Large Decadal Changes in Air-Sea CO₂ Fluxes in the Caribbean Sea

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Abstract

Sixteen years of surface water CO₂ data from autonomous systems on cruise ships sailing in the Caribbean Sea and Western North Atlantic show marked changes on interannual timescales. The measured changes in fugacity (partial pressure) of CO₂ in surface water, fCO₂, are based on over a million observations. Seasonally the patterns are similar to other oligotrophic subtropical regions with an amplitude of fCO₂ of ≈40 µatm with low wintertime values, causing the area to be a sink, and high summertime values making it a source of CO₂ to the atmosphere. On annual scales there was negligible increase of fCO₂ from 2002 to 2010 and a rapid increase from 2010 to 2018. Correspondingly, the trend of air-sea CO₂ flux from 2002 to 2010 was strongly negative (increasing uptake or sink) at −0.05 ± 0.01 (mol m⁻² year⁻¹) and positive (decreasing uptake) at 0.02 ± 0.02 (mol m⁻² year⁻¹) from 2010-2018. For the whole period from 2002 to 2018, the fCO₂ lagged the atmospheric CO₂ increase by 24 %, causing an increase in CO₂ uptake. The average flux into the ocean for the 16 years is −0.20 ± 0.16 mol m⁻² year⁻¹ with the uncertainty reflecting the standard deviation in annual means. The change in multiannual trend in fCO₂ is modulated by several factors, notably changes in sea surface temperature and ocean mixed layer depth that, in turn, affected the physical and biological processes controlling fCO₂.

Plain Language Summary

Through a unique collaboration with Royal Caribbean Cruise Lines several cruise ships were outfitted with automated surface water carbon dioxide (CO₂) measurement systems, providing weekly observations in the Caribbean Sea over the past 16 years. From over a million measurements, the increase in surface water CO₂ in response to rising atmospheric levels was accurately monitored. The region is, on average, a carbon dioxide sink with large multiyear differences. For the first 8 years, the surface water levels do not change appreciably causing an increase in difference between the air and sea concentrations. This increased differential drives in increase in the air to sea flux of CO₂. Carbon dioxide levels in surface water in the following 8 years actually increased faster than the atmosphere, thereby decreasing the differential and subsequent flux rate. The cause of changes in trends appears associated with changes in the ocean biogeochemistry, linked to sea surface temperature and mixed layer depth, particularly in the middle part of the record.

1. Introduction

There has been a large increase in measurements of fugacity of carbon dioxide in surface water (fCO₂) over the past two decades after realization that the quality of automated measurements was high enough to quantify global ocean CO₂ uptake (Takahashi et al., 2002). Recommendations to provide sustained systematic fCO₂ measurements in the world’s oceans to estimate global air-sea fluxes were implemented (Bender et al., 2002). The Caribbean Sea was targeted for observations with passenger cruise ships, providing a unique platform for the unattended systems (Figure 1). Through a federal, industry, academic partnership between NOAA, Royal Caribbean Cruise Lines (RCCL) and the University of Miami, the cruise ship Explorer of the Seas (EoS) was outfitted with an automated surface water CO₂ measurement system (Pierrot et al., 2009) providing observations on alternating weekly transects in the Eastern and Western Caribbean. The biweekly occupations of the same locations were optimal to determine regional changes in surface water CO₂ levels in
support of ocean acidification and ocean carbon cycle research in an area with significant coral reefs that are vulnerable to changes in temperature and CO$_2$ levels (Kleypas, 1999).

The cruise ship changed homeport from Miami FL to Port Elizabeth NJ and the routes changed in 2008, foregoing the Western Caribbean occupations and adding cruises to Bermuda. In 2012, the EoS was repositioned to the Pacific and other RCCL ships were outfitted with underway CO$_2$ systems that covered similar transects as the EoS but on more irregular and seasonal basis. From 2015 onward, two cruise ships, the Celebrity Equinox and Allure of the Seas, have covered the area.

From 2002 to 2018 a total of 510 voyages with automated fCO$_{2w}$ measurements were completed by the RCCL cruise ships. They cover a region spanning 16–28°N and 88–62°W (Figure 1). This represents the northern Caribbean Sea and the western Atlantic, north of the Caribbean island chain. Over the 16 years, there are at least biweekly occupations at the beginning of the record, from 2002 to 2007, and towards the end, from 2014 to 2018, with gaps in the years in between. The temporal and spatial gaps were filled by regressions with position, remotely sensed sea surface temperature (SST), modeled sea surface salinity (SSS), and modeled mixed layer depth (MLD). This does not unduly impact the quantification of conditions and changes in fCO$_{2w}$ and air-sea CO$_2$ fluxes over the time period.

The Caribbean Sea is largely oligotrophic with low nutrient concentrations in surface water. However, several local environmental factors can impact its nutrient and carbon dynamics. Episodic riverine influence from the Orinoco River increase biological productivity in the southwestern part of the region, based on observed lower salinity and remotely sensed color (Lopez et al., 2013). Runoff and other coastal influences from the islands have an impact on the local CO$_2$ dynamics and flux estimates (Melendez Oyola et al., 2018). Hurricanes leave an imprint on fCO$_{2w}$ through large effluxes during the extreme wind events and subsequent decreases in fCO$_{2w}$ caused by lower SST and enhancement of biological productivity due to enhanced nutrient exchange from below the mixed layer (Huang & Imberger, 2010; Wanninkhof et al., 2007). The riverine, island, and hurricane influences are significant at local and short time scales but do not show a large impact on seasonality or the regional scale patterns and multidecadal trends of air-sea CO$_2$ fluxes for the whole region.

The canonical view that the surface inorganic carbon dynamics in the region are largely driven by temperature and processes impacted by temperature holds largely true. However, like the ocean time series stations
in the oligotrophic gyres, near Hawaii (HOT) and Bermuda (BATS), there are multiyear trends that cause the surface water CO\textsubscript{2} increases to deviate from the expected rate of increase (Bates et al., 2014; Dore et al., 2009). The divergent multiyear trends observed in the Caribbean are significantly greater than observed at BATS and HOT for the time period in the references listed. This is unexpected because the shallow MLD, relatively low productivity, and strong stratification should cause the fCO\textsubscript{2w} increases to be largely controlled by atmospheric CO\textsubscript{2} increases, leading to fCO\textsubscript{2w} increases of about 2 \muatm/year.

The multiyear changes impact the rate of ocean acidification (OA), which is of concern in the region where coral reef cover has declined by over 50% in the last 5 decades due to multiple human stressors (Jackson et al., 2014). The current observation-based estimates of OA for the region rely on empirical correlations between inorganic carbon chemistry versus SST and SSS from the earlier years of this observation program from which an ocean acidification product suite was developed (Gledhill et al., 2008). In this model, annual increases in atmospheric fCO\textsubscript{2} are directly coupled to surface ocean acidification rates with subannual variability governed primarily by SST. The results of the current effort indicate that this approach can be fine-tuned with the 16-year record of observations showing the multiannual anomalies in trends in fCO\textsubscript{2w}.

Seasonal changes in fCO\textsubscript{2w} in the Caribbean were first detailed by Olsen et al. (2004) based on the first year (2002) of cruise data from the EoS. The fCO\textsubscript{2w} levels were closely related to SST and, thus, had lower values in winter and higher in the summer. This annual cycle of fCO\textsubscript{2w} is such that in the wintertime the region is a CO\textsubscript{2} sink and during the summer is a CO\textsubscript{2} source. Olsen et al. (2004) created a simple but robust algorithm relating fCO\textsubscript{2w} to SST and location based on the 2002 data:

\[
\text{fCO}_{2w} = 10.18 \times \text{SST} + 0.5249 \times \text{Lat} - 0.2921 \times \text{Lon} + 52.19, \quad r^2 = 0.87
\]

where SST is in °C; Lat is latitude in fractional degrees (North); and Lon is longitude as negative degrees (East). The root-mean-square (RMS) uncertainty of calculated fCO\textsubscript{2w} was 5.7 \muatm.

Equation (1) reflects the strong relationship of fCO\textsubscript{2w} with SST. It also indicates that at constant SST, fCO\textsubscript{2w} will be on average 5 \muatm higher in the southern part (15°N) than in the northern part (25°N) and 8 \muatm higher in the western part (88°W = −88°E) compared to the eastern boundary of the region (62°W). These spatial gradients are relatively small compared to the ≈40 \muatm seasonal amplitude associated with a 4 °C change in SST from winter to summer in the region.

Park and Wanninkhof (2012) analyzed 8 years of fCO\textsubscript{2w} data from 2002 to 2009, in the northeastern part of the region, north of Puerto Rico, predominantly from the EoS. During this time period the fCO\textsubscript{2w} increased ≈9 \muatm, compared to expectations that it should have changed by ≈18 \muatm in response to atmospheric CO\textsubscript{2} increases. The stagnation was attributed primarily to wintertime fCO\textsubscript{2w} increasing very slowly over this time period. This was caused by anomalously high wintertime SST in 2002–2003 and lower wintertime SST in 2008–2009. Winter MLD also increased toward the end of the time period. As a result, the air-water fugacity of CO\textsubscript{2} difference, ΔfCO\textsubscript{2}, became more negative in wintertime causing an increase of the CO\textsubscript{2} sink strength in the region for the 2000–2009 period.

Here we build on the previous work, and extend the time series and analyses from 2002 to the spring of 2018. Data products such as wind speed needed to determine the fluxes along with remotely sensed SST, modeled SSS, Normalized Fluorescence Line Height (NFLH), and modeled MLD are used to interpret to results. The sources of these data are detailed in the data section. Instrumental procedures, time series analyses, mapping procedures, and flux calculations are described in the methods section. The seasonal cycles and multiyear trends of the variables of interest are described, and air-sea CO\textsubscript{2} fluxes over the region are quantified. The marked changes from a stagnant annual average fCO\textsubscript{2w} over the first part of the record from 2002 to 2010 to a larger than atmospheric rate of increase in fCO\textsubscript{2w} from 2010 to 2018 are quantified. The causes thereof are discussed and deconvolved into temperature, gas exchange, and a residual attributed to biological and mixing (B&M) components.

2. Data and Data Products

In this paper, three different datasets are used and compared. The following nomenclature is used: observational datasets from the cruise ships using the time and position of measurements are referred to as
observations; these data are binned and averaged on a 1° × 1° space scale and monthly timescale (1° × 1° × mo) and are called gridded data (product). The gridded data are useful to determine biases between observed SST and SSS, and remotely sensed SST and modeled SSS. They are also used to determine if gridding and extrapolation procedures impact the interpretation. For the whole region spanning 16°N–28°N and 88°W–62°W, the fCO$_{2w}$ data are extrapolated using multilinear regressions (MLRs) with position, SST, SSS, and MLD as described below. They are used in conjunction with monthly remotely sensed SST, wind, and NFLH, and modeled SSS on the same grid and are referred to as the (gridded) mapped product. This product is the basis of the trend estimates and the explanation of trends. Using the gridded mapped product, the trends in fCO$_{2w}$ and flux for the whole time period and area can be determined without artifacts caused by data gaps.

The field observations were obtained from the three cruise ships listed above totaling over 1 million data points over the 16-year period from March 2002 to 2018. They are posted on the website: www.aoml.noaa.gov/occi/ocdweb/occ.html per cruise and ship, along with pertinent metadata. The data are archived in the NCEI Ocean Carbon Data System (www.nodc.noaa.gov/ocads/). Data are also collated annually in the Surface Ocean Carbon ATlas, SOCAT (Bakker et al., 2016) and LDEO (Takahashi et al., 2017) data products. Here we use the original data as served from the AOML website. The fCO$_{2}$ data are the same for all sites but the quality control procedures and flags differ slightly. Only data with quality control flag 2 (= good data) are used, which are over 98 % of the observations.

For the determination of the fugacity of CO$_2$ in air, fCO$_{2a}$, the average monthly atmospheric CO$_2$ mole fraction (XCO$_{2a}$) product based on weekly measurements at the stations at Key Biscayne, FL, USA (KEY; 26°N, 80°W), and Ragged Point, Barbados (RPB; 13°N, 59°W) are used. They are obtained from the flask sampling network of the Global Monitoring Division of the Earth System Research Laboratory (GMD/ERSL/NOAA; Dlugokencky et al., 2017). These values are quality controlled at GMD and fit to the annual seasonal cycle (see www.esrl.noaa.gov/gmd/ccgg/mlb/data.php). The monthly product is considered representative for the whole region. The difference in monthly mean between the KEY and RPB products from 2002–2018 is 0.6 ± 0.6 ppm (n = 189) with KEY values being on average higher.

Support data used for analyses are on the same 1° × 1° × mo grid as the mapped fCO$_{2w}$. Wind speeds are from the updated cross-calibrated multiplatform wind product 2 (Atlas et al., 2011). The 1° × 1° × mo mean neutral wind at 10-m height $<u_{10}>$, and its second moment $<u_{10}^2>$ is determined from the ¼ degree, 6-hourly product from Remote Sensing Systems (www.remss.com). The SST product is the Optimum Interpolated SST, OISST (Reynolds et al., 2007). It uses data from ships, buoys, and satellites to generate the analyzed fields. For the OISST, the reference SST is from buoys, and the SST from ships in this product are adjusted to the buoy data by subtracting 0.14 °C. The OISST is on average 0.25 ± 0.40 °C (n = 8,191) lower than SST measurements from the cruise ships when comparing the 1°-gridded data product for the particular month with OISST product. The SST probes on the cruise ships have accuracies better than 0.01 °C, and while the offset is within the standard deviation, it is assumed the difference is real and attributed to the near-surface cool skin (Soloviev & Schlüssel, 1996).

No MLD determinations were made on the cruises and limited observational estimates from other sources are available. The profiling float program, Argo, produces a global MLD product in near real time (Holte et al., 2017). However, Argo only has limited deployments in the Caribbean and no MLD product is available for the region. The MLDs used here are from a numerical model (HYCOM; https://hycom.org/regional). The MLDs are based on a density contrast of 0.03 between surface and subsurface (de Boyer Montégut et al., 2004).

The SSS data used were also extracted from this model, SSS$_{hycom}$. The SSS$_{hycom}$ is on average 0.1 ± 0.28 (n = 8,191) greater than the ship-based 1° × 1° × mo data product. The difference does not impact the results or interpretation as the SSS has a weak influence on the fCO$_{2w}$ calculated from the MLRs.

NFLH, which is used as a proxy for biological productivity, was retrieved from the moderate resolution imaging spectrophotometer, MODIS. MODIS flies on the satellites AQUA and TERRA. NFLH is a relative measure of water-leaving radiance associated with chlorophyll fluorescence. The level 3 data used are at 1° × 1° × mo resolution and are obtained from http://oceancolor.gsfc.nasa.gov/cgi/l3 website.
3. Methods

3.1. Instrumentation

The fCO$_{2w}$ levels are measured by infrared (IR) analysis of headspace gas equilibrated with water in an equilibration chamber. The gas in the headspace is partially dried ($\approx$75%) before measurements. The procedures and instrumentation are based on a community design described in Pierrot et al. (2009). The instruments are manufactured by General Oceanics INC, and have performed to high accuracy specifications. The instruments have shown agreement to within 1 μatm with other state-of-art systems in intercomparison studies (Y. Nojiri et al., personal communication, February 2009). The IR sensor (LI-COR 6262) is calibrated every 4.5 hr against four standard gases supplied by the GMD/ESRL/NOAA, traceable to the WMO CO$_2$ mole fraction scale. The CO$_2$ concentrations in the standards span the range of values encountered along the ship tracks. Several different versions of the instrument have been deployed on the ships over the years but overall measurement principles and accuracies, estimated at better than 2 μatm, have remained the same.

Pressure in the headspace and temperature of the water in the equilibrator and near the seawater intake, assumed equivalent to SST, are measured at high accuracy. Equilibrator temperature is obtained with a Hart RS thermistor accurate to ±0.002 °C. The pressure is measured by a Setra model 270 barometer which is accurate to 0.1 mbar (hPa). SST and salinity are determined by a Seabird thermosalinograph accurate to 0.01 °C and 0.01 based on factory specifications. As conductivity values used to calculate salinity change due to fouling, the in situ observations probably are less accurate than specification but the uncertainty in salinity has negligible impact on the calculation of fCO$_{2w}$.

The water intake for the underway pCO$_2$ instrument and thermosalinograph is near the bow thruster cavity on the ships at a depth of 1.5 m. The shallow depth of the intake and operation of the bow thruster can cause bubble entrainment near ports and in heavy seas. Data obtained under these conditions are flagged and not used in the analyses. Cruises follow a fixed route with occasional deviations due to weather, such as averting heavy seas during and immediately after hurricanes. Typical cruise speeds are 22 knots that with sampling every 2.5 min, yields a sample approximately every 1.7 km except for 35 min every 4.5 hr when four calibration gases and a CO$_2$-free reference gas are analyzed followed by four atmospheric CO$_2$ measurements. This causes a distance of 24 km ($\approx$1/4°) without fCO$_{2w}$ measurements. The air for the atmospheric measurements is obtained from an intake mounted near the bow of the ship and piped to the infrared analyzer. Air measurements do not cover the entire record. They were started on the EoS in 2008 and on the Equinox in May 2015. Due to challenges installing the airline that needs to be strung up eight decks and forward in the ship’s interior there is no air intake line on the Allure of the Seas. As there are extensive periods when no air measurements are available, the values used here were determined from two locations in the MBL CO$_2$ product as described in the data section.

3.2. Calculation of fCO$_2$

The calibrated values of XCO$_2$ from the IR analyzer are accurate to within 0.2 ppm based on the certified values of the calibration gases that are traceable to the WMO scale, and uncertainties in their interpolation. The XCO$_2$ values are converted to fCO$_{2eq}$ (μatm) values following Pierrot et al. (2009):

$$ fCO_{2eq} = XCO_{2eq} \times (P_{eq} - pH_2O) \times f(T_{eq} P_{eq}) $$  \hspace{1cm} (2)  

where eq refers to equilibrator conditions. $P_{eq}$ is the pressure in equilibrator headspace and the $pH_2O$ is the water vapor pressure calculated according to equation (10) in Weiss and Price (1980). The $T_{eq}$ is the temperature of water in the equilibrator. The function $f(T,P)$ is the fugacity correction (Weiss, 1974). The fugacity is the partial pressure of CO$_2$ corrected for nonideality of the gas. The virial coefficients used in the correction as well as the appropriateness of the correction are under debate. The magnitude of the correction ($f(T,P)$ $\approx$0.997) for converting from pCO$_2$ to fCO$_2$ is about $\approx$1 μatm at the fCO$_2$ ranges encountered, so the uncertainties in the correction have no effect on the conclusions. The fCO$_{2eq}$ is corrected to surface water values using the intake temperature (SST) according to the empirical relationship that Takahashi et al. (1993) developed for North Atlantic surface waters:

$$ fCO_{2w} = fCO_{2eq} \times e^{(0.0423(SST - T_{eq}))} $$  \hspace{1cm} (3)  

on average $SST - T_{eq} = 0.12 \pm 0.28$ °C ($n = 667,865$). The SST is higher than $T_{eq}$ because of the warm SST in
the region and lower air temperatures in the air-conditioned ship, in contrast to most ships where water warms once inside. The fCO$_{2a}$ (μatm) are calculated from XCO$_{2a}$ according to:

$$fCO_{2a} = XCO_{2a} \times (P-pH_{2}O) \times f(SST, P)$$  \hspace{1cm} (4)

here P is the ambient pressure at sea level, pH$_{2}O$ is determined at SST, and f (SST, P) is the fugacity correction.

### 3.3. Calculation of Air-Sea CO$_2$ Flux

For the determination of the air-sea CO$_2$ flux, F$_{CO2}$ (mol m$^{-2}$ year$^{-1}$), a bulk formulation is applied to the data from the gridded mapped product:

$$F_{CO2} = ks \times \Delta fCO_2$$  \hspace{1cm} (5)

where $\Delta fCO_2$ is ($fCO_2$w$−fCO_2$a), s is the seawater CO$_2$ solubility (Weiss & Price, 1980), and k is the gas transfer velocity parameterized as a function of wind speed (Wanninkhof, 2014):

$$k = 0.251 \times <u_{10}^2> \times (Sc/660)^{-1/2}$$  \hspace{1cm} (6)

where $<u_{10}^2>$ is the monthly second moment of the 6-hourly wind speeds reported in cross-calibrated multi-platform wind product 2, accounting for the impact of variability of the wind speed on k. The Sc is the Schmidt number of CO$_2$ in seawater determined as a function of temperature from Wanninkhof (2014).

### 3.4. Gridded Mapped fCO$_{2w}$ Product Using Multilinear Regressions

Monthly fCO$_{2w}$ fields for the region were created in the following fashion. Annual MLRs were determined from the fCO$_{2w}$ data that were binned and averaged on a 1° × 1° × mo grid similar to the approach of Olsen et al. (2004). To take advantage of improved regional products of salinity and mixed layer depth these variables were included in the MLRs. For SST both a linear and logarithmic dependence were checked but little difference was observed in the fit, and a linear SST dependence was used. The functional form for the MLR fit is as follows:

$$fCO_{2w,mapped} = a \text{Lon} + b \text{Lat} + c \text{SST} + d \text{MLD} + e \text{SSS} + f$$  \hspace{1cm} (7)

MLRs were determined for each year, so no assumptions about annual fCO$_{2w}$ trends had to be made. The coefficients and their standard errors for each annual MLR are provided in Table 1. SST and location are the strongest predictors. Multicollinearity was checked for the MLRs created for each year. To determine if two or more of the independent parameters (predictors) were correlated, variance inflation factors (VIFS) were calculated and presented in Table 1. The VIFS were well below 5, considered the threshold for multicollinearity. The exception is for 2008 when the VIF for Latitude was 5.7. The residuals of mapped fCO$_{2w,mapped}$ versus the gridded fCO$_{2w}$ data product (1° × 1° × mo) for pixels with fCO$_{2w}$ observations show no spatial or seasonal patterns.

### 3.5. Determination of controls of fCO$_{2w}$

For investigation of processes controlling fCO$_{2w}$, the monthly changes in fCO$_{2w}$ are decomposed into a temperature term, an air-sea CO$_2$ flux term, and a residual that is controlled by biological processes and mixing:

$$\delta fCO_{2w}(Total) = \delta fCO_{2w}(SST) + \delta fCO_{2w}(FCO2) + \delta fCO_{2w}(B&M)$$  \hspace{1cm} (8)

where $\delta$ is the monthly time derivative such that $\delta fCO_{2w}$ (Total) is the monthly change in fCO$_{2w}$. The $\delta fCO_{2w}$ (SST) is the monthly change in fCO$_{2w}$ due to temperature change:

$$\delta fCO_{2w}(SST) = fCO_{2w}(SST_{i+1})−fCO_{2w}(SST_{i}) \times e^{0.0423 \Delta SST}$$  \hspace{1cm} (9)

This is referred to as the thermodynamic effect, where the subscripts i and i + 1 are the values for consecutive months. $\delta fCO_{2w}$ (FCO2) is the monthly change in fCO$_{2w}$ due to the air-sea CO$_2$ flux. This is determined in the
following manner. First the change in total dissolved inorganic carbon, DIC in the mixed layer is calculated as $F_{\text{CO}_2}/\text{MLD}$. The DIC change is converted to $f_{\text{CO}_2}$ change by combining it with the total alkalinity (TAlk) estimated according to Cai et al. (2010): 

$$\text{TAlk} = 57.3 \times \text{SSS} + 296.4.$$ 

Using $f_{\text{CO}_2}(i)$, TAlk$(i)$, SST$(i)$, and SSS$(i)$ as input functions in the CO2SYS program (V2.2, Pierrot et al., 2006), the DIC$(i)$ is

### Table 1

| Year | a   | b   | c   | d   | e   | f   | $r^2$ | #points |
|------|-----|-----|-----|-----|-----|-----|-------|---------|
| 2002 | −0.32 | 0.45 | 10.29 | −0.04 | 0.77 | 24.4 | 4.7 | 0.90 | 537 |
|      | 0.04 | 0.11 | 0.18 | 0.02 | 0.72 | 25.8 |     |       |     |
|      | 1.5  | 1.7  | 1.3  | 1.1  | 1.2  |     |     |       |     |
| 2003 | −0.56 | 0.32 | 9.24  | −0.09 | 1.11 | 32.6 | 5.2 | 0.86 | 731 |
|      | 0.03 | 0.09 | 0.17 | 0.02 | 0.66 | 24.6 |     |       |     |
|      | 1.2  | 1.4  | 1.3  | 1.2  | 1.2  |     |     |       |     |
| 2004 | −0.42 | 0.82 | 10.34 | −0.20 | 2.78 | −55.4 | 5.2 | 0.92 | 740 |
|      | 0.03 | 0.09 | 0.17 | 0.01 | 0.63 | 23.6 |     |       |     |
|      | 1.2  | 1.5  | 1.8  | 1.4  | 1.4  |     |     |       |     |
| 2005 | −0.43 | 0.49 | 8.71  | −0.07 | 7.30 | −172.0 | 6.6 | 0.85 | 664 |
|      | 0.04 | 0.12 | 0.18 | 0.02 | 0.85 | 32.1 |     |       |     |
|      | 1.1  | 1.5  | 1.6  | 1.4  | 1.6  | 1.1  |     |       |     |
| 2006 | −0.31 | 1.13 | 9.60  | −0.19 | 2.56 | −26.6 | 5.0 | 0.89 | 670 |
|      | 0.03 | 0.08 | 0.16 | 0.02 | 0.70 | 26.1 |     |       |     |
|      | 1.3  | 1.8  | 1.5  | 1.4  | 1.9  | 1.3  |     |       |     |
| 2007 | −0.62 | 1.12 | 10.56 | −0.36 | 2.75 | −77.8 | 7.0 | 0.79 | 483 |
|      | 0.05 | 0.15 | 0.32 | 0.03 | 1.33 | 49.0 |     |       |     |
|      | 1.2  | 2.3  | 1.8  | 1.4  | 2.2  | 1.2  |     |       |     |
| 2008 | −0.12 | 1.12 | 10.58 | −0.30 | 3.16 | −55.8 | 5.7 | 0.95 | 107 |
|      | 0.43 | 0.31 | 0.45 | 0.05 | 1.39 | 59.8 |     |       |     |
|    | 3.7  | 5.7  | 3.0  | 2.5  | 1.8  | 3.7  |     |       |     |
| 2009 | −0.66 | 0.30 | 7.18  | −0.47 | 0.11 | 131.9 | 6.9 | 0.84 | 125 |
|      | 0.22 | 0.24 | 0.53 | 0.07 | 1.49 | 58.8 |     |       |     |
|      | 1.5  | 2.3  | 2.3  | 1.8  | 2.1  | 1.5  |     |       |     |
| 2010 | −0.53 | 2.02 | 8.48  | −0.23 | 2.12 | −10.5 | 9.3 | 0.85 | 323 |
|      | 0.17 | 0.26 | 0.41 | 0.05 | 1.48 | 51.7 |     |       |     |
|      | 1.5  | 3.9  | 3.3  | 2.8  | 2.8  | 1.5  |     |       |     |
| 2011 | −0.32 | 0.98 | 6.65  | −0.28 | 3.06 | 46.8 | 7.2 | 0.76 | 305 |
|      | 0.13 | 0.19 | 0.34 | 0.04 | 1.51 | 59.7 |     |       |     |
|      | 1.3  | 3.0  | 2.6  | 1.5  | 3.1  | 1.3  |     |       |     |
| 2012 | −0.13 | 1.66 | 10.81 | −0.33 | 5.56 | −154.1 | 7.3 | 0.91 | 358 |
|      | 0.10 | 0.16 | 0.28 | 0.04 | 1.36 | 50.9 |     |       |     |
|      | 1.1  | 2.4  | 2.4  | 1.6  | 2.3  | 1.1  |     |       |     |
| 2013 | −0.43 | 1.30 | 11.45 | −0.47 | 4.45 | −137.5 | 8.2 | 0.83 | 219 |
|      | 0.18 | 0.23 | 0.48 | 0.05 | 1.63 | 55.5 |     |       |     |
|      | 1.4  | 2.6  | 1.9  | 1.3  | 2.0  | 1.4  |     |       |     |
| 2014 | −0.60 | 0.62 | 9.48  | −0.18 | 3.26 | −45.2 | 7.0 | 0.78 | 362 |
|      | 0.08 | 0.15 | 0.29 | 0.03 | 1.06 | 37.7 |     |       |     |
|      | 1.6  | 2.3  | 1.4  | 1.7  | 1.8  | 1.6  |     |       |     |
| 2015 | −0.84 | 0.14 | 7.76  | −0.18 | 1.20 | 70.0 | 6.0 | 0.82 | 455 |
|      | 0.04 | 0.11 | 0.27 | 0.02 | 0.90 | 34.5 |     |       |     |
|      | 1.0  | 2.1  | 1.6  | 1.0  | 1.9  | 1.0  |     |       |     |
| 2016 | −0.79 | 0.37 | 8.72  | −0.14 | 4.17 | −62.7 | 6.7 | 0.82 | 1001 |
|      | 0.03 | 0.08 | 0.20 | 0.01 | 0.45 | 17.1 |     |       |     |
|      | 1.1  | 1.3  | 1.5  | 1.4  | 1.2  | 1.1  |     |       |     |
| 2017 | −0.42 | 0.56 | 9.49  | −0.22 | 5.65 | −112.6 | 6.2 | 0.87 | 974 |
|      | 0.03 | 0.08 | 0.15 | 0.02 | 0.58 | 17.1 |     |       |     |
|      | 2.2  | 3.2  | 3.5  | 1.5  | 2.1  | 2.2  |     |       |     |

Note. The first row (in bold) gives the coefficients for each variable in the MLR. The second row (in italics) for each annual entry is the error of the coefficient. The third row gives the variance inflation factor.

These regressions are used to create the mapped $f_{\text{CO}_2}$ fields for each year using the $1^\circ \times 1^\circ \times$ mo gridded data product.

$\text{fCO}_2$,mapped = $a$ Longitude + $b$ Latitude + $c$ SST + $d$ MLD + $e$ SSS + $f$
calculated. Then $\text{DIC}_{(i+1)} = \text{DIC}_{(i)} + \text{FCO}_2/\text{MLD}$. Accordingly, $\text{FCO}_{2w(i+1)}$ is calculated with CO2SYS using $\text{DIC}_{(i+1)}$, $\text{TAlk}_{(i+1)}$, $\text{SST}_{(i+1)}$, and $\text{SSS}_{(i+1)}$. The change in $\text{FCO}_{2w}$ due to $\text{FCO}_2$ is as follows:

$$
\delta \text{FCO}_{2w}(\text{FCO}_2) = \text{FCO}_{2w(i+1)} - \text{FCO}_{2w(i)}
$$

(10)

The $\delta \text{FCO}_{2w}(\text{B&M})$ is determined as the residual:

$$
\delta \text{FCO}_{2w}(\text{B&M}) = \delta \text{FCO}_{2w}(\text{Total}) - \delta \text{FCO}_{2w}(\text{SST}) - \delta \text{FCO}_{2w}(\text{FCO}_2)
$$

(11)

The $\delta \text{FCO}_{2w}(\text{B&M})$ is the monthly change caused by other processes that impact $\text{FCO}_{2w}$, including biological drawdown due to biological production, and remineralization and respiration in the mixed layer, the sum which is referred to as net community production. The term also includes effects of mixing and entrainment of waters with different DIC concentrations (and $\text{FCO}_{2w}$) from below the mixed layer and from lateral transport. The $\delta \text{FCO}_{2w}(\text{B&M})$ also implicitly contains processes that change alkalinity in the mixed layer. Alkalinity increases will lower the $\text{FCO}_{2w}$. While some of the alkalinity effects could, in principle, be determined from changes in salinity due to the strong relationship between salinity and alkalinity, month to month salinity changes are small such that the alkalinity effects on $\text{FCO}_{2w}$ are not studied separately but rather included in this residual term.

The $\text{FCO}_{2w}$ is not a state variable and the deconvolution described in (8) is not strictly additive. For instance, the $\delta \text{FCO}_{2w}(\text{SST})$ does not change the state variable DIC, while the $\delta \text{FCO}_{2w}(\text{FCO}_2)$ and $\delta \text{FCO}_{2w}(\text{B&M})$ terms do impact DIC. However, on monthly basis where the changes are relatively small, a linear approximation as used here is justified. More formal deconvolutions to tease out the processes influencing $\text{FCO}_{2w}$ have been performed (Metzl et al., 2010; Takahashi et al., 1993; Thomas et al., 2008) using the state variables DIC and TAlk, and the derivatives of $\text{FCO}_{2w}$ with DIC, TAlk, temperature, and salinity. In our case where DIC and TAlk were not measured but rather estimated using salinity and $\text{FCO}_{2w}$, the uncertainty in the estimated DIC and TAlk changes, along with use of a modeled mixed layer, warrant the simpler approach of using a residual B&M term that includes several processes.

The uncertainties in the deconvolution cannot be easily quantified. They include uncertainty in the flux of $\approx 20\%$–$30\%$ (Wanninkhof, 2014), uncertainty in MLD estimate, spatial and temporal variability in the grid cell, and nonlinearity of processes changing $\text{FCO}_{2w}$. Therefore, no uncertainty estimate is provided due to challenges in properly determining and propagating the errors in each term. Biases in $\delta \text{FCO}_{2w}(\text{Total})$, $\delta \text{FCO}_{2w}(\text{SST})$, and $\delta \text{FCO}_{2w}(\text{FCO}_2)$ will be reflected in the $\delta \text{FCO}_{2w}(\text{B&M})$ term. Accordingly, this is interpreted with caution.

### 3.6. Deseasonalization Using Harmonic Fits

The seasonal cycle is the primary mode of variability in the region and it exhibits a regular pattern. The $\text{FCO}_{2w}$ and SST can be well represented with a harmonic fit. In turn, this fit can be subtracted from the monthly values to more clearly discern the multiannual trends. The seasonal cycles of $\text{FCO}_{2w}$ and SST of the gridded data product ($1\degree \times 1\degree \times \text{mo}$) are fit to a harmonic function as detailed in Gruber et al. (1998) and Park and Wanninkhof (2012). For $\text{FCO}_{2w}$,

$$
\text{FCO}_{2w, \text{fit}} = \text{FCO}_{2w, \text{mean}} + a \sin (2 \pi \text{mo}/12) + b \cos (2 \pi \text{mo}/12)
$$

(12)

where $\text{FCO}_{2w, \text{mean}}$ is the average of all the $\text{FCO}_{2w}$ values for all grid points over the 16-year period (373.6 $\mu$atm), mo is the month (1–12), and $a$ and $b$ are fitting parameters optimized through a cost function. Using $a = -20.4$ and $b = -10.6$ results in an average root-mean-square error, RMSE between the data and the fit of 1.3 $\mu$atm. The same procedure is used for SST, with an average SST of 26.95 °C and a RMSE of 0.005 °C. The monthly average values of $\text{FCO}_{2w}$ and SST determined with harmonic fits covering the 16-year period are subtracted from the monthly $\text{FCO}_{2w}$ and SST values for the region to obtain a seasonality detrended, or deseasonalized, product. Using the deseasonalized residuals of $\text{FCO}_{2w}$ and SST improve the detection of longer-term trends and increases the confidence level of the trends. The detrending procedures do not bias the results and the same multiannual patterns are discerned with the original nondeseasonalized observations and mapped products.
levels are in February (Figures 3a and 3e). and SST from March 2002 to values are in the northwestern part of the region. The relationship of can be directly affected by in the Southeastern Gulf of Mexico cycles at are not clearly observed, except in the southeastern part of the region and air (Table 1) that show strong dependencies with position. Based on the MLRs, the on daily scale for the Caribbean data set is attributed to and SST data from 2002 to 2018. The and SST are smaller than the extremes in the seasonal cycle (Figures 4a and 4b). Local data near Bermuda by Bates et al. (2001) that showed a with SST is apparent in the patterns. The SSS shows spatial and seasonal variations (Figures 3e and f), but the correspondence with fCO2w is high in the southwest and lower values to the northeast. In summertime the signal not being resolved at regional scale of analyses and the ships covering the region at a set schedule. That is, the cruise ships cover the same locations at the same time of day for each cruise itinerary, and the ships spend more time at sea during nighttime such that the diurnal SST and fCO2w cycles at are not well resolved.

### 4. Results and Discussion

The fCO2w levels are influenced by processes operating over a range of temporal and spatial scales, from hours to decades, and from local to regional. The patterns of fCO2w and air-sea CO2 fluxes in time and space are discussed for the study region shown in Figure 1 nominally covering 16° N–28°N and 88°W–62°W. Unless noted, the mapped product is used which is on a (1° × 1° × mo) grid from the annually derived MLRs (Table 1).

The lack of observations in the Southeastern Gulf of Mexico north of 25°N can cause the interpolation with MLRs to yield erroneous results in this area as it is affected by other influences than typical for the Caribbean Sea. For example, shelf specific processes on the West Florida shelf, and Mississippi outflow can impact the fCO2w in the Southeastern Gulf of Mexico (Robbins et al., 2018; Xue et al., 2016; Yang et al., 2015). The area is included in the calculation of the total air-sea CO2 flux, but the results might not be fully representative.

The complete observational dataset of fCO2w and SST from March 2002 to 2018 are plotted against time in Figure 2. The power spectrum of fCO2w shows a singular strong peak at annual time scales. This corresponds with the power spectrum of temperature. This differs from the analysis of mooring based CO2 data near Bermuda by Bates et al. (2001) that showed a strong daily peak and a broad peak centered on a week based on analysis of monthly data. In both the Bermuda case and this study, the fCO2w and SST spectra match closely, reflecting the role of SST as a direct and indirect driver of fCO2w. The lack of a peak in SST and fCO2w on daily scale for the Caribbean data set is attributed to the signal not being resolved at regional scale of analyses and the ships covering the region at a set schedule. That is, the cruise ships cover the same locations at the same time of day for each cruise itinerary, and the ships spend more time at sea during nighttime such that the diurnal SST and fCO2w cycles at are not well resolved.

#### 4.1. Spatial Differences

Spatial differences are small compared to seasonal changes but they are observable. This can also be inferred from the MLRs of fCO2w (Table 1) that show strong dependencies with position. Based on the MLRs, the average fCO2w values decrease towards the north and toward the east. As temperature is the strongest predictor in the MLRs and there is a correlation between location and temperature, the spatial trends are primarily related to SST. Maps of fCO2w, SST, and SSS are shown in Figures 3a–3f using February and August 2017 as representative examples. The largest spatial differences in fCO2w are apparent in the wintertime (Figure 3a) with high values in the southwest and lower values to the northeast. In summertime (Figure 3b), the highest fCO2w values are in the northwestern part of the region. The relationship of fCO2w with SST is apparent in the patterns. The SSS shows spatial and seasonal variations (Figures 3e and f), but the correspondence with fCO2w is not clearly observed, except in the southeastern part of the region where lower SSS waters typically also have lower fCO2w.

The differences in the annual cycle for four representative locations shown in Figure 1 are highlighted in Figure 4 where the gridded fCO2w, SST, SSS, and MLD products are plotted versus month in 2017. Monthly mean gridded data products from the Florida Current, South of the Florida Keys (pixel centered at 24.5°N, 81.5°W); the Outer Bahamas Banks (Turks and Caicos, 21.5°N, 70.5°W); the Lesser Antilles (18.5°N, 64.5°W); and the western Caribbean (Cayman Islands, 19.5°N, 81.5°W) are shown. The spatial differences in fCO2w and SST are smaller than the extremes in the seasonal cycle (Figures 4a and 4b). Local anomalies are apparent, particularly in SSS (Figure 4c) where a strong minimum is observed in the Lesser Antilles region in the summer and fall, attributed to a combination of Orinoco river outflow and regional precipitation anomalies (Ibáñez et al., 2017). Annual SSS anomalies are seen in other locations as well, such as the minimum observed in summer at the Florida current location. The fCO2w can be directly affected by
dilution with rainfall (Turk et al., 2010) or, in case of riverine runoff, indirectly as well though enhanced biological productivity from excess nutrients or alkalinity carried by rivers. The low $f_{CO_{2w}}$ levels near the Lesser Antilles appear largely caused by dilution as the observed decrease in $f_{CO_{2w}}$ corresponds to the thermodynamic effect that is 17.5 μatm per unit salinity change (Takahashi et al., 1993). For the low salinities in the Florida current, there is no corresponding reduction in $f_{CO_{2w}}$, likely because of the increasing SST. The model derived MLD for four locations are given in Figure 4d. All show a seasonal cycle with late summer MLD minima. The Florida current site shows MLD as shallow as 15 m during the summer. The monthly MLD product shows significantly deeper MLD in the western Caribbean compared to the other locations throughout much of 2017.

Local deviations from basin-wide patterns are apparent for other years as well. For example, a maximum in $f_{CO_{2w}}$ of 445 μatm that was observed in the Florida current in July 2016 (not shown) was ≈20 μatm higher than values observed in the adjacent months of 2016 and the $f_{CO_{2w}}$ values observed in July 2017 (Figure 4a). The corresponding SST in July 2016, of 30.3 °C, is only ≈0.4 °C higher than July 2017. Thus, the $f_{CO_{2w}}$ anomaly in July 2016 is about twofold larger than expected from a thermodynamic temperature effect of 4.23% °C$^{-1}$. This high $f_{CO_{2w}}$ value is attributed to entrainment of West Florida Shelf water and is an example of local anomalies that are present throughout the record. However, while local monthly anomalies in SST, SSS, and $f_{CO_{2w}}$ are apparent for different locations, and for different years, the

**Figure 3.** Contour plots of surface water properties in February (left panels) and August (right panels) 2017: $f_{CO_{2w}}$ (a, b); SST (c, d); and SSS (e, f). Color bars with ranges are at the bottom of each panel.
region as a whole behaves similarly and, as elaborated upon below, can be viewed as a single entity to assess seasonal to interannual variability and trends.

4.2. Subannual (Seasonal) Patterns

Different factors influence the seasonal fCO$_{2w}$ cycle but seasonal changes in SST are the dominant influence on fCO$_{2w}$. The thermodynamic dependence of fCO$_{2w}$ to SST is 0.0423°C$^{-1}$ (4.23% per degree centigrade) due to decreasing solubility and shifting of carbonate equilibrium constants favoring fCO$_{2w}$ increase. The SST changes by ≈4 °C over the annual cycle in the Caribbean and thus has the potential to change fCO$_{2w}$ by ≈60 μatm.

Seasonal changes in MLD can affect fCO$_{2w}$ in differing and opposing ways. Deepening mixed layers can entrain colder water that will thermodynamically decrease the fCO$_{2w}$. However, the waters can contain higher DIC, which will increase fCO$_{2w}$. Entrainment and mixing can also supply nutrients, and the resulting primary production can decrease fCO$_{2w}$. Variability of fCO$_{2w}$ due to mixed layer dynamics and nutrient supply are not well understood (Fawcett et al., 2014). The gradients in inorganic carbon and nutrients below the mixed layer up to the maximum wintertime MLD are small based on GO-SHIP line A22 cruise data (cchdo.

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**Figure 4.** Monthly averaged surface water properties at four locations in the Caribbean based on the gridded data product (1° × 1° × mo) in 2017: Lesser Antilles (18.5°N, 64.5°W) red circles, medium-dashed line; western Caribbean (19.5°N, 81.5°W), blue plusses, long-dashed line; outer Bahamas Bank (21.5°N, 70.5°W), brown diamonds, short-dashed line; and Florida Current (24.5°N, 81.5°W), black open circles, solid black line. fCO$_{2w}$ (a); SST (b); SSS (c); and MLD from the HYCOM model output (d). The vertical lines give the sd of the observations for the particular month in 2017 for the 1° × 1° pixel. For clarity only half the line depicting sd are shown for some points.
uscd.edu) in the eastern part of the region such that entrainment of DIC and nutrients is thought to have a small effect. Lowering of temperature and longer response time to gas transfer are the dominant responses to MLD deepening (Park & Wanninkhof, 2012).

The average seasonal cycle for MLD, NFLH, SST, and fCO$_2$ is based on the monthly mapped products for the entire region are shown in Figure 5 with the sd of the monthly values over the 16 years depicted as error bars. Multiannual trends have not been subtracted. The MLD (Figure 5a) shows shallow depths of $21 \pm 6$ m from May to September and rapid deepening thereafter reaching a maximum average depth of $54 \pm 14$ m in January after which a shallowing occurs in early spring. The NFLH (Figure 5b) shows a minimum from March to May. The decrease corresponds roughly with the mixed layer shallowing and possibly caused by limited diffusion of nutrients from below during this time.

The seasonal signal in SST (Figure 5c) and fCO$_2$ is well represented using a harmonic function (equation (12)). The seasonal range in fCO$_2$ is $40 \mu$atm and $4 ^\circ$C in SST with minimum values in February and maximum values in August/September. There is no distinct annual cycle in salinity (not shown).

The close correspondence in phase and shape of the seasonal cycle of SST and fCO$_2$ indicates a strong correlation between them. Figure 6 shows the monthly averaged fCO$_2$ plotted against SST following Lefèvre

Figure 5. The 16-year monthly averages of the mapped products in the Caribbean with error bars showing the sd of the monthly values. (a) Mixed layer depths from the HYCOM model; (b) Normalized Fluorescent Line Height from MODIS; (c) SST (dashed line) with harmonic fit (thick line); and (d) monthly fCO$_2$ (dashed line) with harmonic fit (thick line). The horizontal dashed line in c and d are the 16-year averages of SST and fCO$_2$. 

10.1029/2019JC015366
and Taylor (2002). This diagram illustrates the factors influencing the seasonal cycle in fCO$_{2w}$. The average change in fCO$_{2w}$ with respect to temperature based on a linear regression of monthly fCO$_{2w}$ versus SST is 9.5 μatm °C$^{-1}$ (2.5% °C$^{-1}$). This is about 60% of the thermodynamic trend, which is shown as a dashed line in Figure 6 indicating that there are counteracting factors impacting the seasonal cycle of fCO$_{2w}$.

The progression of SST and fCO$_{2w}$ through the year can be delineated by periods of warming from March to August, cooling from September to February, and transitions in February–March, and August–September, respectively (Figure 5c). The seasonal trends in fCO$_{2w}$ are caused by interplay of temperature changes, gas transfer, mixing, and biologically mediated processes. The partitioning of the processes (equation (8)) for the seasonal cycle using gridded product are shown in Figure 7. The deconvolution of fCO$_{2w}$ for the full 16-year record is presented in the attribution section below.

As depicted in Figures 6 and 7, during periods of warming (March–September) the fCO$_{2w}$ increases by ≈8–10 μatm/mo or 2.5%–3.1% °C$^{-1}$. The thermodynamic effect, which is predominant in the annual cycle, increases fCO$_{2w}$. The net effect of B&M is a drawdown of fCO$_{2w}$ during this warming period. The effect of air-sea gas exchange is of the same magnitude as the B&M impacts (≈1 to 3 μatm/mo) but changes sign during the warming period as it depends on the sign of the ΔfCO$_{2}$ gradient. It contributes to increasing fCO$_{2w}$ when the area is a net sink from March to May and to decreasing fCO$_{2w}$ from June to September.
During the cooling period from September to February, fCO$_{2w}$ decreases by $\approx8$–10 $\mu$atm/mo, 2.5%–3% °C$^{-1}$. Again, the thermodynamic effect dominates, with counteracting effects of B&M. The effect of air-sea fluxes reverses from a CO$_2$ sink in October to a small CO$_2$ source for the remainder of the cooling period. The B&M effect increases fCO$_{2w}$ during the cooling period except in February. In the transition month from cooling to warming (February–March), the flux into the ocean and B&M are of equal but opposite magnitude. In September, when the transition from warming to cooling occurs, gas evasion counteracts the small net warming with a small contribution of B&M.

The B&M term is calculated as a residual (equation (11)) between the observed monthly fCO$_{2w}$ change, the thermodynamic effect, and the effect of gas transfer. The observed monthly change in fCO$_{2w}$ and the thermodynamic effect are well constrained but biases in the gas flux can be significant at 20%–30%. Generally, the air-sea CO$_2$ flux term is a smaller contributor. The $\delta$fCO$_{2w}$\text{BM}$^{\text{BM}}$ is negative from February to August and positive from September to January. This does not match the NFLH annual progression that shows a minimum in spring and a maximum in December (Figure 5b). However, $\delta$fCO$_{2w}$\text{BM}$^{\text{BM}}$ does show broad correspondence with MLD (Figure 5a). This suggests that during periods of shallow mixed layers the biological production is lower but still draws down the fCO$_{2w}$ while during times of mixed layer deepening nutrients are entrained, enhancing productivity and elevating the NFLH. However, due to the deeper MLD the effect on fCO$_{2w}$ is smaller. Also, with deeper MLD remineralized carbon in the form of DIC are brought to the surface that causes the B&M effect to increase fCO$_{2w}$.

4.3. Temporal Trend

There are clear trends and patterns on multiannual scales in the region. The fCO$_{2w}$ observations versus time show a well-defined seasonal cycle and long-term trends (Figure 2). For the entire observational record from March 2002 to February 2010, the observed fCO$_{2w}$ is $1.30 \pm 0.003$ $\mu$atm/year but there is a large change in the middle part of the 16-year record, such that it can represented as two linear segments (Figure 2). From March 2002 to February 2010, the observed fCO$_{2w}$ decreases by $-1.37 \pm 0.017$ $\mu$atm/year with a reversal in trend to $3.69 \pm 0.011$ $\mu$atm/year from March 2010 to February 2018. The uncertainties are the standard errors in the slope and are due to the large number of data points. As data coverage is sparser from 2007 to 2011 than at the beginning and end of record, the exact year of turnaround in trend is not well defined in the observations.

A similar trend in the mapped product for the whole region is seen using the deseasonalized monthly residuals, albeit with lower, and only positive trends (Figure 8a). It shows an increase of $1.66 \pm 0.09$ $\mu$atm/year for the entire record. The trend is $0.60 \pm 0.21$ $\mu$atm/year from 2002 to 2010 and $2.55 \pm 0.28$ $\mu$atm/year from 2010 to 2018. While the trends for the observations and mapped values differ, the pattern of little change or decreases in fCO$_{2w}$ for the first 8 years followed by a strong positive trend is the same. The differences between the observations and the gridded mapped product are attributed to a combination of stronger trends in fCO$_{2w}$ and SST in the central region along the tracks where the observations are taken and the use of MLRs in the mapped product that causes features to be smoothed.

To determine the timeframe when the trend changes, 3-year increments are compared, using the mapped product (Table 2). This follows the approach of Fay and McKinley (2013) and allows trends for different increments and lengths to be compared. There is a broad minimum in fCO$_{2w}$ trends centered on the 2008–2011 time frame that cannot be attributed to a single anomalous year. The residuals of the detrended/deseasonalized fCO$_{2w}$, data (Figure 8a) show large positive and negative anomalies with similar anomalies in SST (Figure 8b) in the 2008–2011 period. SST and fCO$_{2w}$ show negative anomalies (Figure 8) in the
fall/winter of 2010 and in 2011. A strong positive anomaly in SST and fCO$_{2w}$ occurs in the summer of 2010. These anomalies have a determining impact on the multi-year trends.

The trends can be put in context of the rising atmospheric CO$_2$ levels. At steady state, the fCO$_{2w}$ would keep up with the atmospheric CO$_2$ rise of, on average, 2.21 ppm/year from 2002 to 2018 based on the data from the KEY and RPB stations. This rise in mole fraction of CO$_2$ in the marine boundary layer, XCO$_2$, translates to 2.13 μatm/year at 100% humidity and 27 °C for fCO$_{2w}$. Thus, for the first part of the record, the fCO$_{2w}$ shows an appreciable lag while in the second part of the record the increase is greater than expected from invasion of CO$_2$ from the atmosphere. Over the entire 16 years, the fCO$_{2w}$ has increased but it is appreciably less than expected if the Caribbean Sea had kept up with the atmospheric increase leading to a deficit of 13 μatm (24%) compared to the expected trend by the end of the record. The short-dashed line in Figure 8a is the expected trend in the residual if fCO$_{2w}$ followed atmospheric CO$_2$ increase and SST trends (Figure 8b).

### 4.4. Attribution of Trends in fCO$_{2w}$

For the attribution of multiyear trends, we first describe the statistical correlations between the variables in the gridded mapped product. This is followed by the description of how SST and MLD impact the trends. Finally, the effect of temperature, gas exchange, biology, and mixing (B&M) on multiyear trends fCO$_{2w}$ are described. No single overriding cause for the large change in trend in fCO$_{2w}$ is apparent but rather it is a combination of factors, including multiyear seasonal anomalies in the middle part of the record that cause the large changes in fCO$_{2w}$ trends.

The correlation analysis of the gridded data provides a summary of the correlation of fCO$_{2w}$ with the environmental parameters as well as correlations between the different parameters (Figure 9). For the whole mapped product, the strongest dependency of fCO$_{2w}$ is with temperature (positive) but there are also statistically significant correlations (at >90% confidence level) with wind (negative), position (negative), MLD (negative), and NFLH (positive). The fCO$_{2w}$ shows insignificant statistical dependency on SSS. The sign of the correlations indicates that fCO$_{2w}$ increases in response to the SST increase and decrease in response to increasing wind. The fCO$_{2w}$ is negatively related with mixed layer depth indicating that deeper mixed layers lead to lower fCO$_{2w}$. The NFLH shows a positive correlation with fCO$_{2w}$, similar to what was shown in the sub-annual analysis, contrary to the expectation that greater fluorescence would indicate higher productivity and thus lower fCO$_{2w}$. The unexpected signs of the correlations of fCO$_{2w}$ with MLD and NFLH are, in part, due to parameters being cross-correlated such that the statistical relations do not always imply causality. For example, SST and MLD are negatively correlated and the strong dependency of fCO$_{2w}$ on SST will decrease the effect of entrainment of DIC due to mixed layer deepening. The NFLH and mixed layer depth are positively correlated, while fCO$_{2w}$ and MLD are negatively correlated confounding simple causal relationships of the fCO$_{2w}$ with individual parameters. The positive sign and weak correlations suggest that NFLH is not a strong predictor of the effect of productivity on fCO$_{2w}$ in the region.

For the whole time period from 2002 to 2018, there is no statistically significant trend in observed SST but there is a decreasing trend in the observed SST from March 2002 to February 2010 of $-0.178 \pm 0.002$ °C/year ($P$ value < 0.001) and increasing from March 2010 to February 2018 of $0.194 \pm 0.001$ °C/year ($P$ value = 0;
For the OISST 1° × 1° × mo mapped product for the whole region, the trends are smaller but of the same sign, $-0.06 \pm 0.05$ °C/year from March 2002 to February 2010 and $0.075 \pm 0.07$ °C/year for March 2010 to February 2018. The lower trend is attributed to the inherent smoothing of the OISST and compensating differences in the region on a monthly basis compared to along the cruise tracks.

Of the parameters investigated, SST has the greatest influence on $fCO_{2w}$. Based on the relationship of $\partial fCO_{2w} / \partial SST^{-1}$ of 0.042 °C$^{-1}$, which under average conditions in the Caribbean Sea causes $fCO_{2w}$ to change by $\approx 16$ μatm per °C, the trend in observed $fCO_{2w}$ due to surface ocean cooling/warming would be $-2.9$ μatm/year from 2002 to 2010 and $3.1$ μatm/year from 2010 to 2018 for the observed SST, and $-1.0$ and 1.2 μatm/year, using the mapped OISST product. Figure 8a includes trend lines depicting the changes in $fCO_{2w}$ expected due to changes in atmospheric CO$_2$ and SST (thermodynamic effect) for the OISST mapped product in these periods.

The $fCO_{2w}$ trends are affected by seasonal changes in MLD as shown in Park and Wanninkhof (2012). They showed that the wintertime $fCO_{2w}$ decreased in the northeastern part of the study region from 2002 to 2009. This was attributed to longer periods with deep mixed layer depth (>30 m) providing more volume for CO$_2$ uptake and thus small changes in $fCO_{2w}$ due to gas exchange. The summertime $fCO_{2w}$ increased over the same time period. However, on annual scale the $fCO_{2w}$ remained invariant thereby increasing the uptake significantly. Following this study, a more detailed investigation of the seasonal controls on $fCO_{2w}$ trends

| Start/End  | 2002  | 2005  | 2008  | 2010  | 2011  | 2014  | 2017  | 2018  |
|-----------|-------|-------|-------|-------|-------|-------|-------|-------|
| 2002      | 0.7   | -1.3  | -1.8  | -1.5  | -0.6  | 1.1   | 1.3   |       |
| 2005      | -4.1  | -4.0  | -2.1  | -0.3  | 1.9   | 2.0   |       |       |
| 2008      | -5.9  | 1.1   | 2.6   | 4.1   | 4.0   |       |       |       |
| 2010      | 2.3   | 4.2   | 3.9   |       |       |       |       |       |
| 2011      | 3.5   | 4.3   | 3.9   |       |       |       |       |       |
| 2014      | 2.2   | 1.5   |       |       |       |       |       |       |
| 2017      |       |       |       |       |       |       |       |       |
| 2018      |       |       |       |       |       |       |       |       |

*aThe trends are determined from the gridded mapped product and color coded from strongly negative (dark green) to strongly positive (red). For example, from 2011 to 2017 the trend is strongly positive (red) at 4.3 μatm/year.
Table 3
Trends in SST, MLD, and fCO_{2w} for Wintertime and Summertime\textsuperscript{a}

| Range      | Data      | SST Min/Winter | SST Max/Summer | MLD Min/Summer | MLD Max/Winter | fCO_{2w} | fCO_{2w} |
|------------|-----------|----------------|----------------|----------------|----------------|---------|---------|
| 2002–2018  | Mapped product | 0.05 ± 0.02    | 0.03 ± 0.01    | 0.27 ± 0.09    | 0.23 ± 0.14    | 1.72 ± 0.18 | 1.65 ± 0.17 |
|            | Gridded data    | 0.02 ± 0.05    | −0.02 ± 0.03   | 0.28 ± 0.14    | 0.36 ± 0.23    | 1.39 ± 0.64 | 1.44 ± 0.33 |
|            | Mapped@Obs    | 0.04 ± 0.04    | 0.00 ± 0.03    | 0.32 ± 0.16    | −0.06 ± 0.32   | −2.64 ± 1.01 | 1.12 ± 0.65 |
| 2002–2010  | Mapped product | −0.01 ± 0.04   | 0.05 ± 0.03    | −0.27 ± 0.12   | 0.06 ± 0.37    | −0.06 ± 0.32 | 1.37 ± 0.5 |
|            | Gridded data    | −0.06 ± 0.10   | −0.02 ± 0.05   | −0.33 ± 0.14   | 0.99 ± 0.64    | 2.82 ± 0.46 | 2.10 ± 0.48 |
|            | Mapped@Obs    | −0.14 ± 0.08   | −0.01 ± 0.05   | −0.33 ± 0.14   | 0.99 ± 0.64    | 2.82 ± 0.46 | 2.10 ± 0.48 |
| 2010–2018  | Mapped product | 0.12 ± 0.04    | 0.02 ± 0.05    | 0.78 ± 0.24    | 0.55 ± 0.34    | 4.67 ± 0.95 | 3.33 ± 0.74 |
|            | Gridded data    | 0.26 ± 0.07    | 0.16 ± 0.08    | 1.06 ± 0.37    | 0.22 ± 0.47    |         |         |
|            | Mapped@Obs    | 0.24 ± 0.06    | 0.13 ± 0.07    | 1.06 ± 0.37    | 0.22 ± 0.47    |         |         |

\textsuperscript{a}Values are per year (SST: °C/year, MLD: m/year, fCO_{2w}: μatm/year)

The mapped monthly fCO_{2w}, MLD, and SST products are averaged into wintertime (December–February) and summertime (June–August) values. As with the annual analysis, there is a change in the midpoint of the record in winter and summer patterns around 2010. The winter and summer trends for the 2002 to 2010 and 2010 to 2018 time frames are shown in Table 3.

The wintertime and summertime data are also investigated to determine differences in trends between gridded data products and mapped data for SST and MLD. For the 1° × 1° gridded data, the trends in situ measured SST are compared with the OISST, denoted in Table 3 as Obs and Mapped@Obs, respectively. This indicates if there are biases in seasonal trends using the MLRs to estimate fCO_{2w}, compared to measured fCO_{2w}.

Over the entire period 2002–2018, the trends during summer and winter for the mapped product and gridded data are consistent but with some differences (Table 3). Wintertime SST shows a slight warming for the mapped product and gridded data of 0.02 to 0.05 °C/year. The summertime OISST for the whole region show an increase as well of 0.03 ± 0.01 °C/year while observations along the cruise tracks are showing a non-significant cooling with a change of −0.02 ± 0.03 °C/year. The OISST at the sampling locations show no trend but overall the observations, gridded data, and mapped products agree to within their uncertainties. The MLD shows an appreciable increase of ≈0.3 m/year over the 16-year time period with general correspondence between summer and winter.

When breaking down the summer/winter trends into two time periods, differences are more apparent between the gridded data and the gridded mapped products. From 2002 to 2010, the gridded SST data along the cruise tracks show a decrease of ≈−0.16 °C/year in wintertime and no significant change in summer. The whole region, however, does not show a strong trend in OISST in winter. The differences are in the western region, which was not well sampled by the ships during this time period. The area shows a slight warming in the OISST product. The MLDs show a decrease of 0.3 m/year in summer and a large increase of 1 m/year in summertime at the observation. Summertime shallowing is the same but there is no deepening in the mapped product. This wintertime MLD increase at the observational grid points correlates with the SST decrease during this period and similarly the lack of SST and MLD change in the mapped product. As a result, the fCO_{2w} shows a decrease of −0.06 (regional mapped product) to −2.64 (mapped) μatm/year during the wintertime and an increase of 1.37 (mapped) to 1.12 μatm/year (gridded) in the summer. Thus, during 2002–2010, the steady (mapped) or decreasing fCO_{2w} (gridded) are largely driven by the trends in wintertime similar to the results for one location as presented in Park and Wanninkhof (2012).

For the 2010–2018 time period, there are coherent positive trends for both winter and summer with SST increasing 0.2 °C/year. The MLD increases, particularly strongly in the summertime, and the fCO_{2w} increases on the order of 2 to 4 μatm/year. Winter and summertime trends for fCO_{2w} are also more similar compared to the 2002–2010 time frame where the seasonal trends diverged. The increases in fCO_{2w} and SST are significantly higher for the gridded data than for the mapped product for the whole region, again indicating that the trends in fCO_{2w} and forcing functions are stronger in the areas along the cruises’ tracks.
To further elucidate the physical and biogeochemical factors contributing to the observed trends in fCO$_{2w}$ over the entire period, the same deconvolution is used as for the seasonal attribution (equation (8)). The monthly values of δfCO$_{2w}$(SST), δfCO$_{2w}$(F$_{CO2}$), and δfCO$_{2w}$(B&M) are determined from 2002 to 2018. Then the 16-year average monthly values (Figure 7) are subtracted to deseasonalize the δfCO$_{2w}$ data in order to determine the monthly anomalies over the 16-year record. To discern seasonal anomalous patterns, 3-month averages are used (Figure 10). The deseasonalized record shows a general anticorrelation of δfCO$_{2w}$(SST) and δfCO$_{2w}$(B&M) for the times with large deviations. This suggests that during anomalous warming, there is increased biological drawdown, and during cooling higher fCO$_{2w}$ likely caused by mixing. However, for small δfCO$_{2w}$, the δfCO$_{2w}$(SST), and the δfCO$_{2w}$(B&M) can act synergistically or antagonistically.

The δfCO$_{2w}$(F$_{CO2}$) shows smaller seasonal anomalies compared to δfCO$_{2w}$(SST) and δfCO$_{2w}$(B&M). However, the δfCO$_{2w}$(F$_{CO2}$) shows persistent trends and is negative at the start of the record from 2002 to 2007, that is, air-sea gas flux causes a net loss of carbon in the mixed layer. It is predominantly positive from 2008 up to 2013. It does not show strong positive or negative values from 2013 onward.

Figure 10 shows large positive and negative excursions of seasonal δfCO$_{2w}$(SST) and δfCO$_{2w}$(B&M) in the middle of the record, from 2009 to 2011. The large changes in deseasonalized δfCO$_{2w}$(SST) are opposed by δfCO$_{2w}$(B&M). From 2009 to 2010, the springtime positive δfCO$_{2w}$(SST) is compensated by a negative late fall/winter time δfCO$_{2w}$(SST). The δfCO$_{2w}$(B&M) shows an opposite phase to the large excursions in δfCO$_{2w}$(SST), but they are smaller magnitude to δfCO$_{2w}$(SST).

The change in trend in fCO$_{2w}$ in the middle part of the record (2009–2011) is strongly influenced by anomalous physical and associated biogeochemical conditions in this time period. In the 2009–2011 transition, there are 3 consecutive years of large positive changes in δfCO$_{2w}$(SST) in Spring/Summer (Figure 10) and large negative δfCO$_{2w}$(SST) in fall/winter for 2010 and 2011. These changes are partially offset by opposing δfCO$_{2w}$(B&M). The net effect is that the processes lead to low fCO$_{2w}$ from 2009 to 2011. This fCO$_{2w}$ minimum has a large influence on the multiyear trends before and after. The period from 2010 to 2018 shows significantly smaller seasonal anomalies in the processes controlling fCO$_{2w}$ as indicated by the smaller δfCO$_{2w}$(SST) and δfCO$_{2w}$(B&M) values.

Changes in MLD are an important contributor to the changes in δfCO$_{2w}$ through its effect on SST, F$_{CO2}$, and B&M. The MLD influences δfCO$_{2w}$(SST) through cooling during MLD deepening and accelerated warming during shallowing. The δfCO$_{2w}$(F$_{CO2}$) is impacted by MLD as deeper mixed layers cause smaller changes in fCO$_{2w}$ due to the volume effect. The MLD deepening impacts δfCO$_{2w}$(B&M) through differing and opposing means. Increased nutrient supply can enhance productivity that lowers fCO$_{2w}$ but increased DIC supply will increase fCO$_{2w}$. As shown in Figure 11, during the first part of the record from 2002 to 2010, the winters of 2004/2005 and 2008/2009 show mixed layers than are 5 m deeper than average. From 2010 to 2018, there is a general trend of increasing wintertime MLD with large increases (≈6 m) for the winters of 2015 and 2016 (Figure 11). The MLD increases appear a major contributor to the increase in trend of fCO$_{2w}$ for the 2010–2018 time period.
4.5. Air-Sea CO₂ Fluxes in the Caribbean

The air-sea CO₂ fluxes in the region are quantified by using the fCO₂ₘₐₚₚ mapped product (fCO₂ₘₐₚₚ,mapped). It is merged with the monthly averaged fCO₂ₘₚₚ determined from the MBL product to determine the monthly ΔfCO₂. The OISST, SSSbscom, and the second moment of the winds, Δ<ub>, are used to determine the monthly fluxes according to equations (5) and (6).

Overall, the seasonal progression in fCO₂ₘₚₚ controls an air-sea CO₂ flux pattern that is characteristic for the subtropical and tropical regions of CO₂ outgassing in the summer and invasion in the winter (Bates et al., 2014; Takahashi et al., 2002). The monthly averaged CO₂ flux, Δ<ub>, and ΔfCO₂ over the 16-year period are shown in Figure 12 along with the annual means for the region. Winds show maxima in wintertime that are 50% greater than summertime values. The long-term trends in fluxes are in accord with the trends in ΔfCO₂, and changes in wind play a secondary role. There is no trend in Δ<ub> from 2002 to 2018, but there are some multiannual anomalies. Of note is the increase in Δ<ub> of 1.3% year⁻¹ during the wintertime from 2002 to 2010 and the weak increase in Δ<ub> of 0.2% year⁻¹ in summertime from 2010 to 2018. For the wintertime from 2002 to 2010, the trend in flux is −0.08 (mol m⁻² year⁻¹)year⁻¹ of which −0.01 (mol m⁻² year⁻¹)year⁻¹ or 17% can be attributed to changes wind speed. For the weak positive trend in summertime fluxes from 2010 to 2018, the wind speeds contributed about the same (18%) despite the weaker trend Δ<ub> as the absolute trend of efflux was smaller.

The multiannual trends in fluxes follow the same pattern as ΔfCO₂ with an increasing uptake from 2002 to 2010 and a decrease in uptake from 2010 to 2016. There is a reversal to an increase in uptake over the last year of the record (2017). For the whole region the average flux (uptake) is −11.1 ± 8.8 Tg/year from 2002 to 2018 where the uncertainty is the sd of the annual values. The average annual specific fluxes range from of 0.14 mol m⁻² year⁻¹ in 2003, the only year of net outgassing, to −0.40 mol m⁻² year⁻¹ in 2017 (Figure 12a). The total annual fluxes range from a net release of 8.0 Tg C year⁻¹ in 2003 to a strong uptake −22.4 Tg C year⁻¹ in 2017. The differences are primarily caused by differences in uptake during the winter months. In summertime, the region is a CO₂ source and shows less year-to-year variability than wintertime.

Annual anomalies of note are the maxima in uptake in 2013 and 2017 that are caused predominantly by large wintertime uptake. In 2017, there also is reduced summertime evasion (Figure 12a). While high wintertime uptake for 2013 and 2017 are mostly caused by the lower ΔfCO₂ in 2017 higher than average winter time winds contribute to the larger uptake as well. In addition to the effect high wintertime winds on the flux in 2017, low winds in the summer of 2015 significantly decreased the CO₂ evasion (Figures 12a and 12b).

The large changes in annual fluxes over preclude a clear indication that the CO₂ exchange in this marginal sea is appreciably different than the surrounding oceans as suggested in the ocean-dominated margin hypotheses of Dai et al. (2013). The hypothesis is that in waters the Caribbean Sea exchange with the open ocean through the passages and the subsequent vertical mixing causes slightly elevated fCO₂ₘₚₚ and effluxes compared to the Atlantic Ocean basin. This was, in part-based interpretation of the results of Olsen et al. (2004), where Dai et al. (2013) suggest that fCO₂ₘₚₚ are about 5 μmol higher in the Caribbean Sea than in the open ocean. As shown in Figure 12, 2003 had higher ΔfCO₂ and fluxes than seen in the following
14 years. The Takahashi climatology (Takahashi et al., 2002) shows that on average the 14°N to 18°N latitude band in the open ocean is a weak source while the 18°N–26°N band is a net sink. These meridional trends and values are similar to observed in the region of the current study Caribbean Sea.

The means of determining the total CO₂ flux over the area has an impact on the results. All global and regional air-sea CO₂ flux estimates rely on interpolation of sparse data such that the large Caribbean dataset, and gridded data product can be used to provide a view of the differences. To estimate the effect of different temperatures, the average difference between the OISST product and measured temperatures on the ship of 0.25 °C is added the OISST and the mapped fCO₂w was determined with the MLRs in Table 1. This leads to mapped fCO₂w values that are on average 2 μatm higher and the average specific uptake decreases by 25% from −0.20 to −0.15 mol m⁻² year⁻¹. This illustrates that accurate temperature measurements and means to interpolate the fCO₂w and SST from the intake depth to the water surface are important (Woolf et al., 2016).

The fluxes determined using gridded ΔfCO₂ data, and the mapped ΔfCO₂ based on the MLRs yield differences as well. The former only covers part of the region and has temporal gaps. Qualitatively, the two products agree. Both show similar large seasonal variations with the gridded data showing slightly greater amplitudes (Figure 12a). However, the average fluxes for the entire period differ greatly. They are −0.19 ± 0.70 mol m⁻² year⁻¹ for the MLR based mapped product and −0.05 ± 0.70 mol m⁻² year⁻¹ for estimate based on the gridded data. The differences in magnitudes of fluxes are attributed to lack of observations in the winter months, along with the use of observed SST in the gridded observations and OISST in the gridded mapped product. The OISST yields more negative fluxes as described above.

4.6. Relationship With Climate Reorganizations

The changes in fCO₂w and CO₂ fluxes on seasonal to interannual timescales are closely related to SST and MLD. In turn, the regional scale SST anomalies are associated with changes in the physical environment influenced by large-scale climate reorganizations. The climate cycles in the Caribbean are linked, through teleconnections, to the El Niño–Southern Oscillation, ENSO; the Atlantic Multidecadal Oscillation, AMO; and North Atlantic Oscillation, NAO (Birkmark, 2014; Ibánhez et al., 2017; Lefèvre et al., 2013, 2014; Thomas et al., 2008). Since the meteorological and oceanic responses to the climate indices are complex and sometimes lagged, only a broad view of responses of air-sea CO₂ fluxes to these climate patterns are investigated using the seasonal indices that have the greatest impact, for the NAO this is the wintertime (December–February), for the AMO spring (March–May), and for the ENSO the early winter (November–January).

Time series of these seasonal climate indices and annual CO₂ fluxes are shown in Figure 13, each showing significant anomalies in the middle part of the record (2009–2011) where the change in flux trends occurred. The annual fluxes in Figure 14 are plotted at year’s end to reflect the expected lag between the indices and the annual cumulated flux. The overall trend in the NAO is positive for the time period, much like the preceding two decades (Thomas et al., 2008), but it shows a large minimum in 2010 that corresponds to the minimum in CO₂ flux (maximum uptake). The AMO shows a large negative followed by a positive excursion in 2009/2010 similar to the reversals in SST and fCO₂w anomalies, and the ΔpCO₂w (SST) (Figure 11) observed during this time. These changes in annual fluxes in response to the AMO are opposite to the changes observed in the Equatorial Atlantic (Ibánhez et al., 2017). The air-sea CO₂ fluxes show a positive relationship with ENSO with greater uptake during the negative phase of the ENSO (El Niño conditions). This is synergistic with the decreased evasion in the Equatorial Pacific albeit fivefold less in magnitude compared to the El Niño induced depressed outgassing in the Equatorial Pacific.
upwelling region (Feely et al., 2006). The mechanisms for the lower fluxes in the Equatorial Pacific and Caribbean differ as well. The increased uptake in the Caribbean in response El Niño is due to prevailing lower SSTs. In the Equatorial Pacific, the decrease fCO₂ does not change as much as fCO₂ux. Indeed, the midpart of the record show that all indices have large anomalies (Figure 13) corresponding with a minimum in fCO₂ux and fCO₂uw. This suggests that the 3- to 8-year trends are strongly influenced by large single-year anomalies in the climate indices.

The effect of the AMO in forcing of the fluxes is less apparent but the large swing in 2009/2010 corresponds to large changes in the trend of fCO₂uw. Indeed, the midpart of the record show that all indices have large anomalies (Figure 13) corresponding with a minimum in fCO₂uw and largest uptake. However, the multifaceted impacts and monthly variation of the indices along with the large seasonal cycle of fCO₂uw precludes a simple statistical or strong causal relationship for the entire period.

5. Conclusions

Through a corporate, academic and federal partnership the Caribbean Sea has changed from an undersampled area for surface water CO₂ and air-sea CO₂ fluxes to one of the best-observed regions in the world. The high sampling density and robust interpolation methods utilizing the strong correlation of fCO₂uw with SST and other variables make it possible to discern strong decadal trends for the whole area in spite of seasonal variability that is 10 times greater. The region shows an average net uptake of CO₂ of ~11.1 Tg C year⁻¹ for 2002–2018 with annual fluxes ranging from +8.2 (evasion) in 2003 to −22.4 Tg C year⁻¹ in 2017 in contrast to the annual climatological values in the region of 0.4 Tg C year⁻¹ (Takahashi et al., 2009). Our results show that the general view of net outgassing in low latitude seas needs to be reassessed. Moreover, decadal variability of regional air-sea fluxes in oligotrophic marginal seas should be accounted for. In this work, much lower than expected fCO₂uw increases prevailed for 8 years with major transition in patterns and trends occurring around 2010, closely associated with changes in SST trends and MLD that have a direct impact and indirect effect, though biological processes, on fCO₂uw. The fCO₂uw minima corresponds to anomalous low wintertime SSTs and extrema in several major climate indices, most notably the NAO. The fCO₂uw hardly changed from 2002 to 2010 and strongly increased from 2010 to 2018. Over the entire time period the surface ocean fCO₂uw lagged the atmospheric CO₂ increase by 24% causing the uptake to increase over time. Annual fluxes show increasing uptake from 2002 to 2010 and a small decrease in uptake thereafter with a reversal in the last year full of the record (2017) to larger uptakes. The changes in flux are less dramatic that the fCO₂ux trend from 2010 to 2018 as the xCO₂ux and fCO₂ux are both increasing such that the ΔfCO₂ux and thus, the flux does not change as much as fCO₂ux. The trends and pattern are similar for the observations and the interpolated products, but magnitudes vary indicating that quantitatively assessing changes in fCO₂uw and air-sea CO₂ fluxes requires both high density and quality data without temporal gaps, and robust approaches for spatial and temporal interpolation.

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