

**Air-sea exchange of
CO₂ at a Northern
California coastal site**

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Air-sea exchange of CO₂ at a Northern California coastal site along the California Current upwelling system

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Abstract

Uncertainty in the air-sea CO₂ exchange (CO₂ flux) in coastal upwelling zones is attributed to high temporal variability, which is caused by changes in ocean currents. Upwelling transports heterotrophic, CO₂ enriched water to the surface and releases CO₂ to the atmosphere, whereas the presence of nutrient-rich water at the surface supports high primary production and atmospheric CO₂ uptake. To quantify the effects of upwelling on CO₂ fluxes, we measured CO₂ flux at a coastal upwelling site off of Bodega Bay, California, during the summer of 2007 and the fall of 2008 using the eddy covariance technique and the bulk method with *p*CO₂ measurements from November 2010 to July 2011. Variations in sea surface temperatures (SST) and alongshore wind speeds suggest that the measurement period in 2007 coincided with a typical early-summer upwelling period and the measurement period in 2008 was during a typical fall relaxation period. A strong source of CO₂ ($\sim 1.5 \pm 7$ SD (standard deviation) g C m⁻² day⁻¹) from the ocean to the atmosphere during the upwelling period was concurrent with high salinity, low SST, and low chlorophyll density. In contrast, a weak source of CO₂ flux ($\sim 0.2 \pm 3$ SD g C m⁻² day⁻¹) was observed with low salinity, high SST and high chlorophyll density during the relaxation period. Similarly, the sink and source balance of CO₂ flux was highly related to salinity and SST during the *p*CO₂ measurement periods; high salinity and low SST corresponded to high *p*CO₂, and vice versa. We estimated that the coastal area off Bodega Bay was likely a source of CO₂ to the atmosphere based on the following conclusions: (1) the overall CO₂ flux estimated from both eddy covariance and *p*CO₂ measurements showed a source of CO₂; (2) although the relaxation period during the 2008 measurements were favorable to CO₂ uptake, CO₂ flux during this period was still a slight source, (3) salinity and SST were found to be good predictors of the CO₂ flux for both eddy covariance and *p*CO₂ measurements, and historical data of daily averaged SST and salinity between 1988 to 2011 show that 99% of the data falls within the range of our observation in May–June 2007, August–September 2008 and November 2010–July 2011 indicating that our data set was representative of the

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annual variations in the sea state. Based on the developed relationship between $p\text{CO}_2$ and SST and salinity, the average annual CO_2 flux between 1988 and 2011 was estimated to be $\sim 35 \text{ mol C m}^{-2} \text{ yr}^{-1}$. The peak monthly CO_2 flux of $\sim 7 \text{ mol C m}^{-2} \text{ month}^{-1}$ accounted for about 30 % of the dissolved inorganic carbon in the surface mixed-layer.

1 Introduction

Intensive efforts have been underway over the last few decades to better understand and quantify global air-sea CO_2 exchange. These investigations reveal that the oceans absorb approximately one-third of the global anthropogenic CO_2 emissions (Gruber et al., 2009; Takahashi et al., 2009), and have accumulated 118 (± 19) PgC of carbon emitted by fossil fuel combustion, land use change, and cement production between 1800 and 1994 (Sabine et al., 2004). However, current estimates of the global ocean CO_2 uptake still contain considerable uncertainty, and more observations are necessary to better understand this critical carbon reservoir (Doney et al., 2009). One of the largest uncertainties in the global carbon cycle is the role of coastal seas, which potentially account for 30 % of the net global sink of CO_2 from the atmosphere to the ocean (Chen and Borges, 2009). Current global estimates do not account for the variability of the coastal ocean due to the difficulties of quantifying coastal CO_2 fluxes (Takahashi et al., 2009). In order to better understand the dynamics of coastal CO_2 fluxes, we must understand the physical and biological processes unique to coastal waters that control these fluxes.

Coastal upwelling is an important process that conflates the physical and biological influences on the coastal carbon cycle (Dugdale et al., 2006; Gattuso et al., 1998). Upwelling conveys deep nutrient-rich water to the surface and supports diverse coastal ecosystems. Although the surface of coastal upwelling zones covers only 1 % of the global ocean, these areas support approximately 50 % of the fishery industry because of their high primary productivity (Gattuso et al., 1998). Observational studies indicate that upwelling along the California coast has increased over the past 30 yr

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(Garcia-Reyes and Largier, 2010). This intensification of upwelling is likely related to increasing greenhouse gases, as the increasing temperature difference between land and sea accelerates coastal upwelling (Bakun, 1990; Diffenbaugh et al., 2004).

The sign of the effects of coastal upwelling on CO₂ fluxes has been debated. Upwelling provides a source of CO₂ from the ocean to the atmosphere because deep water is enriched with respired CO₂. At the same time, upwelling carries nutrients to the surface that enhance the biological uptake of CO₂ (Borges and Frankignoulle, 2002). Thus, the net CO₂ flux in upwelling zones averaged for inter-seasonal scales can be a sink of CO₂ from the atmosphere to the ocean (Evans et al., 2011; Gago et al., 2003; Hales et al., 2005; Ianson and Allen, 2002). The source and sink pulses of CO₂ flux reported in past studies range from -10 to 30 molCm⁻²yr⁻¹ (Copin-Montegut and Raimbault, 1994; Friederich et al., 2002; Goyet et al., 1998; Hales et al., 2005; Ianson and Allen, 2002; Ianson et al., 2009; Kelley and Hood, 1971; Lendt et al., 2003; Simpson and Zirino, 1980; Torres et al., 1999), using the sign convention of positive fluxes into the atmosphere from the ocean, and they are frequent two orders magnitude greater than the global average net CO₂ flux of approximately -0.4 molCm⁻²yr⁻¹ (Takahashi et al., 2009). This emphasizes the importance of understanding CO₂ fluxes in the complex coastal zones where upwelling occurs. Despite its importance, uncertainty in determining the net CO₂ flux in upwelling zones remains due to the spatial and temporal sparsity of the available measurements.

Another uncertainty in the CO₂ flux of coastal upwelling zones is attributed to the fact that a standard measurement technique for air-sea CO₂ flux is still under development. Quantifications of the spatial and temporal variations of CO₂ flux in upwelling zones were made, for example, by Friederich et al. (2002) for the California coast and Evans et al. (2011) for the Oregon coast. However, these past studies estimated the CO₂ flux using the bulk method based on the partial pressure of CO₂ (pCO₂) measured in surface waters. Currently, the gas transfer coefficient required for the bulk method is determined solely by empirical relationships between gas transfer coefficients and wind speed, which were developed in the open ocean where wind speeds are generally

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much higher than coastal areas (Ho et al., 2006; Wanninkhof and McGillis, 1999). Under lower wind speeds, the effects of factors such as surface films and rain may be important as well for determining gas transfer properties (Frew et al., 2004; Jähne et al., 1987), and the effects of breaking waves and bubbles may be especially influential in coastal seas (Keeling, 1993).

The eddy covariance technique utilized in this study is considered to be the most direct CO₂ flux measurement method available. The application of the technique to ocean fluxes is still challenging due to the fact that data processing introduces high uncertainty to the measurement of smaller fluxes (less than 1 gCm⁻² day⁻¹) and the sensors are sensitive to contamination in marine environments (Else et al., 2011; Iwata et al., 2005; Prytherch et al., 2010). However, frequent maintenance and the expected large magnitude of the CO₂ fluxes from coastal upwelling zones enabled us to employ the open path eddy covariance technique.

The objective of this study was to quantify the CO₂ flux from the wind-driven upwelling zone near Bodega Bay, California, and evaluate the physical and biological effects of upwelling on the observed near-shore CO₂ flux. We hypothesized that the sink or source characteristics of the coastal region are determined by a balance between the transports of CO₂ enriched water to the surface and high primary productivity. To test this hypothesis, we measured CO₂ fluxes using the eddy covariance technique and the bulk transfer method with pCO₂ measurements and compared the observed CO₂ fluxes with environmental variables related to upwelling events (e.g. wind speed, salinity, sea temperature) and primary production (e.g. light and nutrient availability and chlorophyll density).

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2 Methods

2.1 Site descriptions

The study site was located at the intertidal zone in the vicinity of the Bodega Bay Marine Laboratory (BML) in Northern California (Fig. 1). The coastal sea of Northern California is classically characterized by northerly winds in the summer due to the lateral pressure gradient (Dorman et al., 2000) established by the Pacific High offshore. The predominant alongshore wind causes Ekman transport along with lateral shear vorticity, which generates positive curl in the surface stress, resulting in strong coastal upwelling. Spatial distributions of water temperature indicated that our study area was particularly favorable for wind-driven upwelling events (Day and Faloon, 2009). The region's wind-driven upwelling cycle, nutrient availability and subsequent primary production have been described in CoOP WEST (Wind Events and Shelf Transport) studies (Dugdale et al., 2006; Largier et al., 2006; Wilkerson et al., 2006). The region's typically strong winds are periodically interrupted by a period with weaker winds known as a relaxation period, which lasts for ~3–7 days. The repeating cycles of upwelling and relaxation create a unique biological system. Upwelling carries deep water that is relatively cold and nutrient-rich to the surface, but increased turbidity (Day and Faloon, 2009) and lateral exports of nutrients inhibit production, while nutrients brought up to the surface are consumed during the subsequent relaxation period (Largier et al., 2006; Wilkerson et al., 2006).

The year of 2007 and 2008, when the eddy covariance measurements operated, were neutral to El Niño-Southern Oscillation. The winter of 2010, when $p\text{CO}_2$ measurements were operated, was characterized as a moderate El Niño condition.

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2.2 Eddy covariance measurements

2.2.1 Data collection

Eddy covariance CO₂ flux measurements were recorded from 28 May to 5 June in 2007 and from 31 August to 25 September in 2008. A three-dimensional sonic anemometer (C-SAT3, Campbell Scientific, USA) and an open path infrared gas analyzer (ATDD, NOAA, USA in 2007 (Auble and Meyers, 1992) and LI-7500, Li-Cor, USA in 2008) were mounted on a 13 m tower within an intertidal zone (38°18'58" N, 123°04'20" W). The density of CO₂ and H₂O, three dimensional wind components, and sonic temperature were recorded at 10 Hz. The data were stored in a compact flash memory card in a datalogger (CR1000; Campbell) and manually collected.

2.2.2 CO₂ flux calculations

Mixing ratios of CO₂ and H₂O to dry air were computed for each 10 Hz data point based on the ideal gas law; air temperature at each 10 Hz data point was computed from the sonic temperature and the vapor pressure calculated from the H₂O density, and the dry air pressure at each 10 Hz data point was computed from the vapor pressure and atmospheric pressure. High frequency data were not available for atmospheric pressure, so standard atmospheric pressure was used.

Each 10 Hz measurement of the mixing ratios of CO₂ and H₂O, 3-D wind velocity, and sonic temperature were filtered by a de-spiking algorithm to remove outliers when a datum was greater or less than the median of the 20 adjacent data points $\pm 6\times$ their standard deviation (Burba and Anderson, 2010). About 0.02 % of the 10 Hz data points were filtered out and interpolated.

The data stream was divided into 30-min intervals, with the validity of the 30-min averaging period tested using an Ogive function (Foken and Wichura, 1996). The first and second axis-rotation schemes were applied on the three dimensional wind speeds (Baldocchi et al., 1988) for each 30-min data set. The first axis-rotation calculates a wind

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vector along a mean wind direction, and the second axis-rotation computes 3-D wind vectors to align the mean vertical wind speed to zero. After the axis rotation, the time lag between the infrared gas analyzer and the sonic anemometer was estimated individually for every 30 min period. Within a -2.0 to $+2.0$ s range, the lag was chosen to maximize the covariance between the CO_2 mixing ratio and the vertical wind speed. On average, the CO_2 mixing ratio measurement had to be advanced by 0.6 s to maximize the correlation with the vertical wind speed measurement. The CO_2 flux was calculated as the covariance between the CO_2 mixing ratio and the vertical wind speed every 30 min (Webb et al., 1980).

2.2.3 Corrections

The effects of the sensor-heating correction (Burba et al., 2008) and cross-sensitivity correction were evaluated. Since the temperature of the infrared gas analyzer detector cell is maintained above the ambient temperature and radiant heating can also increase the temperature of the sensor, the surface of the analyzer can be warmer than its surroundings. Fluctuations of air temperature caused by the hot surface of the analyzer can cause a negative correlation between CO_2 density and the vertical wind speed, resulting in a false sink of CO_2 flux (Burba et al., 2008). The sensor-heating correction (method (4) described in Burba et al., 2008) was applied to all data. Some uncertainties in applying the correction are as follows; (1) the sensor was oriented nearly vertically but not perfectly, whereas the correction method developed by Burba et al. (2008) assumes that the sensor is exactly vertical; (2) we did not measure the temperature of the sensor surface, and it had to be estimated by empirical equations (method (4) in Burba et al., 2008); (3) the NOAA gas analyzer was used for the measurement in 2007 instead of the LI-7500 gas analyzer, and an appropriate correction for the NOAA gas analyzer is most probably different from that for the LI-7500 gas analyzer. Regardless of these uncertainties, we used the same correction as for the LI-7500 to be consistent. The relationship between CO_2 flux with heat correction and CO_2 flux without the heat

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correction was as follow.

$$F_{cc} \text{ (g C m}^{-2} \text{ day}^{-1}) = 1.03 \times F_{cw} + 0.76 \text{ (} R^2 = 0.99 \text{)} \quad (1)$$

for the NOAA gas analyzer, and

$$F_{cc} \text{ (g C m}^{-2} \text{ day}^{-1}) = 0.97 \times F_{cw} + 0.89 \text{ (} R^2 = 0.92 \text{)} \quad (2)$$

for the LI-7500 gas analyzer. F_{cc} is CO_2 flux with heat correction and F_{cw} is CO_2 flux without heat correction.

Contamination, especially by sea spray, on the mirror surface of the infrared gas analyzer can potentially cause a false negative correlation between CO_2 density and relative humidity (Prytherch et al., 2010). Prytherch et al. (2010) attributed the high magnitude of the CO_2 flux often observed by the eddy covariance technique over the ocean to this cross-sensitivity, and suggested an algorithm to estimate the uncontaminated slope of the CO_2 mixing ratio to humidity. However, our sensors were cleaned at least every 3 days, and the sensitivity of the CO_2 mixing ratio to humidity in our data ($\sim 0.0025 \text{ (mmol}_{\text{CO}_2} \text{ mol}_{\text{air}}^{-1}) (\%_{\text{humid}} 100^{-1})$) was much less than that reported in Prytherch et al., (2010) ($\sim 0.07 \text{ (mmol}_{\text{CO}_2} \text{ mol}_{\text{air}}^{-1}) (\%_{\text{humid}} 100^{-1})$). Else et al. (2011) reported that the application of the cross-sensitivity correction lead to large scatter in the data. We also encountered the same problem, (CO_2 flux with cross-sensitivity correction) = $0.63 \times (\text{CO}_2 \text{ flux without the correction}) + 0.80$, ($R^2 = 0.15$). Consequently the correction was not applied to our study.

2.2.4 Quality controls

Data quality was assessed using diagnostic values provided by the infrared gas analyzer and the sonic anemometer (9%: numbers in parentheses indicate the data amount filtered out by each process). After the sonic anemometer data was filtered using the diagnostic value, data during offshore winds, wind directions of 310° – 160° (where a wind direction of 0° indicates a northerly winds and the degree increases

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clockwise), were filtered out (39 %). The double-differenced time series using the median of absolute deviation about the median was then used to filter out outliers (10 %) (Sachs, 1996), with the parameter z in the outlier detector set as 4 (Papale et al., 2006). Finally, apparent outliers (larger than $30 \text{ g C m}^{-2} \text{ day}^{-1}$ or lower than $-30 \text{ g C m}^{-2} \text{ day}^{-1}$) were also filtered out (1 %). After all data filtering processes were complete, 48 % of the CO_2 fluxes remained for further analysis.

Cospectra of CO_2 and temperature with the vertical wind speed were evaluated to assess data quality (Fig. 2). Based on surface layer similarity theory, within the inertial subrange the logarithmic cospectra of scalar and vertical wind speed follows a $-4/3$ slope against normalized frequency (Kaimal et al., 1972). Although the theoretical characteristics became obscure under less stationary conditions, the overall structure resembled the ideal curve (Fig. 2). The cospectra did not vary noticeably with atmospheric stability, based on analysis of z/L , where z is measurement height and L is the Monin-Obukhov length.

Footprint analysis (Schuepp et al., 1990) performed on the quality-controlled fluxes indicated that 90 % of the fluxes originated from within 3 km of the coast. The 30 m of land between the tower base and the ocean accounted for less than 0.01 % of the flux footprint, even under conditions with the smallest flux.

2.3 Environmental data and the Bodega Ocean Observing Node (BOON)

Wind speed and wind direction were measured with the same sonic anemometer used for the eddy covariance measurements. Datasets of air temperature, wind speed, wind direction, Photosynthetic Photon Flux Density (PPFD), sea surface temperature (SST), salinity, and chlorophyll density were obtained from the Bodega Ocean Observing Node (BOON, <http://www.bml.ucdavis.edu/>). Air temperature, wind speed, wind direction, and PPFD had been recorded every hour at a meteorological tower located 30 m inland from the intertidal zone ($38^\circ 19' \text{ N}$, $123^\circ 04' \text{ W}$) since 1988, and SST, salinity, and chlorophyll density had been measured on a buoy (BML buoy) located 1.3 km offshore ($38^\circ 18' \text{ N}$, $123^\circ 04' \text{ W}$) of our study site. SST and salinity had been measured

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since 1988, and chlorophyll data was available only from 2005 to 2008. The $p\text{CO}_2$ was measured every hour by a SAMI- $p\text{CO}_2$ (Sun-Burst Sensors, USA) colorimetric sensor mounted on the buoy from November 2010 to July 2011. The measurement was temporarily halted due to a system malfunction in December 2010 through February 2011.

5 The sensor was calibrated by the factory within the range 150–700 μatm . All the data used in this study are summarized in Table 1.

2.4 CO_2 flux estimation with $p\text{CO}_2$ and atmospheric CO_2

CO_2 flux (F_c) was calculated from $p\text{CO}_2$ (C_{pw}), SST, salinity, and atmospheric CO_2 pressure (C_{pa}) with the bulk method for each $p\text{CO}_2$ measurement based on the follow-
10 ing equation (e.g. Wanninkhof and McGillis, 1999).

$$F_c = \alpha k [C_{pa} - C_{pw}] \quad (3)$$

The solubility, α , was computed by SST and salinity based on Weiss (1974); the gas transfer coefficient, k , was determined to be the average value calculated following Wanninkhof and McGillis (1999) (hereafter, W&M99), Wanninkhof (1992) (hereafter, W92), Ho et al. (2006) (hereafter, Ho06), Nightingale et al. (2000) (hereafter, N00),
15 and Sweeney et al. (2007) (hereafter, Sw07). The equations for k (md^{-1}) are as follows:

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[W&M99]

$$k = 0.0068u^3 \times (Sc/660)^{-0.5}, \quad (4)$$

[W92]

$$k = 0.074u^2 \times (Sc/660)^{-0.5}, \quad (5)$$

[Ho06]

$$k = 0.061u^2 \times (Sc/660)^{-0.5}, \quad (6)$$

[N00]

$$k = [0.053u^2 + 0.024u] \times (Sc/660)^{-0.5}, \quad (7)$$

[Sw07]

$$k = 0.065u^2 \times (Sc/660)^{-0.5}, \quad (8)$$

where, u is wind speed and Sc is Schmidt number (Wanninkhof, 1992).

Data collected monthly by the NOAA/ESRL Carbon Cycle Greenhouse Gases (CCGG) group's aircraft program were used to estimate C_{pa} . The nearest routine sampling location is offshore near Trinidad Head, CA, which is located about 300 km north of Bodega Bay ($\sim 41^\circ 10' N$, $124^\circ 20' W$). The lowest altitude data (near 300 m a.s.l.) from each month since September 2003 were used to estimate the average monthly atmospheric CO_2 concentration in the marine boundary layer throughout the region.

The CO_2 concentration was converted to the pressure, C_{pa} under the standard atmosphere.

2.5 Effective upwelling and National Data Buoy Center 46013 data

The National Data Buoy Center (NDBC) 46013 buoy is located off of Bodega Bay, California ($38^\circ 14' N$, $123^\circ 18' W$). The Effective Upwelling Index (EUI) was introduced by Garcia-Reyes (2011) as a proxy for the cumulative NO_3 . It is estimated using the sea surface temperature measured at the NDBC 46013 buoy (SST_{N13}) from the National

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Data Buoy Center (<http://www.ndbc.noaa.gov>). To identify upwelling periods, NO_3 concentrations were estimated based on the NO_3 - SST_{N13} relation (Garcia-Reyes, 2011). EUI was then calculated as a summation of a temporal differential of NO_3 estimated while the daily averaged alongshore wind speed (AWS) was greater than 5 ms^{-1} . EUI was set back to zero when daily AWS was less than 5 ms^{-1} for over 3 days.

2.6 Statistic analysis

2.6.1 For the eddy covariance data set

A t-test was used to test the significance of differences between the 2007 and 2008 CO_2 fluxes and environmental parameters (i.e. SST, salinity, PPFD, wind speeds, AWS, friction velocity, chlorophyll density).

2.6.2 For the $p\text{CO}_2$ data set

Linear correlation analysis was used to investigate relationships between environmental parameters (i.e. SST, salinity, PPFD, wind speeds and AWS) and $p\text{CO}_2$ and CO_2 fluxes calculated from $p\text{CO}_2$. Since diurnal variations in $p\text{CO}_2$ were minimal (less than 10 % of the total variations), these data were averaged for 24-h periods.

2.7 Estimates of dissolved inorganic carbon (DIC) and cumulative monthly CO_2 flux and net primary production (NPP)

To evaluate the monthly net CO_2 exchange between the air-water interface and the monthly net primary production (NPP) and to compare them with the dissolved inorganic carbon (DIC) pool, monthly CO_2 fluxes and NPP were estimated and compared with estimated DIC per unit area within the surface mixed layer.

Since daily averaged $p\text{CO}_2$ was most correlated with SST and salinity, as will be demonstrated later, daily $p\text{CO}_2$ from 1988 to 2011 was estimated based on the SST

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and salinity data collected from the BML buoy with the following equation:

$$C_{pw} = -142S_T + 115S_s - 1473 \quad (9)$$

where, S_T is STT and S_s is salinity. CO_2 fluxes were calculated in the same manner as explained above. DIC (C_{dic}) was computed based on the estimated pCO_2 , SST, salinity and pH (pp 324–325 in Sarmiento and Gruber, 2006):

$$C_{dic} = C_{pw}K_0[1 + K_1[H^+]^{-1} + K_1K_2[H^+]^{-2}] \quad (10)$$

where, K_0 is the solubility of CO_2 ($mol\ kg^{-1}\ atm^{-1}$), K_1 and K_2 are dissociation constants of inorganic carbon species ($mol\ kg^{-1}$) determined from temperature and salinity, and $[H^+]$ is hydrogen ion concentration ($mol\ kg^{-1}$) and equal to 10^{-pH} . Hauri et al. (2012) reported seasonal cycles of pH off the Central California coast, and based on these results we approximated the pH seasonal cycle with a sine curve. The influence of the seasonal variation of pH on the DIC calculation was minimal (less than 3%). Similarly, the NPP off Central California reported in Kahru and Mitchell (2002) was approximated by a sine curve. The surface mixed-layer depth was approximated by wind stress estimated from wind speeds (Lentz, 1992).

3 Results and discussions

High CO_2 efflux to the atmosphere was observed during the measurement period in 2007. The average CO_2 flux during this period was $1.5 (\pm 7\ SD)\ g\ C\ m^{-2}\ day^{-1}$ with high variability (Fig. 3a, Table 2). Although a relatively strong sink of CO_2 was observed with high wind speeds on 7–12 September 2008, the overall CO_2 flux measured in 2008 was a slight source of CO_2 , with a magnitude of $0.2 (\pm 3\ SD)\ g\ C\ m^{-2}\ day^{-1}$ (Fig. 4a, Table 2), and the average CO_2 flux during the 2008 measurement season was not statistically different from zero (Table 2).

Contrasting differences were found in SST, salinity, and chlorophyll density between the 2007 and 2008 measurement seasons (Table 2). No difference in AWS between the 2007 and 2008 measurement periods was apparent. However, AWS on longer time scales strengthened in late spring to early summer corresponding with a drop in sea surface temperatures (Fig. 5a, b). The AWS then weakened in fall leading to an increase in sea surface temperature (Garcia-Reyes, 2011). A high EUI ($\sim 20 \mu\text{M}$) was still observed during fall 2008, but a negative AWS ($\sim -5 \text{ms}^{-1}$) was frequently observed, creating conditions favorable to relaxation (Fig. 5a, c). The relatively lower SSTs ranging from 10 to 12 °C in May–June and higher SSTs ranging from 15 to 18 °C in August–September shown in Fig. 5b are compatible with the typical seasonal variations in SST, where upwelling intensifies in early summer and relaxes in fall (Garcia-Reyes, 2011). The upwelling season is also characterized by high temporal variations in CO_2 flux, which was also observed in the 2007 measurements, due to the highly dynamic shifts of water chemistry caused by upwelled water with high CO_2 and subsequent CO_2 uptake by primary production (Dugdale et al., 2006; Evans et al., 2011; Gago et al., 2003).

Relaxation periods are favorable to stratification of the upper water column, which was reflected in lower salinity (~ 32 psu) and higher SST (~ 17 °C) during the 2008 measurement period. Stratification further enhances nutrient depletion on the surface and increases primary production, resulting in higher chlorophyll density. Interestingly, an increasing trend of chlorophyll density was observed during the 2008 measurement period (Fig. 4d). A sink of CO_2 , $\sim 2 \text{gCm}^{-2} \text{day}^{-1}$, was consistently observed on 15 September 2008 when chlorophyll density approached the maximum of 2.3mgm^{-3} . The measurement period in 2008 fell directly between high upwelling events with the peaks of EUI in mid-July and October 2008 (Fig. 5c). Therefore, high nutrient levels were brought to the surface by the preceding upwelling event, increasing the chlorophyll density until the nutrients were depleted. Kahru and Mitchell (2002) reported high NPP of $2.4 \text{gCm}^{-2} \text{day}^{-1}$ (ca. $5 \times 10^{12} \text{gCmonth}^{-1}$ for the area of $6.2 \times 10^4 \text{km}^2$) in summer when nutrient availability was high near our study site. However, the overall CO_2

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flux was near zero or a slight source during the 2008 measurement period despite the favorable conditions for CO₂ uptake. This was because the period preceded a high upwelling period that stimulated higher chlorophyll density and probable relaxation conditions, as inferred from the negative AWS.

5 The overall CO₂ flux estimated from $\Delta p\text{CO}_2$ was also a source of 0.4 (± 0.6 SD) gC m⁻² day⁻¹ from November 2010 to July 2011 (Fig. 6f). The $p\text{CO}_2$ data were often out of the calibration range (150–700 μatm), and therefore the data may not have been accurate at higher $p\text{CO}_2$ values (Fig. 6e). However, $p\text{CO}_2$ higher than 1000 μatm is likely in our study area (personal communication with John Largier of University of California, Davis, 2007). Correlation analysis showed that $p\text{CO}_2$ was most correlated with SST ($R = -0.54$, $p < 0.001$) followed by salinity ($R = 0.50$, $p < 0.001$), and CO₂ flux was strongly correlated to wind speeds ($R = 0.58$, $p < 0.001$) and AWS ($R = 0.51$, $p < 0.001$) followed by SST ($R = -0.40$, $p < 0.001$) and salinity ($R = 0.34$, $p < 0.001$). Similar to the flux data obtained by the eddy covariance technique, higher salinity and lower SST corresponded with higher $p\text{CO}_2$ and a source of CO₂ (Fig. 7). Salinity and SST buoy data from 1988–2011 showed that salinity reached its maximum level and SST reached its minimum level concurrently, as AWS peaked during the upwelling season in late spring and early summer (Fig. 8). Therefore, although AWS did not reflect the CO₂ flux on short time scales (~ 1 h), it likely reflected the CO₂ fluxes on a seasonal scale. Interestingly, the seasonal change of the atmospheric CO₂ showed a very similar pattern to AWS, (Fig. 8a) with a higher peak during a high upwelling season in late spring to early summer; however, because the atmospheric changes in CO₂ concentration were so much smaller than those induced in the surface ocean this periodicity did not influence the overall flux significantly.

25 The CO₂ flux estimated from the eddy covariance technique and from the bulk method both showed that positive CO₂ fluxes were larger than negative fluxes, and that this balance was tightly reflected in salinity and SST. Considering the fact that 99% of the historical data set of salinity and SST fell within the range of our observations, supporting the notion that the data is representative of the region in general,

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our data indicates that the overall exchange of CO₂ in this area is likely a net source of CO₂ to the atmosphere. Table 3 lists our CO₂ flux data together with the CO₂ flux in upwelling zones reported in past studies. Although the annual net CO₂ flux in some upwelling zones (e.g. North Pacific and Galician coast) show a sink of CO₂ and high primary production (Evans et al., 2011), particularly in strong downwelling conditions (Ianson et al., 2009), other studies conducted in North-Central California (Feely et al., 2008; Friederich et al., 2002; Wilkerson et al., 2006) further support our conclusion that the upwelling zone off of Bodega Bay is an overall source of CO₂ to the atmosphere. Despite the high chlorophyll density of 11 mgm⁻³ observed in the early summer of 2000 by Wilkerson et al. (2006), their pCO₂ measurements still indicated that the area was a source of CO₂ to the atmosphere (Table 3). Their study also showed that the magnitude of high pCO₂ was similar over the cross-shelf distance of at least 50 km offshore (Wilkerson et al., 2006).

The estimation of CO₂ flux and DIC based on the relationship of pCO₂ with SST and salinity revealed that the monthly CO₂ flux reached its maximum in May ($\sim 7 \text{ mol C m}^{-2} \text{ month}^{-1}$), which is equal to about 30 % of DIC within the surface mixed-layer (hDIC) (Fig. 9). The annual CO₂ flux was estimated to be about $35 \text{ mol C m}^{-2} \text{ yr}^{-1}$. The fraction of monthly NPP to hDIC became highest ($\sim 50\%$) in late summer, and the annual NPP was estimated to be about $40 \text{ mol C m}^{-2} \text{ yr}^{-1}$. This simple estimation suggested that CO₂ flux and NPP were about the same magnitude and together accounted for significant flux compared to the stock of DIC.

4 Summary

The CO₂ flux from the coastal waters of Bodega Bay was measured during a typical upwelling season in 2007 and during a relaxation season in 2008. CO₂ flux was also estimated from pCO₂ measured from November 2010 to July 2011. Contrasting patterns in CO₂ flux and environmental parameters were observed between the two measurement periods in 2007 and 2008. Measurements in 2007 were during a typical

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upwelling period, as indicated by the seasonal variations of AWS and EUI in early summer, and a source of CO₂ to the atmosphere was observed concurrently with low SST, high salinity, and low chlorophyll density. The measurements in 2008 were, on the contrary, made during a relaxation period with high SST and low salinity that likely led to water stratification and high chlorophyll density. A strong upwelling event preceding the measurements in 2008 likely created favourable conditions for CO₂ uptake. However, the average CO₂ flux over the 2008 measurements was a small source of CO₂ overall. The magnitude of the observed sources CO₂ to the atmosphere estimated by the bulk method were also much higher than that of sinks of CO₂ during the pCO₂ measurements. Similar to the fluxes observed by the eddy covariance technique, those estimated by the pCO₂ measurements were closely related to salinity and SST. The historical data set of salinity and SST (1988–2011) indicated that 99% of data fell in the range of our observations. Therefore, the area was likely a source of CO₂ to the atmosphere annually. Past studies further support the assertion that the overall conditions are favourable to producing a source of CO₂ in the coastal upwelling area off of Bodega Bay at least within 50 km of the coast. Although AWS did not reflect pCO₂ or CO₂ flux on shorter time scales, high AWS together with low SST and high salinity suggests that AWS likely drives the seasonal cycle of CO₂ flux. Based on the pCO₂ to SST and salinity relationship, the average annual CO₂ flux between 1988 and 2011 was estimated to be 35 molCm⁻²yr⁻¹, and the amount of CO₂ transferred vertically and consumed by NPP accounted for significant amounts of carbon in comparison to the local dissolved carbon stock.

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Table 1. Data summary.

Parameter	Date range	Location	Position
Eddy covariance tower			
CO ₂ flux			
Friction velocity	28 May–5 Jun 2007,	Bodega Bay, CA	38°20' N, 123°04' W
Wind speed	31 Aug–25 Sep 2008		
Wind direction			
BML meteorological tower			
Air temperature			
Wind speed	1988–2011	Bodega Bay, CA	38°19' N, 123°04' W
Wind direction			
PPFD			
BML buoy			
Sea surface temperature	1988–2011	Near Bodega Bay, CA	38°18' N, 123°04' W
Salinity			
Chlorophyll density	2007–2008		
$p\text{CO}_2$	Nov 2010–Jul 2011		
NDBC 40613 buoy			
Wind speed			
Wind direction		Near Bodega Bay, CA	38°14' N, 123°18' W
AWS (calculated from WS and WD)	2007–2011		
Sea surface temperature			
Aircraft CO ₂ measurement			
Atmospheric CO ₂	Sep 2003–Jan 2012	Near Trinidad Head, CA	41°05', 124°17' W (~ 4 km in altitude)

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Table 2. Mean, standard deviation, maximum, and minimum of CO₂ flux (F_c) in gCm⁻²day⁻¹, wind speed (U) in ms⁻¹, friction velocity (U^*) in ms⁻¹, air temperature (T_a) in °C, PPFD in μmolm⁻²s⁻¹, chlorophyll density (CHL) in mgm⁻³, salinity (SS) in psu, sea surface temperature (SST) in °C, sea surface temperature measured at the NDBC 46013 buoy (SST_{N13}) in °C and alongshore wind speeds (AWS) in ms⁻¹ from (a) 28 May–4 June 2007 and (b) 30 August–24 September 2008.

	F_c	U	U^*	T_a	PPFD	CHL	SS	SST	SST _{N13}	AWS
(a) 28 May–4 Jun 2007										
Mean	1.5	4.5	0.33	13.6	609	0.4	34	11.5	10.3	2.6
Standard deviation	7.4	2.7	0.3	3.2	522	0.0	0.5	0.4	0.6	2.0
Maximum	28.3	11.7	2.52	21.9	2011	0.5	34	12.2	12.4	9.3
Minimum	-29.1	0.6	0.04	8.3	0	0.0	28	9.6	9.1	0.1
(b) 30 Aug–24 Sep 2008										
Mean	0.2*	2.8	0.19	12.5	523	1.8	32	16.8	14.0	3.9
Standard deviation	2.5	1.4	0.1	1.2	484	0.7	0.2	0.3	1.0	4.7
Maximum	16.5	8.1	1.82	16.4	1645	3.5	33	17.7	17.5	17.0
Minimum	-21.0	0.2	0.02	10.7	0	0.7	32	15.9	11.7	-5.8
	**	**	**	**		**	**	**	**	

* Statistically significant difference between the periods (a) and (b) in each parameter ($p < 0.01$).

** not statistically different from zero at $p = 0.01$.

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Table 3. CO₂ flux, pCO₂, wind speed, sea surface temperature (SST), salinity (SS) and chlorophyll density (CHL) observed in coastal upwelling zones.

Location	Time	CO ₂ flux gCm ⁻² d ⁻¹	pCO ₂ µatm	U ms ⁻¹	SST °C	SS psu	CHL mgm ⁻³	Technique	Study
Oregon coastal ocean	May–Aug 2001	-0.24	-130 ^b	6	10–11	30–31	–	Bulk method	(1) Hales et al. (2005)
Central California coast	Spring, 1999	1.64	~ +480 ^b	–	–	–	–	Bulk method	(2) Friederich et al. (2002)
Central California coast	1998–1999	0.072	+50 ^b	5.8	–	–	–	Bulk method	(2) Friederich et al. (2002)
Cabo Nazca, Peru	Spring, 1976	-0.24–0.21 ^a	120–980	–	20–23	–	0–33	Bulk method	(3) Simpson and Zirino (1980),
Vancouver Island, Canada	typical year	-0.35–0.25	220–560	–	–	–	–	Box model	(4) Ianson and Allen (2002)
Vancouver Island, Canada	typical year	-0.014	–	–	–	–	–	Box model	(5) Ianson et al. (2009)
Galician coast, Spain	1997–1999	0.04–0.084	315–345	4–11	14–20	35–36	–	Bulk method	(6) Borges and Frankignoulle (2002)
St. Lawrence Island	Summer, 1968	0.07 ^a	+108 ^b	–	4.5–9	–	–	Bulk method	(7) Kelly and Hood (1971)
Peru coast	Aug 1986	0.03–0.50 ^a	400–1000	–	14.5–16.5	35	~ 1.7	Bulk method	(8) Copin-Montegut and Raimbault (1993)
Bodega Bay, CA	1–3 Jun 2000	0.07 ^a	470	5 ^c	9	34	11	Bulk method	(9) Wilkerson et al. (2006)
Bodega Bay, CA	19–20 May 2001	0.09 ^a	625	-4 ^c	10	33.9	3.5	Bulk method	(9) Wilkerson et al. (2006)
Bodega Bay, CA	15–17 Jun 2002	1.05 ^a	800	9 ^c	9.5	34	1.5	Bulk method	(9) Wilkerson et al. (2006)
Bodega Bay, CA	May–Jun 2007	1.94	–	4.29	11.5	33.7	0.4	Eddy covariance	this study
Bodega Bay, CA	Sep 2008	0.22	–	3.32	16.7	32.5	1.7	Eddy covariance	this study
Bodega Bay, CA	Nov 2010–Jul 2011	0.40	+402 ^b	5.0	11	33	–	Bulk method	this study

^a CO₂ flux estimated by reported pCO₂ with the bulk method (average of Nightingale et al., 2000 and Wanninkhof 1992).

^b dpCO₂ (atmospheric CO₂–pCO₂).

^c Alongshore wind speeds.

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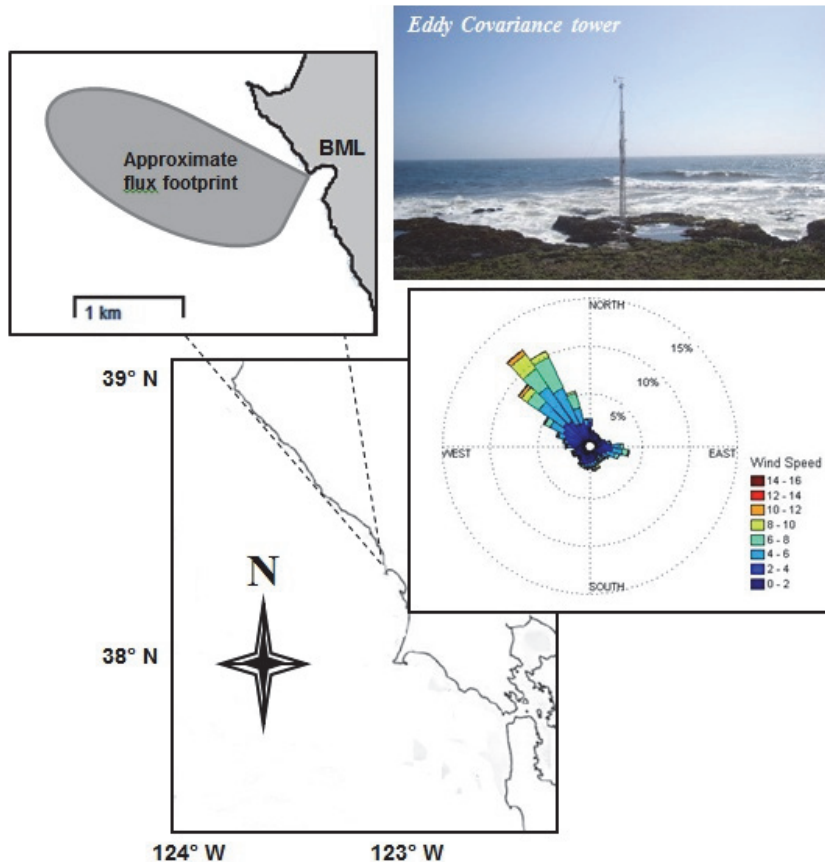


Fig. 1. Study site with the approximate 90% flux footprint, and the wind rose of daily averaged wind directions and speeds (1990–2008) in Bodega Bay, California ($38^{\circ}18'58''$ N, $123^{\circ}04'20''$ W).

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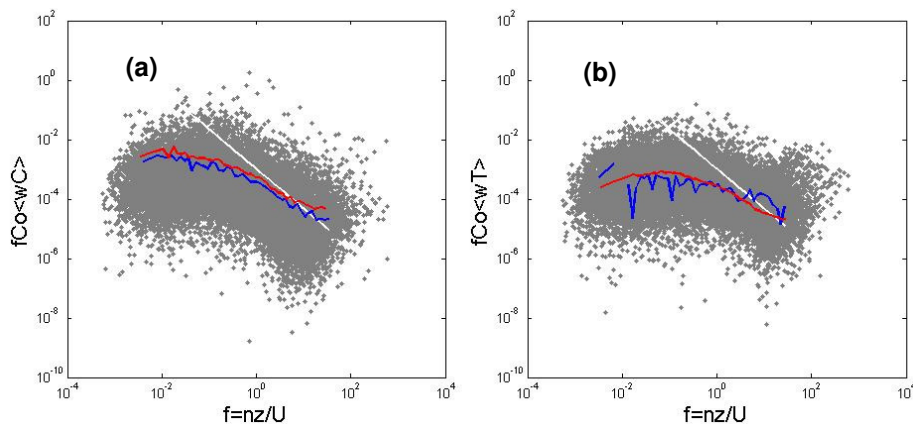


Fig. 2. Normalized cospectra of the vertical wind speed and the CO₂ mixing density, $f_{Co < wC >}$ (a) and the vertical wind speed and temperature, $f_{Co < wT >}$ (b). The blue line indicates the average cospectra for $z/L > -0.1$ and the red line indicates the average for $z/L < -0.1$. The white line shows $-4/3$ at log-log scale.

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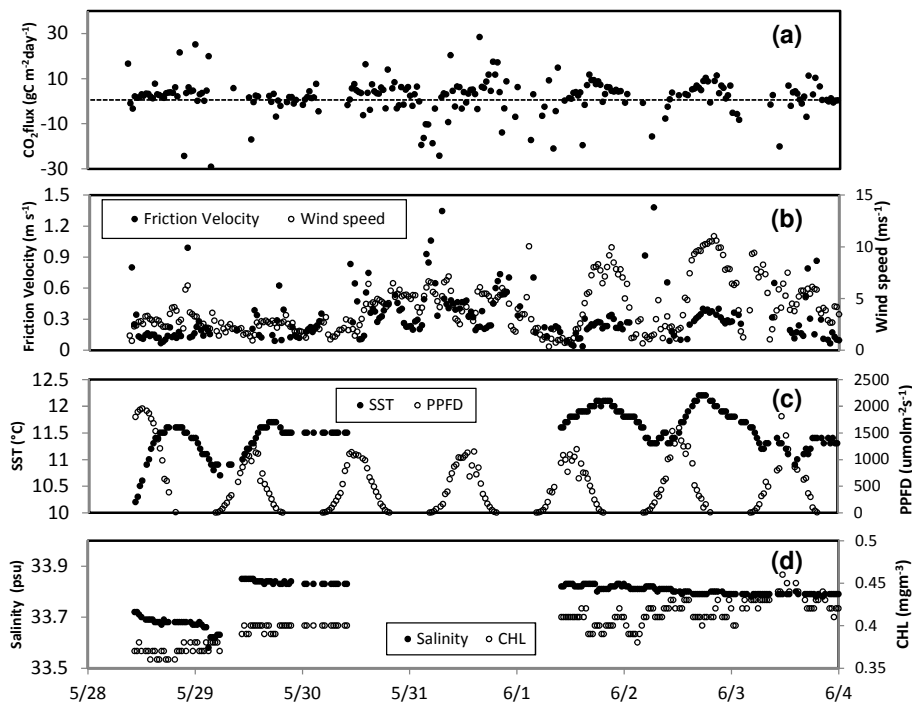


Fig. 3. CO₂ flux (a), friction velocity and wind speed (b), SST and PPFD (c), salinity and chlorophyll density (CHL) (d) measured from 28 May to 4 June 2007. All data were plotted every 30 min.

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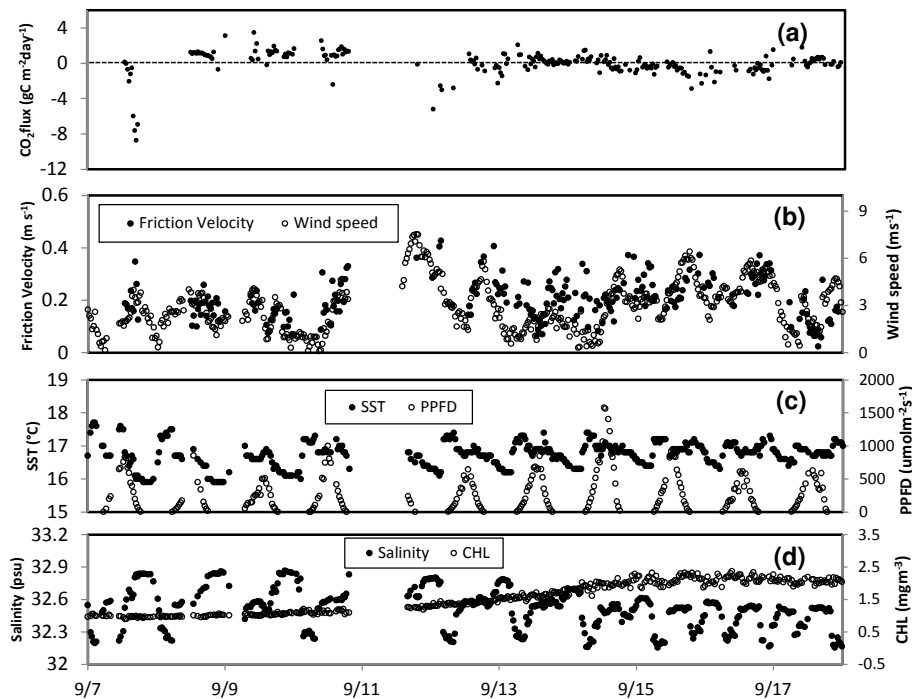


Fig. 4. CO₂ flux (a), friction velocity and wind speed (b) SST and PPFD (c), salinity and chlorophyll density (CHL) (d) measured from 30 August to 24 September 2008. All data were plotted every 30 min.

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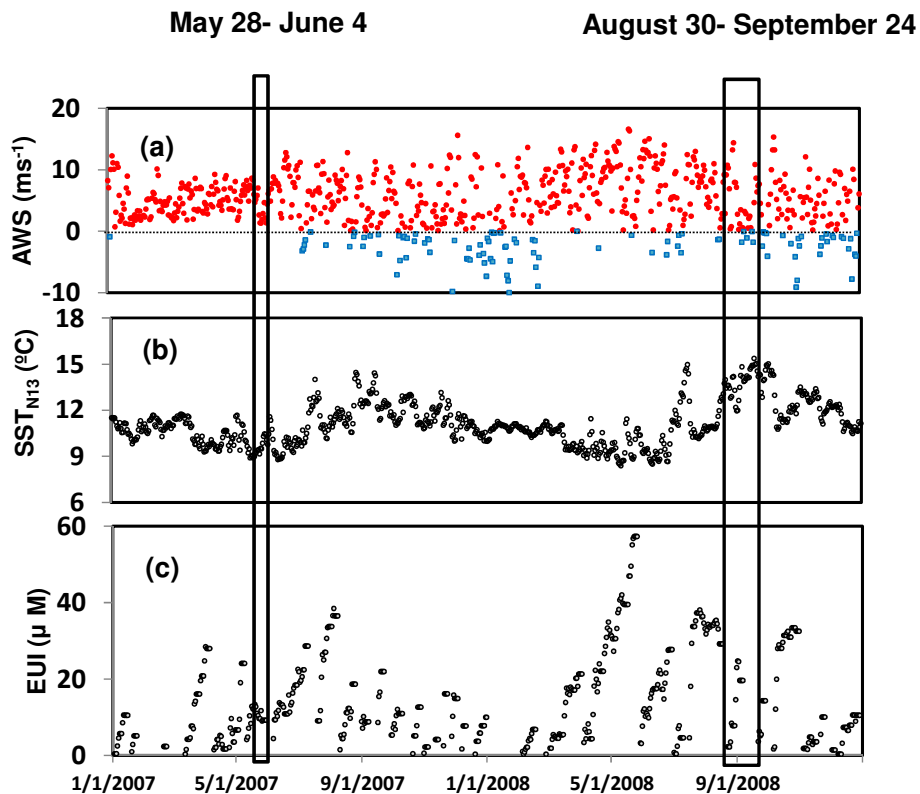


Fig. 5. Alongshore wind speed calculated from wind speed and direction at the NDBC 46013 buoy, with positive AWS (red dots) and negative AWS (blue dots) differentiated by color **(a)**, sea surface temperature at the NDBC 46013 buoy (SST_{N13}) **(b)**, and effective upwelling index (EUI) **(c)**. All data were averaged daily and plotted. The two boxes indicate the periods when eddy covariance measurements were available.

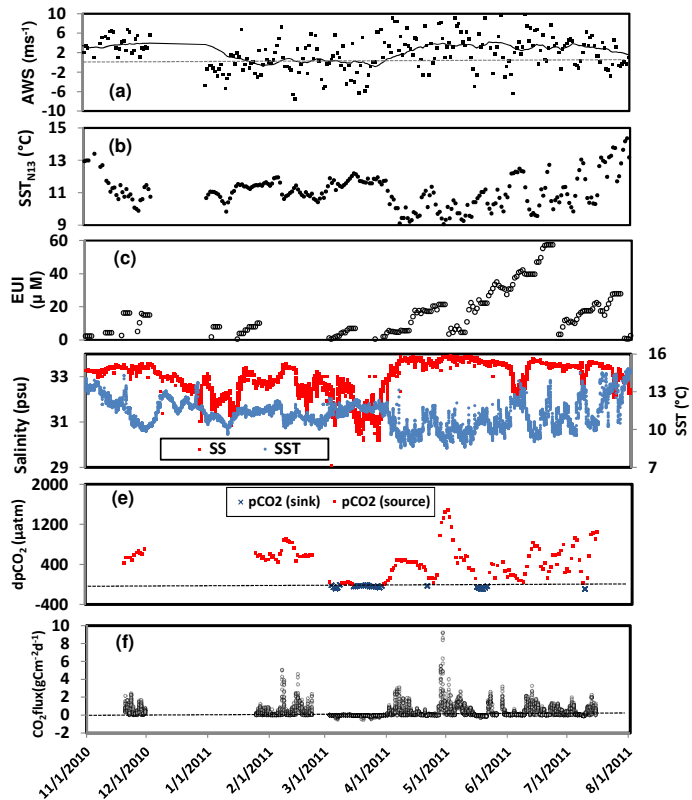


Fig. 6. Hourly alongshore wind speed (AWS) calculated from wind speed and direction at the NDBC 46013 buoy (black squares), with the 30 point running average (black line) also shown (a), sea surface temperature at the NDBC 46013 buoy (SST_{N13}) (b), and effective upwelling index (EUI) (c), salinity and SST (d), $dpCO_2$ (pCO_2 –atmospheric CO_2) (e) and CO_2 flux estimated by the bulk method (f) from November 2010 to July 2011. (a)–(c) were plotted daily and (d)–(f) were plotted hourly.

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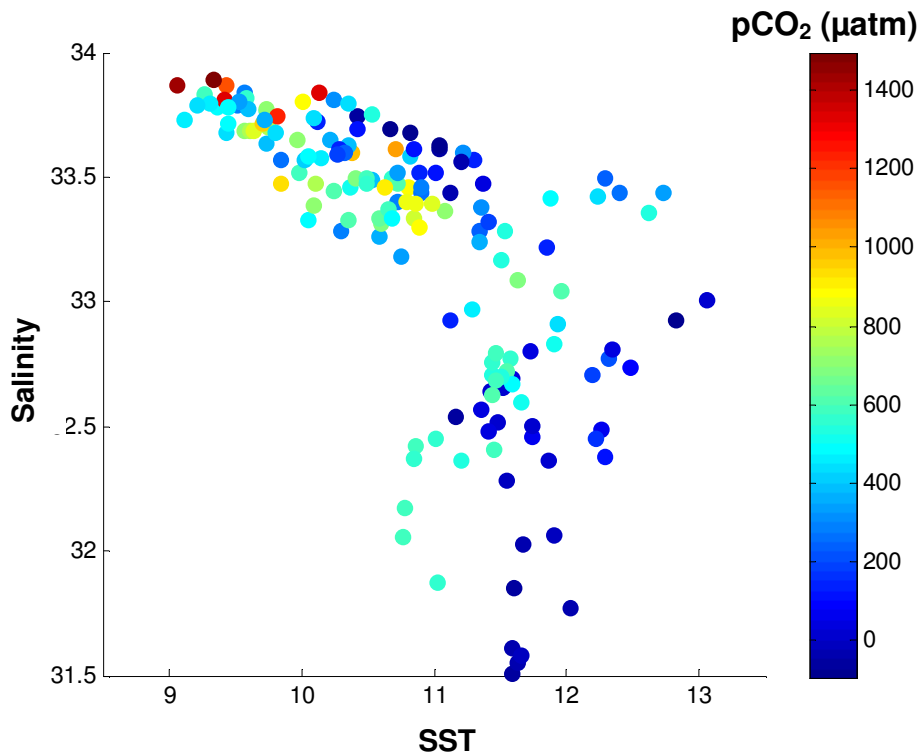


Fig. 7. Distribution of daily averaged $p\text{CO}_2$ in relation to salinity (psu) and seas surface temperature (SST) ($^{\circ}\text{C}$) measured from November 2010 to July 2011. A multiple linear regression was applied for $p\text{CO}_2$ against both salinity and SST ($R = 0.57$, $p < 0.001$).

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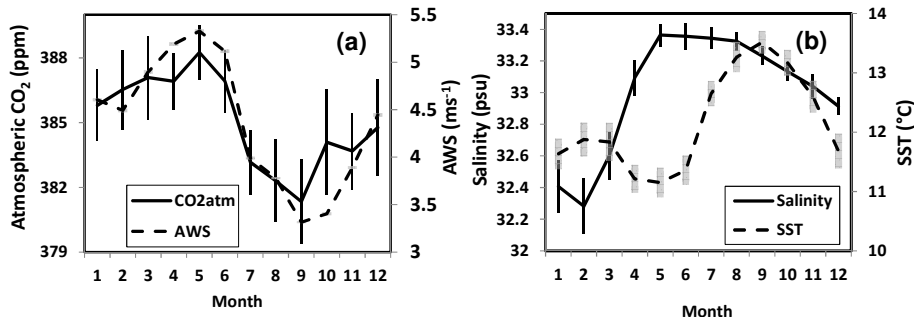


Fig. 8. Average seasonal variations of atmospheric CO_2 concentration between September 2003 to August 2012 obtained near Trinidad Head, CA and alongshore wind speed (AWS) between 1988 to 2011 **(a)** and average seasonal variations of salinity and sea surface temperature (SST) from 1988 to 2011 obtained from the BML buoy **(b)**. Vertical bars show standard errors (SE). The average and SE for each month were calculated from the monthly averages available for all years.

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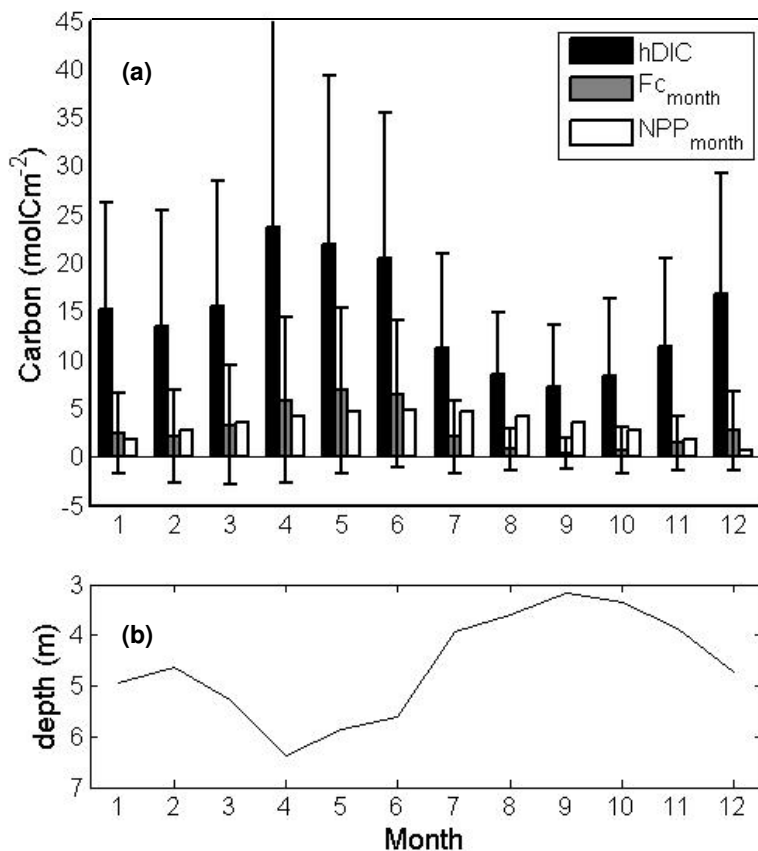


Fig. 9. Monthly averaged seasonal variations of DIC per unit area within the surface mixed layer (hDIC), monthly CO₂ flux ($F_{c,month}$), and monthly NPP (NPP_{month}) **(a)** and the depth of the surface mixed layer depth **(b)** estimated from 1988–2011 data. The vertical lines indicate standard deviations.

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