced photosynthesis probably cannot compensate the CO_2 increase due to atmospheric CO_2 invasion and upwelling of CO_2 -enriched subsurface water; this may be because the relatively low temperature in the California Current System inhibits primary production or the eddies transport nutrients out of the immediate coastal area [Gruber *et al.*, 2011; Renault *et al.*, 2016]. The CO_2 enrichment due to upwelling is so strong that the 30–40°N nearshore area in California Current System has even become an increasing CO_2 source (Figure S5), which could further exacerbate the effects of ocean acidification in this upwelling-dominated system [Gruber *et al.*, 2012; Hauri *et al.*, 2009].

Taken together, our results revealed the heterogeneity in CO_2 sink/source evolution in global ocean margins and the mechanisms that control $f\text{CO}_2$ trends in these systems. The findings have significant implications to understanding the temporal changes in CO_2 sinks and sources in global ocean margins and adjacent boundary current-influenced areas on multidecadal time scales. Due to the current sparse $f\text{CO}_2$ data coverage in the ocean margins and the highly heterogeneous nature of these margins, continued efforts in collecting and compiling additional data and incorporating them into the SOCAT database are necessitated. These efforts will lead to a better understanding of ocean margins and their role in the global carbon budget under future climate change conditions.

Acknowledgments

Funding for this study was provided by the Gulf of Mexico Research Initiative (RFP-II, GoMRI-020). Partial funding to H. W. as a graduate research assistant was provided by NOAA's NOS National Centre for Coastal Ocean Science (contract NA15NOS4780185). W.-J.C. thanks NSF, NASA, and NOAA for supporting his coastal ocean carbon cycle research. The Surface Ocean CO_2 Atlas (SOCAT) is an international effort, endorsed by the International Ocean Carbon Coordination Project (IOCCP), the Surface Ocean Lower Atmosphere Study (SOLAS) and the Integrated Marine Biogeochemistry and Ecosystem Research program (IMBER), to deliver a uniformly quality-controlled surface ocean CO_2 database. The SOCAT Version 3 Coastal databases used for this paper could be downloaded from <http://cdiac.ornl.gov/ftp/oceans/SOCATv3/>. The many researchers and funding agencies responsible for the collection of data and quality control are thanked for their contributions to SOCAT. We thank M. McCutcheon for her editorial assistance. Special thanks are given to the two anonymous reviewers for their insightful and constructive suggestions and comments, which helped to significantly improve the quality of this manuscript.

References

- Andersson, A. J., and F. T. Mackenzie (2004), Shallow-water oceans: A source or sink of atmospheric CO_2 ?, *Front. Ecol. Environ.*, 2(7), 348–353, doi:10.1890/1540-9295(2004)002[0348:SOASOS]2.0.CO;2.
- Bakker, D. C., B. Pfeil, C. S. Landa, N. Metz, K. M. O'Brien, A. Olsen, K. Smith, C. Cosca, S. Harasawa, and S. D. Jones (2016), A multi-decade record of high-quality $f\text{CO}_2$ data in version 3 of the Surface Ocean CO_2 Atlas (SOCAT), *Earth Syst. Sci. Data*, 8, 383–413, doi:10.5194/essd-8-383-2016.
- Bakun, A. (1990), Global climate change and intensification of coastal ocean upwelling, *Science*, 247(4939), 198–201.
- Bakun, A., D. B. Field, A. Redondo-Rodriguez, and S. J. Weeks (2010), Greenhouse gas, upwelling-favorable winds, and the future of coastal ocean upwelling ecosystems, *Global Change Biol.*, 16(4), 1213–1228, doi:10.1111/j.1365-2486.2009.02094.x.
- Bates, N., Y. Astor, M. Church, K. Currie, J. Dore, M. Gonaález-Dávila, L. Lorenzoni, F. Muller-Karger, J. Olafsson, and M. Santa-Casiano (2014), A time-series view of changing ocean chemistry due to ocean uptake of anthropogenic CO_2 and ocean acidification, *Oceanography*, 27(1), 126–141, doi:10.5670/oceanog.2014.16.
- Bauer, J. E., W.-J. Cai, P. A. Raymond, T. S. Bianchi, C. S. Hopkinson, and P. A. Regnier (2013), The changing carbon cycle of the coastal ocean, *Nature*, 504(7478), 61–70, doi:10.1038/nature12857.
- Bourgeois, T., J. C. Orr, L. Resplandy, J. Terhaar, C. Ethé, M. Gehlen, and L. Bopp (2016), Coastal-ocean uptake of anthropogenic carbon, *Biogeochemistry*, 13(14), 4167–4185, doi:10.5194/bg-13-4167-2016.
- Cai, W.-J. (2011), Estuarine and coastal ocean carbon paradox: CO_2 sinks or sites of terrestrial carbon incineration?, *Annu. Rev. Mar. Sci.*, 3(1), 123–145, doi:10.1146/annurev-marine-120709-142723.
- Dai, M., Z. Lu, W. Zhai, B. Chen, Z. Cao, K. Zhou, W.-J. Cai, and C.-T. A. Chen (2009), Diurnal variations of surface seawater $p\text{CO}_2$ in contrasting coastal environments, *Limnol. Oceanogr.*, 54(3), 735–745, doi:10.4319/lo.2009.54.3.0735.
- Demarcq, H. (2009), Trends in primary production, sea surface temperature and wind in upwelling systems (1998–2007), *Prog. Oceanogr.*, 83(1–4), 376–385, doi:10.1016/j.pocean.2009.07.022.
- Ezer, T., L. P. Atkinson, W. B. Corlett, and J. L. Blanco (2013), Gulf Stream's induced sea level rise and variability along the U.S. mid-Atlantic coast, *J. Geophys. Res. Oceans*, 118, 685–697, doi:10.1002/jgrc.20091.
- Fay, A. R., and G. A. McKinley (2013), Global trends in surface ocean $p\text{CO}_2$ from in situ data, *Global Biogeochem. Cycles*, 27, 541–557, doi:10.1002/gbc.20051.
- Feely, R. A., C. L. Sabine, J. M. Hernandez-Ayon, D. Ianson, and B. Hales (2008), Evidence for upwelling of corrosive “acidified” water onto the continental shelf, *Science*, 320(5882), 1490–1492, doi:10.1126/science.1155676.
- Frankignoulle, M., and A. Borges (2001), The European continental shelf as a significant sink for atmospheric carbon dioxide, *Global Biogeochem. Cycles*, 15(3), 569–576, doi:10.1029/2000GB001307.
- Grubbs, F. E. (1969), Procedures for detecting outlying observations in samples, *Technometrics*, 11(1), 1–21, doi:10.1080/00401706.1969.10490657.
- Gruber, N., C. Hauri, Z. Lachkar, D. Lohrer, T. L. Frölicher, and G.-K. Plattner (2012), Rapid progression of ocean acidification in the California Current System, *Science*, 337(6091), 220–223, doi:10.1126/science.1216773.
- Gruber, N., Z. Lachkar, H. Frenzel, P. Marchesiello, M. Munnich, J. C. McWilliams, T. Nagai, and G.-K. Plattner (2011), Eddy-induced reduction of biological production in eastern boundary upwelling systems, *Nat. Geosci.*, 4(11), 787–792, doi:10.1038/NGEO1273.
- Guo, X., M. Dai, W. Zhai, W.-J. Cai, and B. Chen (2009), CO_2 flux and seasonal variability in a large subtropical estuarine system, the Pearl River Estuary, China, *J. Geophys. Res.*, 114, G03013, doi:10.1029/2008JG000905.
- Gutiérrez, D., et al. (2011), Coastal cooling and increased productivity in the main upwelling zone off Peru since the mid-twentieth century, *Geophys. Res. Lett.*, 38, L07603, doi:10.1029/2010GL046324.
- Hales, B., P. G. Strutton, M. Saraceno, R. Letelier, T. Takahashi, R. Feely, C. Sabine, and F. Chavez (2012), Satellite-based prediction of $p\text{CO}_2$ in coastal waters of the eastern North Pacific, *Prog. Oceanogr.*, 103, 1–15, doi:10.1016/j.pocean.2012.03.001.
- Hauri, C., N. Gruber, G.-K. Plattner, S. Alin, R. A. Feely, B. Hales, and P. A. Wheeler (2009), Ocean acidification in the California Current System, *Oceanography*, 22(4), 60–71, doi:10.5670/oceanog.2009.97.
- Huang, W.-J., W.-J. Cai, Y. Wang, S. E. Lohrenz, and M. C. Murrell (2015), The carbon dioxide system on the Mississippi River-dominated continental shelf in the northern Gulf of Mexico: 1. Distribution and air-sea CO_2 flux, *J. Geophys. Res. Oceans*, 120, 1429–1445, doi:10.1002/2014JC010498.

- Jacox, M. G., A. M. Moore, C. A. Edwards, and J. Fiechter (2014), Spatially resolved upwelling in the California Current System and its connections to climate variability, *Geophys. Res. Lett.*, *41*, 3189–3196, doi:10.1002/2014GL059589.
- Kitidis, V., I. Brown, N. Hardman-Mountford, and N. Lefèvre (2016), Surface ocean carbon dioxide during the Atlantic Meridional transect (1995–2013); Evidence of ocean acidification, *Prog. Oceanogr.*, doi:10.1016/j.pocean.2016.08.005, in press.
- Lachkar, Z., and N. Gruber (2011), What controls biological production in coastal upwelling systems? Insights from a comparative modeling study, *Biogeosciences*, *8*(10), 2961–2976, doi:10.5194/bg-8-2961-2011.
- Landschützer, P., N. Gruber, and D. C. E. Bakker (2016), Decadal variations and trends of the global ocean carbon sink, *Global Biogeochem. Cycles*, *30*, 1396–1417, doi:10.1002/2015GB005359.
- Laruelle, G. G., R. Lauerwald, B. Pfeil, and P. Regnier (2014), Regionalized global budget of the CO₂ exchange at the air-water interface in continental shelf seas, *Global Biogeochem. Cycles*, *28*, 1199–1214, doi:10.1002/2014GB004832.
- Le Quééré, C., et al. (2015), Global carbon budget 2015, *Earth Syst. Sci. Data*, *7*(2), 349–396, doi:10.5194/essd-7-349-2015.
- Levitus, S., J. I. Antonov, T. P. Boyer, R. A. Locarnini, H. E. Garcia, and A. V. Mishonov (2009), Global ocean heat content 1955–2008 in light of recently revealed instrumentation problems, *Geophys. Res. Lett.*, *36*, L07608, doi:10.1029/2008GL037155.
- Liu, K.-K., L. Atkinson, R. Quiñones, and L. Talaue-McManus (2010), *Carbon and Nutrient Fluxes in Continental Margins: A Global Synthesis*, Springer, Berlin, doi:10.1007/978-3-540-92735-8_1.
- McGregor, H. V., M. Dima, H. W. Fischer, and S. Mülitz (2007), Rapid 20th-century increase in coastal upwelling off Northwest Africa, *Science*, *315*(5812), 637–639, doi:10.1126/science.1134839.
- McKinley, G. A., A. R. Fay, T. Takahashi, and N. Metzl (2011), Convergence of atmospheric and North Atlantic carbon dioxide trends on multidecadal timescales, *Nat. Geosci.*, *4*(9), 606–610, doi:10.1038/NNGEO1193.
- McKinley, G. A., D. J. Pilcher, A. R. Fay, K. Lindsay, M. C. Long, and N. S. Lovenduski (2016), Timescales for detection of trends in the ocean carbon sink, *Nature*, *530*(7591), 469–472, doi:10.1038/nature16958.
- O'Brien, T. (2015), Indicator: Ocean chlorophyll concentrations. [Available at <http://www.globalchange.gov/browse/indicators/indicator-ocean-chlorophyll-concentrations>.]
- Pachauri, R. K., M. R. Allen, V. Barros, J. Broome, W. Cramer, R. Christ, J. Church, L. Clarke, Q. Dahe, and P. Dasgupta (2014), Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC.
- Patti, B., C. Guisande, I. Riveiro, P. Thejll, A. Cuttitta, A. Bonanno, G. Basilone, G. Buscaino, and S. Mazzola (2010), Effect of atmospheric CO₂ and solar activity on wind regime and water column stability in the major global upwelling areas, *Estuar. Coast. Shelf Sci.*, *88*(1), 45–52, doi:10.1016/j.ecss.2010.03.004.
- Pfeil, B., A. Olsen, D. C. Bakker, S. Hankin, H. Koyuk, A. Kozyr, J. Malczyk, A. Manke, N. Metzl, and C. Sabine (2012), A uniform, quality controlled Surface Ocean CO₂ Atlas (SOCAT), *Earth Syst. Sci. Data Discuss.*, *5*(2), 735–780.
- Renault, L., C. Deutsch, J. C. McWilliams, H. Frenzel, J.-H. Liang, and F. Colas (2016), Partial decoupling of primary productivity from upwelling in the California Current system, *Nat. Geosci.*, *9*(7), 505–508, doi:10.1038/ngeo2722.
- Robson, J., P. Ortega, and R. Sutton (2016), A reversal of climatic trends in the North Atlantic since 2005, *Nat. Geosci.*, *9*(7), 513–517, doi:10.1038/ngeo2727.
- Santos, F., M. Gomez-Gesteira, M. deCastro, and I. Alvarez (2012), Differences in coastal and oceanic SST trends due to the strengthening of coastal upwelling along the Benguela current system, *Cont. Shelf Res.*, *34*, 79–86, doi:10.1016/j.csr.2011.12.004.
- Signorini, S. R., A. Mannino, R. G. Najjar, M. A. Friedrichs, W. J. Cai, J. Salisbury, Z. A. Wang, H. Thomas, and E. Shadwick (2013), Surface ocean pCO₂ seasonality and sea-air CO₂ flux estimates for the North American east coast, *J. Geophys. Res. Oceans*, *118*, 5439–5460, doi:10.1002/jgrc.20369.
- Stocker, T., D. Qin, G. Plattner, M. Tignor, S. Allen, J. Boschung, A. Nauels, Y. Xia, B. Bex, and B. Midgley (2013), IPCC, 2013: Climate change 2013: The physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge Univ. Press.
- Takahashi, T., et al. (2002), Global sea–air CO₂ flux based on climatological surface ocean pCO₂, and seasonal biological and temperature effects, *Deep Sea Res. Pt II*, *49*(9–10), 1601–1622, doi:10.1016/S0967-0645(02)00003-6.
- Takahashi, T., S. C. Sutherland, R. Wanninkhof, C. Sweeney, R. A. Feely, D. W. Chipman, B. Hales, G. Friederich, F. Chavez, and C. Sabine (2009), Climatological mean and decadal change in surface ocean pCO₂, and net sea–air CO₂ flux over the global oceans, *Deep Sea Res., Part II*, *56*(8), 554–577, doi:10.1016/j.dsr2.2008.12.009.
- Wang, D., T. C. Gouhier, B. A. Menge, and A. R. Ganguly (2015), Intensification and spatial homogenization of coastal upwelling under climate change, *Nature*, *518*(7539), 390–394, doi:10.1038/nature14235.
- Wang, H., X. Hu, and B. Sterba-Boatwright (2016), A new statistical approach for interpreting oceanic fCO₂ data, *Mar. Chem.*, *183*, 41–49, doi:10.1016/j.marchem.2016.05.007.
- Weiss, R., and B. Price (1980), Nitrous oxide solubility in water and seawater, *Mar. Chem.*, *8*(4), 347–359.
- Weiss, R. F. (1974), Carbon dioxide in water and seawater: The solubility of a non-ideal gas, *Mar. Chem.*, *2*(3), 203–215, doi:10.1016/0304-4203(74)90015-2.
- Wu, L., et al. (2012), Enhanced warming over the global subtropical western boundary currents, *Nat. Clim. Chang.*, *2*(3), 161–166, doi:10.1038/nclimate1353.
- Yang, H., G. Lohmann, W. Wei, M. Dima, M. Ionita, and J. Liu (2016), Intensification and poleward shift of subtropical western boundary currents in a warming climate, *J. Geophys. Res. Oceans*, *121*, 4928–4945, doi:10.1002/2015JC011513.