A 17-year dataset of surface water fugacity of CO_2, along with calculated pH, aragonite saturation state, and air-sea CO_2 fluxes in the Northern Caribbean Sea

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Abstract. A high-quality dataset of surface water partial pressure/fugacity of CO_2 (pCO_{2w}/fCO_{2w})[1], comprised of over a million observations, and derived products are presented for the Northern Caribbean Sea covering the timespan from 2002 through 2018. Prior to installation of automated pCO_2 systems on cruise ships of the Royal Caribbean Cruise Lines and subsidiaries, very limited surface water carbon data were available in this region. With this observational program, the Northern Caribbean Sea has now become one of the best sampled regions for pCO_2 of the world's ocean. The dataset, and derived quantities are binned and averaged on a 1-degree monthly grid and are available at http://accession.nodc.noaa.gov/0207749, DOI:10.25921/2swk-9w56 (Wanninkhof et al., 2019a). The derived quantities include total alkalinity (TA), acidity (pH), aragonite saturation state (Ω_{Ar}) and air-sea CO_2 flux, and cover the region from 15° N to 28° N and 88° W to 62° W. The gridded data and products are used for determination of status and trends of ocean acidification, for quantifying air-sea CO_2 fluxes, and for ground truthing models. Methodologies to derive the TA, pH and Ω_{Ar}, and to calculate the fluxes from fCO_{2w} temperature and salinity are described.

Introduction

Over the past 20 years a rapidly expanding program of measurements of surface water partial pressure of carbon dioxide (pCO_{2w})[1], or the fugacity of CO_2 (fCO_{2w})[1] has provided data to determine air-sea CO_2 fluxes and rates of ocean acidification on local to global scales (e.g. Boutin et al., 2008; Degrandpre et al., 2002; Evans et al. 2015; Schuster et al., 2012; Takahashi et al., 2014; Wanninkhof et al. 2019 a, b). Marginal seas, that historically had a dearth of measurements, have been targeted for increased observations. Through an industry, academic, and federal partnership between the U.S. National Oceanic and Atmospheric Administration (NOAA), Royal Caribbean Cruise Lines (RCCL), and the University of Miami, cruise ships were outfitted with automated surface water pCO_2 systems, also called underway pCO_2 systems (Pierrot et al., 2009). In 2002, the RCCL ship *Explorer of the Seas* (*EoS*) was equipped with an underway pCO_2 setup providing observations on alternating weekly transects from Miami, FL to the Northeastern and Northwestern Caribbean. In 2015, the *EoS* was repositioned out of the Atlantic and the *Celebrity Equinox* (*Eqnx*) was outfitted with an underway pCO_2 system. Additionally, an underway pCO_2 system was installed on the *Allure of the Seas (ALoS)* in 2016. The *Eqnx* and *ALoS* covered similar transects as the *EoS* but on more irregular and seasonal basis. A total of 582 cruises covered the region from 2002 through 2018. A map of all cruise tracks is shown in Figure 1. The number of cruises per year covering the Caribbean Sea and adjacent Western Atlantic are provided in Figure 2. There are fewer cruises in the middle part of the record when the *EoS* was diverted to other routes outside the area, and eventually repositioned.

[1] fCO_2 is the pCO_2 corrected for the non-ideal behavior of CO_2 (Weiss 1974). In surface water $fCO_{2w} \approx 0.997\ pCO_{2w}$

The surface water pCO_2 observational dataset and derived products including total alkalinity (TA), acidity (pH), aragonite saturation state (Ω_{Ar}), and air-sea CO_2 flux are of importance for determining the anthropogenic carbon uptake, and to assess trends and impacts of ocean acidification. The observational data are provided to the global surface ocean carbon atlas (SOCAT) (Bakker et al., 2016) and global CO_2 climatology (Takahashi et al., 2009; 2018). These data are the main source of fCO_2 observations available in the region, and the high frequency of measurements provides a seasonally resolved picture of changing fCO_{2w}. This effort has made the Northern Caribbean one of the few places in the world's ocean where such regional observational density has been established. The data and mapped products are interpreted in Wanninkhof et al. (2019b) who show large decadal changes in trends of surface water fCO_2 and associated changes in air-sea CO_2 fluxes.

The data from the first part of this record form the basis of the Caribbean ocean acidification product suite that maps ocean acidification conditions in the Caribbean (Gledhill et al., 2008; https://www.coral.noaa.gov/accrete/oaps.html; https://cwcgom.aoml.noaa.gov/erddap/griddap/miamiacidification.graph). The large, high quality, and well-resolved dataset is also used to validate models (Gomez et al., 2020).

For optimal application, datasets and associated data products are fully documented here, and are readily accessible according to the findable, accessible, interoperable and reusable (FAIR) principles (Wilkinson et al., 2016). The documentation of the cruises, the sampling methodology, and data reduction techniques are presented in brief. This is followed by a description of the approaches to calculate the different inorganic carbon system parameters. The procedure is to bin and average the fCO_{2w} on a 1˚ by 1˚ grid on monthly scales, referred to as gridded observations. Then the so-called second inorganic carbon system parameter is calculated. We estimate the total alkalinity (TA) based on robust relationships of TA with salinity (Cai et al., 2010; Takahashi et al., 2014; Lee et al., 2006; Millero et al., 1998), and use a software program CO2SYS (Pierrot et al., 2006) to calculate the other inorganic carbon system parameters of interest, in this case pH and Ω_{Ar}. These data products are presented at monthly scales binned and averaged on a 1˚ by 1˚ grid, referred to as gridded products. Annual multi-linear regressions (MLR) are developed between the gridded fCO_{2w} data, and sea surface temperature (SST), sea surface salinity (SSS), location (latitude (Lat) and longitude (Lon)), and mixed layer depth as independent variables. These regressions are applied at monthly and 1˚ by 1˚ spatial resolution to the region between 15˚ N and 28˚ N, and 88˚ W and 62˚ W using remotely sensed or modeled independent variables, and used to calculated air-sea CO_2 fluxes. These calculated parameters are called gridded mapped products. The procedures and datasets, including uncertainty analyses, and tables of column headers of the product created are provided below.

1 Observations

The observational program is described in terms of the ships, voyages, and instrumentation. The ships predominantly sailed in the Caribbean Sea, but also had tracks outside the region, including in the Northeast of the USA, to Bermuda and in the Mediterranean. The description of operations, data, and products presented here cover the Northern Caribbean Sea and Western Atlantic, north of the Caribbean islands chain covering the region from 15˚ N to 28˚ N and -62˚ to -88˚ (= 62˚ W to 88˚ W)

(Fig. 1). The homeports of the ships, where passengers embark and disembark, are Miami and Fort Lauderdale, FL. The *Explorer of the Seas (EoS*) changed homeport from Miami, FL to Cape Liberty Cruise Port, NJ in 2008, and changed its routes at that point to include cruises with Bermuda as a port of call. In 2015 the *EoS* was repositioned to the Pacific and the underway pCO_2 system was removed. From 2015 onward the *Celebrity Equinox (Eqnx)* and from 2016 onward the *Allure of the Seas* (*ALoS)* covered the area. The *Eqnx* spent the summers of 2015 and 2016 in the Mediterranean, causing seasonal data gaps in the Caribbean.

1.2 Cruises

The *EoS* had 331 cruises from 2002 to 2015; the *Eqnx* completed 135 cruises from 2015 through 2018; and the *ALoS* performed 116 cruises in the study area from 2016 through 2018. Temporal coverage over the 17 years shows at least bi-weekly occupations at the beginning of the record from 2002 to 2007, and at the end from 2014 through 2018, with fewer occupations in the years in between (Fig. 2). The cruises lasted between 7 and 14 days, and made about a half a dozen ports of call. The ships generally were in port from early morning to late afternoon, and transited between ports at night, except for long runs (e.g., from Miami to San Juan) when the ship sailed continuously for several days. Ports are listed in the metadata accompanying the original data.

The systems were installed in different locations for the three ships, but each had a dedicated seawater intake near the bow. The *EoS* had an intake in the bow thruster tube (≈3 m depth) that was non-optimal due to bubble entrainment during bow thruster operations and heavy seas. These observations have been culled from the datasets. The *ALoS* and *Eqnx* had their intake at ≈5 m depth, but forward of the bow thruster and had fewer issues with bubble entrainment. On the *EoS,* the underway pCO_2 instrument was initially in a dedicated science laboratory built for purpose on the ship, and located amidships about 100 m from the intake. In 2008, a new system was placed in the engineering space closer to the bow with no apparent change in performance. For the *ALoS* and *Eqnx,* the instruments were near the bow intake in the engineering space, about 5 m from the intake. The *EoS* had an air intake mounted on a mast at the forward most point of the main deck in August of 2008. The *Eqnx* had an air intake near the bow one level below the main deck since the initial installation in March 2015. The underway pCO_2 systems on these ships made marine boundary layer (MBL) air observations (xCO_{2a}) as described in Wanninkhof 2019b but these are not used in the data products presented. Typical cruise speeds were 22 knots which, with sampling every 2.5 minutes, yielded a fCO_{2w} sample approximately every 1.7 km except for 35 minutes every 4.5 hours when four calibration gases, and a CO_2-free reference gas were analyzed, followed by 5 atmospheric CO_2 measurements for the ships with air intakes. This created a gap of 24 km (≈1/4˚) without fCO_{2w} measurements

1.3 Instrumentation

1.3.1 pCO_2 system

The instrumentation is based on a community design described in Pierrot et al. (2009). The instruments were manufactured by General Oceanics Inc. and have performed to high accuracy specifications (Wanninkhof, 2013). The surface water drawn from the intake at about 4 l min^{-1} went through a 1.1 l sprayhead equilibrator with a water volume of 0.5 l and a headspace of 0.6 l. The spray and agitation caused the CO_2 in the headspace to equilibrate with the CO_2 in water with a response time of about 2 minutes (Pierrot et al., 2009; Web et al., 2016). Thus, the air in the headspace reached 99.8 % equilibration in 12 minutes. As the air is recirculated and surface waters were relatively homogeneous on hourly timescales, 100.0 % equilibration was assumed. The four calibration gases supplied by the global monitoring division of the environmental science research laboratory of NOAA (GMD/ ESRL/NOAA) were traceable to the WMO CO_2 mole fraction scale. The CO_2 concentrations of the standards spanned the range of surface water values encountered along the ship tracks (≈ 280- 480 ppm).

The gas entering the analyzer was dried by passing it through a thermo-electric cooler at 5˚ C and a PermaPure drier. The standards did not contain water vapor. The air and equilibrator headspace analyses typically had about 10 % or less humidity. Every 27 hours the CO_2 signal of the LICOR model 6262 infrared analyzer was zeroed with the dry CO_2-free air and spanned with the highest standard. The water vapor channel was zeroed if it showed reading of greater than 0.5 millimol/mol for the dry CO_2-free air. The water vapor channel values are used for a minor correction to the xCO2, and any inaccuracy in the H_2O results in 0.5 ppm change in the analyzer output of dry mole fraction of CO_2 (xCO_2 in parts per million, ppm). xCO_2 as calculated and output by the analyzer, based on measured CO_2 and water vapor levels, were recorded. The seawater circulation in the systems was automatically turned off when the ships entered port, and back-flushed with fresh water removing particles from the inline water filter and thereby alleviating clogging issues in the filter and reducing biofouling of water lines, filter and equilibrator.

Data from the *ALoS* and *Eqnx* were transmitted to shore daily and displayed online in graphical format at https://www.aoml.noaa.gov/ocd/ocdweb/allure/allure_realtime.html and https://www.aoml.noaa.gov/ocd/ocdweb/equinox/equinox_realtime.html, respectively. These updates provided a near-real time opportunity to look at the response of surface water CO_2 to episodic events, such as passage of hurricanes, and yielded timely indications of instrument malfunction that could be remedied by company engineers on board or when the ships returned to port. The instruments have shown agreement to within 1 µatm with other state-of-art systems in intercomparison studies (Nojiri et al., Feb. 2009 pers. com.). 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 (Pierrot et al., 2009; Wanninkhof et al., 2013), were maintained.

1.3.2 Thermosalinograph

Temperature and salinity were measured with a flow-through Seabird SBE45 thermosalinograph (TSG) that was in a seawater flow line parallel to the pCO_2 equilibrator. A SBE38 remote temperature probe was situated near the inlet before the pump, and was used as the SST measurement. The TSGs and temperature probes were maintained by collaborators from the Marine Technical group at the Rosenstiel School of Marine and Atmospheric Sciences at the University of Miami (RSMAS/U. Miami). The TSGs on the ships were factory calibrated on an annual basis. Post-calibrations showed no drift in the temperature sensor but occasionally some drift in the conductivity.

1.3.3 Other instrumentation

The underway effort is part of a larger scientific operation lead by RSMAS called Oceanscope (https://oceanscope.rsmas.miami.edu/). Additional instrumentation onboard the ships include the Marine-Atmospheric Emitted Radiance Interferometer (M-AERI) to assess surface skin temperature retrievals from a number of radiometers on earth-observation satellites (Minnet et al., 2001). The M-AERI's are Fourier-Transform Infrared interferometers situated on the deck viewing the sea surface away from the wake. Acoustic Doppler Current Profilers (ADCP) mounted on the hull of the ships are used to measure ocean currents.

2 Datasets

2.1 pCO_2 data

Full details on data acquisition with these systems and calculation of pCO_2 and fCO_2 can be found in Pierrot et al. (2009). Post-cruise, the xCO_2 data were processed by first linearly interpolating each standard measured every ~4 hours to the time of a sample measurement and then recalculating the air and water xCO_2 values based on the linear regression of the interpolated standard values at the time of sample measurement. For 3 cruises, the analyzer output showed negative water vapor values due to the condition of desiccant chemicals, and thus yielded erroneous dry xCO_2 values. Separate processing routines were developed to correct for these situations (available on request from D. Pierrot). The post-cruise corrected xCO_2 values were used for calculation of pCO_2 and fCO_2 as described in the calculation section.

2.2 Thermosalinograph, sea surface temperature and salinity data

The SST data were obtained from a temperature probe (Seabird, model SBE38) near the intake. The salinity was determined with a thermosalinograph (Seabird, model SBE45), from the measured conductivity and temperature in the unit using the internal software of the SBE45. The SST and SSS data were appended to the pCO_2 data records in real time and also logged via another shipboard computer at more frequent intervals. The SSS data was not quality controlled and no corrections to the

SSS data were made, other than removal of spikes and values that were out of range (<5 and > 40). As salinity has minimal effect on the calculated fCO_2, bad or missing salinities were removed and substituted by linearly interpolated values to eliminate gaps. When SST were not recorded in the CO_2 files, or in error, the SST gaps were filled from the high resolution SST data files maintained by the RSMAS Oceanscope project. On the rare occasion (≈<0.1 %) that SST was not recorded at all, SST data was estimated from the equilibrator temperature data (T_{eq}) after applying a constant offset between T_{eq} and SST using SST data before and after the gap. On average the T_{eq} was 0.12 ± 0.28˚C lower than SST for all the cruises. Per cruise the standard deviation (stdev) of the difference of T_{eq} and SST was on the order of 0.04 ˚C. The in situ pCO_2 data calculated with T_{eq} was flagged with a WOCE quality control flag of 3 which refers to data that is deemed questionable and has a larger uncertainty (https://www.nodc.noaa.gov/woce/woce_v3/wocedata_1/whp/exchange/exchange_format_desc.htm).

For the regionally mapped products on a 1-degree grid and monthly timescale (1˚ by 1˚ by mo), SST and SSS were obtained from the following sources: The SSS data were from a numerical model, the Hybrid Coordinate Ocean Model (HYCOM) (https://HYCOM.org/) and referred to as SSS_{HYCOM}. The SST product for the region is the Optimum Interpolated SST, OISST from https://www.esrl.noaa.gov/psd/., (Reynolds et al., 2007). It uses data from ships, buoys and satellites to generate the fields. For the OISST the reference SST is from buoys, and the other SST data obtained from ships and other platforms were adjusted to the buoy data by subtracting 0.14˚C in the OISST product (Reynolds et al., 2007).

2.3 Wind speed data

Winds were measured on the ships but these data are not used as they are not synoptic for the whole region which is a requirement for the regional flux maps. Instead, wind speeds were obtained from the updated cross-calibrated multi-platform wind product (CCMP-2) (Atlas et al., 2011). The mean scalar neutral wind at 10 m height $\langle u_{10} \rangle$, and its second moment $\langle u_{10}^2 \rangle$ were used to calculate the fluxes. They were determined from the ¼ degree, 6-hourly product that was obtained from Remote Sensing Systems (RSS) (www.remss.com). This product relies heavily on the European Center for Median Weather Forecasting (ECMWF) assimilation scheme that uses in situ and remotely sensed assets, particularly (passive) radiometers on satellites. The directional component uses scatterometer data. The ¼ degree, 6-hourly CCMP-2 product was binned and averaged in 1-degree grid boxes on monthly scales (1˚ by 1˚ by mo). In absence of CCMP-2 data for 2018, the wind product from European Reanalysis, ERA5, was used (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5, Copernicus Climate Change Service, 2017). The ERA5 wind data are at 31-km and 3-hourly resolution but were binned and averaged in the same manner as the CCMP-2 winds. There were no apparent biases between the scalar winds for the two products in the Caribbean.

2.4 Mixed layer depth data

No MLD determinations were made on the cruises and limited observational estimates from other sources are available. The MLDs provided here are from the same numerical model (HYCOM) as used for the mapped SSS, and obtained from http://www.science.oregonstate.edu/ocean.productivity/index.php. The MLDs are based on a density contrast of 0.03 between

surface and subsurface. Mixed layer depths (MLD) are used as an independent variable in the MLRs to map the fCO_{2w} values. They are also needed if mixed layer dissolved inorganic carbon (DIC) inventories are desired, and to determine the effect of mixed layer depth on the changes in fCO_{2w}. As shown in Wanninkhof (2019b, Figure 9), MLDs are negatively correlated with fCO_{2w}. They are provided in the gridded mapped product.

3 Calculations

The calculations of the concentrations and fluxes follow standard procedures as described below. The mapping procedures are detailed. The section on uncertainty includes an example of different possible means of mapping and the effect on the final product.

3.1 Calculation of pCO_2

The starting point in the calculations, which were aided by use of MATLAB routines following the procedures as in Pierrot et al. (2009), were calibrated (dry) xCO_2 values.

The xCO_2 values were converted to pCO_{2eq} (µatm) values:

$$pCO_{2eq} = xCO_{2eq} (P_{eq}\text{-}pH_2O) \quad (1)$$

where eq refers to equilibrator conditions. The P_{eq} is the pressure in the equilibrator headspace and pH_2O is the water vapor pressure calculated according to Eq. 10 in Weiss and Price (1980). The pCO_{2eq} was corrected to surface water values using the intake temperature (SST) and the temperature of water in the equilibrator (T_{eq}) according to the empirical relationship that Takahashi et al. (1993) developed for North Atlantic surface waters:

$$pCO_{2w} = pCO_{2eq}\, e^{(0.0423(SST-Teq))} \quad (2)$$

This empirical correction for temperature is widely used but it is of note that applying the thermodynamic relationships for carbonate dissociation constants yields different temperature dependencies that are a function of temperature. For average SST in the Caribbean of 27.0 ˚C the coefficient of temperature dependence varies from 0.036 to 0.040 using commonly used constants as provided in inorganic carbon system programs such as CO2SYS (Pierrot et al, 2006) compared to the coefficient of 0.0423 (or 4.23 % $˚C^{-1}$) used above. On average the difference between SST and T_{eq} is 0.12 ˚C for all the cruises such that the correction from T_{eq} to SST using the coefficient of 0.0423 in Eq. 2 is 1.9 µatm under average conditions of SST= 27 ˚C and pCO_{2w} =374 µatm. Using a temperature coefficient of 0.036 the temperature correction would be 1.6 µatm, or a 0.3 µatm difference.

3.2 Calculation of fCO_2 in air and water

The fCO_2 is the pCO_2 corrected for non-ideality of CO_2 solubility in water using the virial equation of state (Weiss, 1974). The correction can be expressed as: $fCO_{2a,w} = e^{g(SST,P)}\, pCO_{2a,w}$

and:

$$g(T,P)=[(-1636.75+12.0408T-0.0327957T^2+0.0000316528T^3)+2(1-xCO_2\,10^{-6})^2\,(57.7-0.118T)\,(P/1013.25)]/(82.0575T) \quad (3)$$

where T is in Kelvin, xCO_2 is in ppm, and P is in mbar.

Under average conditions in the Caribbean, the function $e^{g(SST,P)} \approx 0.997$ and fCO_{2w} will be ≈ 1.2 µatm less than pCO_{2w}. As the corrections from partial pressure to fugacity in air and water are approximately the same, the difference between ΔpCO_2 (=pCO_{2w}-pCO_{2a}) and ΔfCO_2 (=fCO_{2w}-fCO_{2a}), that were used to determine the fluxes (Eq. 5), is negligible (≈<0.1 µatm).

3.3 Gridding procedure

Gridding of the observations of fCO_{2w}, SST, and SSS was performed by binning and averaging the data in (1˚ by 1˚ by mo) cells. At typical ship speeds of 22 knots, the ship covered 1˚ in about 2.5 hours taking 60 measurements. This would yield about 250 measurements per month assuming weekly cruises through the area. The actual number of measurements per grid cell ranged from 8 to 500. The higher number of observations per cell were mostly in the latter part of the record, when the *Eqnx* and *ALoS* operated in the same area. The total number of observations from March 2002 through December 2018 was 1.13 million, and the total number of (1˚ by 1˚ by mo) grid cells with observations was 9224.

The gridding facilitated the co-location and merging of products such as MLD_{HYCOM}, SSS_{HYCOM}, OISST, $\langle u^2 \rangle_{CCMP}$, and Marine Boundary Layer (MBL) xCO_2, xCO_{2aMBL}, into the gridded observational dataset for further interpretation. The gridded data aided comparison of *in situ* SST with OISST and SSS with SSS_{HYCOM}. The average difference between SSS and SSS_{HYCOM} for the 2002-2018 data was -0.1 ± 0.28 (n= 9224), and for SST and OISST the difference was 0.25 ± 0.40 ˚C (n= 9224) with the in situ SSS being lower and SST being higher. While both differences include zero within their standard deviation, the temperature difference is in agreement with expected cooler near-surface temperatures that could lead to lower fCO_{2w} which, in turn, have a large impact on the calculated air-sea fluxes.

3.4 Mapping procedures for fCO_{2w} and fluxes

3.4.1 Mapping fCO_{2w} using a multi-linear regression

The gridded observations (1˚ by 1˚ by mo) represented about 10 % of the area and months of investigation from 15 to 28 ˚N and 88 to 62 ˚W over the period of investigation. To interpolate ("map") the data in space (334 grid cells) and time (192 months) a multi-linear regression (MLR) approach was used to determine fCO_{2w} in each grid cell. For each year from 2002 through 2018 the gridded fCO_{2w} observations were regressed against the 1-degree monthly gridded values of position (Lat and Lon), SST, MLD, and SSS. Other permutations of independent parameters were tested but yielded less robust fits (see Appendix A). Annual MLRs were created as fCO_{2w} levels change over time in response to increasing atmospheric CO_2 levels. If fCO_{2w} kept pace with atmospheric CO_2 increase this would translate to a linear trend of fCO_{2w} of 2.13 µatm yr^{-1} over the time period. As shown in Wanninkhof et al. (2019b), multi-year (>4-yr intervals) trends varied from -4 to 4 µatm yr^{-1}, with an average linear trend in fCO_{2w} of 1.4 µatm yr^{-1} from 2002-2018.

The annual MLRs were created of the form:

$$fCO_{2w} = a\ Lon + b\ Lat + c\ OISST + d\ MLD_{Hycom} + e\ SSS_{Hycom} + f \quad (4)$$

with the coefficients for each year, along with the standard error in fCO_{2w} and standard error in each of the coefficients provided in Table 1. The standard error in the fCO_{2wMLR} ranged from 5 to 9 µatm for each year. Using the locations, OISST (the optimal interpolated SST product), MLD_{Hycom} and the SSS_{Hycom} (output of the HYCOM model) the fCO_{2MLR} is determined for each grid cell. There were significant cross-correlations between independent variables such that the effect of the often significant year-to-year differences in coefficients was difficult to interpret. However, the fCO_{2wMLR} from the annual MLRs faithfully reproduced the trends and variability.

The MLRs were produced for each year such that the mapped products could be extended for future years in a straightforward fashion. To determine if there were anomalous discontinuities between subsequent annual MLRs that could impact the timeseries, the difference between fCO_{2w} for subsequent months were plotted versus time in Figure 3. No significant discontinuities were observed between December and January. Only for December and January 2008/2009, 2009/2010, and 2016/2017 there appear to be slight differences in the pattern of monthly progressions but such anomalies are observed during other times of year as well. Using an MLR that includes year as one of the coefficients (see, Appendix A, Eqs. A3 and A4) provides a slightly worse fit. Moreover, using such a fit would necessitate recalculating the mapped products every time a new year is added.

As illustration of differences between gridded and mapped products the results of the fCO_{2wMLR} calculated with the MLRs for 2004, 2011, and 2017 (Table 1) are plotted in Figure 4a along with the gridded observations for the grid cells that span the longitude range from 88˚W to 62˚ W between 23˚ N and 24˚ N (see Fig.1). Figure 4a shows that the mapped product using annual MLRs show the increases in fCO_{2w} over time in the region, and consistent differences in patterns between the East and the West for the three years. The mapped product showed a reasonable correspondence with the gridded observations. Some of the differences between the gridded observations and mapped product were caused by the mismatch between SST and SSS *in situ* with the OISST and SSS_{HYCOM} (Figs. 4b and 4c). In particular, the strong minima in OISST at 79˚ W is not seen in the SST. This is likely because the OISST captured the lower SST near the coast of Cuba. This caused the fCO_{2wMLR} product to be lower as well, as shown in Figure 4a. It illustrates that the mapped fCO_{2wMLR} product is both influenced by the annual MLR, and the gridded MLD_{Hycom}, OISST, and SSS_{Hycom}.

3.4.2 Determining the air-sea CO_2 fluxes for the region

For the determination of the air-sea CO_2 flux (F_{CO2}, mol m^{-2} yr^{-1}), a bulk formulation was applied using the gridded mapped product:

$$F_{CO2} = k\ K_0\ \Delta fCO_2 \quad (5)$$

where ΔfCO_2 is (fCO_{2w}-fCO_{2a}), K_0 is the seawater CO_2 solubility that is a function of temperature and salinity (Weiss and Price, 1980), and k is the gas transfer velocity parameterized as a function of wind speed (Wanninkhof, 2014):

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

where $<u_{10}^2>$ is the monthly 2nd moment of the wind speeds reported in CCMP-2. The 2nd moment accounts for the impact of variability of the wind speed on k. It is determined by taking the monthly average of the sum of squares of the wind speed in CCMP-2 provided at 6-hours and ¼˚ grid resolution. The number of wind speed observation in a (1˚ by 1˚ by mo) grid is 1920. This sample density captures the frequency spectrum of winds in the grid boxes except that extreme wind events such as hurricanes are not fully represented due to the local nature of the extremes and inherent smoothing in the CCMP-2 product. The Sc is the Schmidt number of CO_2 in seawater, defined as the kinematic viscosity of seawater divided by the molecular diffusion coefficient of CO_2. It is determined as a function of temperature from Wanninkhof (2014). At the average temperature of 27 ˚C the Sc_{CO2} equals 475. Over the typical range of SST in the Caribbean from 24 ˚C to 30 ˚C, the $(Sc/660)^{-1/2}$ will vary from 1.1 to 1.27, indicating that the gas transfer velocity will be 27 % higher at an SST of 30 ˚C compared to a SST of 20 ˚C that would correspond to a Sc of 660.

The annual fCO_{2MLR}, and modeled and remote sensed products, OISST, MLD_{HYCOM}, SSS_{HYCOM}, were determined for all (1˚ by 1˚ by mo) grid cells. The monthly fCO_{2aMBL} values were derived from the weekly average xCO_{2aMBL} of the stations on Key Biscayne (KEY) and Ragged Point Barbados (RPB) (CarbonTracker Team, 2019; https://www.esrl.noaa.gov/gmd/ccgg/flask.php). The second moments of the scalar winds, $<u^2>$ from CCMP-2 were averaged on the same grids. As no CCMP-2 product was available in 2018, the ERA5 wind product was used for the last year of the record.

Table 1. Coefficients for the MLR for each year*

3.5 Gridded and mapped products for Alkalinity and pH_T

Total Alkalinity (TA) was determined from salinity. For estimation of TA, several algorithms have been developed with salinity (Fig. 5) (Millero et al., 1998; Lee et al., 2006; Takahashi et al., 2014 and Cai et al., 2010). The relationship of Cai et al. (2010), TA= 57.3 SSS+ 296.4, stdev = 5.5 was used as this was determined from observations that are similar to the conditions in the Caribbean.

The pH_T, the pH on the Total scale at SST, was subsequently determined from the calculated TA and fCO_{2w}. For the gridded products the gridded SSS was used to determined TA. For the pH_T the gridded TA, SST and fCO_{2w}. were used. For the mapped products the gridded OISST and SSS_{Hycom} were applied in the calculations. The pH_T was calculated using the program CO2SYS for Excel V2.2 (Pierrot et al., 2006) with the apparent CO_2 dissociation constants, K_1, K_2 from Lueker et al. (2000); the KSO_4^- dissociation constant from Dickson (1990); the KF dissociation constant from Perez and Fraga (1987); and the total boron relationship with salinity from Uppström (1974).

3.6 Aragonite saturation state (Ω_{Ar})

The aragonite saturation state (Ω_{Ar}) indicates the level of supersaturation or undersaturation of seawater with respect to the mineral aragonite, a polymorph of calcium carbonate, and part of the skeletal structure of many marine calcifiers. When Ω_{Ar} is less than 1 aragonite dissolution is thermodynamically favored and when Ω_{Ar} is greater than 1, it has a tendency to precipitate. The Ω_{Ar} is used as an indicator of ecosystem health with regards to ocean acidification (Mollica et al., 2018). In warm tropical regions surface water saturation states are well above one but no active precipitation takes place except under unusual circumstances in shallow waters, in a precipitation process called whitings (Purkis et al., 2017). The Ω_{Ar} is not measured directly and is defined as the product of calcium and carbonate ion concentrations divided by the solubility product of aragonite:

$$\Omega_{Ar} = [Ca^{2+}]\,[CO_3^{2-}]\,(K_{Ar}'_{sp})^{-1} \quad (7)$$

where $[Ca^{2+}]$ is the total calcium concentration and is derived from salinity: $[Ca^{2+}] = 0.02128 / 40.087 * (SSS/ 1.80655) = 293.84$ S in mol/kg-SW (Riley and Tongudai, 1967). $[CO_3^{2-}]$ is the total carbonate ion concentration determined from two of the inorganic carbon system parameters, and $K_{Ar}'_{sp}$ is the apparent solubility product of aragonite in seawater at a specified salinity, temperature and pressure. In this work $[CO_3^{2-}]$ was determined from the gridded fCO_{2w} and calculated TA using the CO2SYS program. For surface waters, $K_{Ar}'_{sp}$ is

$$pK_{Ar}'_{sp} = -[-171.945 - 0.077993\,T + 2903.293/T + 71.595\log(T) + (-0.068393 + 0.0017276\,T + 88.135/T)\,S^{0.5} - 0.10018\,S + 0.0059415\,S^{1.5}] \quad (8)$$

where $pK_{Ar}'_{sp} = -\log K_{Ar}'_{sp}$, T is temperature in Kelvin (K), and S is salinity (Mucci,1983). As with pH_T, the Ω_{Ar} gridded product was determined from the gridded (1˚ by 1˚ by mo) values of SSS, SST, fCO_{2w} and TA, while the mapped product uses SSS_{Hycom}, OISST, fCO_{2wMLR} and TA.

3.7 Uncertainty of observations, gridded and mapped products

The uncertainty of the products is difficult to quantify due to the many factors, calculations, and interpolations influencing the overall uncertainty. Moreover, the uncertainty estimate includes a random and a systematic component. The latter can have a large influence on interpretations, particularly on the calculated air-sea CO_2 fluxes. Below we address the uncertainty in terms of standard errors in the observations; in the gridded products; and in the mapped products. For the calculated quantities and nomenclature we follow the approach in Orr et al. (2018). The standard uncertainty is characterized by its standard deviation/error of the measured quantities and the standard uncertainties of the input variables. The propagated uncertainty of

a calculated variable is called the combined standard uncertainty. Error propagation for addition is $(e_A^2 + e_B^2 + \ldots..)^{0.5}$ and for multiplication it is $((e_A/A)^2 + (e_B/B)^2 + \ldots.)^{0.5}$, where A and B are the variables, and e_A is the standard error in variable A.

The individual measurements of fCO_{2w} have a combined standard uncertainty of less than 2 µatm based on a propagation of error of instrument response, equilibrator efficiency standardization, and temperatures and pressures at equilibration and at the sea surface (Pierrot et al., 2006). The performance and output data from the $UWpCO_2$ systems have been checked at manufacturer, in intercomparison exercises, and at sea. SST measurements, at point of measurement, are accurate to 0.02 ˚C and SSS to 0.1 based on instrument specifications and annual calibrations.

The combined standard uncertainty in gridded product will vary based on the number of measurements. It includes the actual variability in the 1˚ by 1˚ cells. To estimate the uncertainty per grid cell, the standard deviation of the fCO_{2w} in each cell was determined and then the average of the standard deviations for the 9924 cells with observations was taken. The average standard deviation was 3.4 ± 2.6 µatm (n=9224). The same procedure was followed for SST and SSS and yielded values of 0.22± 0.19 ˚C for SST; and 0.10± 0.10 for SSS. These were relatively small uncertainties compared to the monthly spatial range of ≈ 20 µatm for fCO_{2w}; ≈1 ˚C for SST; ≈1 in SSS. The amplitude of the seasonal cycle of ≈40 µatm for fCO_{2w} and ≈4 ˚C for SST were significantly greater than the standard uncertainties as well.

The calculated parameters for the gridded products, TA, pH_T and Ω_{Ar} have an added uncertainty due to the uncertainty in the constants and parameterizations. The agreement between TA-SSS relationships was good, and choice of TA relationship did not have a determining influence on results. We used TA=57.3 SSS_OBS + 296.4 specifically developed for the Subtropical Western Atlantic (see insert of Figure 10 in Cai et al., 2010). The standard error for the relationship was 5.5 µmol kg^{-1}. The average standard deviation for salinity in the grid cells is 0.1 which translates to an uncertainty gridded TA of 5.7 µmol kg^{-1}. Thus the combined standard uncertainty for TA is $((5.5)^2+(5.7)^2)^{0.5} = 7.9$ µmol kg^{-1}.

pH_T is determined from fCO_{2w} and calculated TA with associated uncertainties. An added uncertainty for the calculated pH_T is the uncertainty in the dissociation constants that are used to calculate pH_T . These uncertainties can be calculated using a modified version of CO2SYS (Orr et al. 2018). As shown in Orr et al. (2018, Figure12), the uncertainty in the constants dominates the calculated pH_T. Using the uncertainty in the constants as presented in the program, an uncertainty of gridded TA of 8 µmol kg^{-1} and 3.4 µatm for fCO_2 yields a combined standard uncertainty in pH of 0.0075. For comparison the uncertainty in pH would be 0.0070 if state-of-the art measurement uncertainties in TA of ±3 µmol kg^{-1} and ±2 µatm for fCO_2 are used. Similarly, the uncertainty Ω_{Ar} using the same approach and uncertainties in independent parameters is ±0.20. Again it is the uncertainty in the dissociation constants that dominate the overall uncertainty.

For the mapped products there is the additional uncertainty due to the use of regressions. The standard errors of the annual regressions in fCO_{2wMLR} and the coefficients of the independent parameters are provided in Table 1. The average standard error for the fCO_{2wMLR} for the 17 years is 6.4 ± 1.2 µatm. Following the approach above for the other independent parameters and propagating these uncertainties yields a combined standard uncertainty of 0.0090 in mapped pH and 0.21 in mapped Ω_{Ar}

Systematic errors are introduced by the different SST and SSS data that are used for the gridded and mapped products, with the former using the gridded measured SST and SSS, and the latter using the SSS_{Hycom} and OISST products. The magnitude of the systematic uncertainty between the mapped and gridded product is estimated from the difference in the gridded and mapped parameters for cells that have both gridded and mapped products (for columns headers, see Table 2). Another possible source of systematic uncertainty is using the gridded and mapped fCO_{2sw}, SSS, and SST to calculate the TA, pH_T and Ω_{Ar} rather than using the in situ values to calculate the parameters and then gridding them. This uncertainty is small based on the low uncertainty of variables in each cell as shown from their standard deviations provided in the gridded products (for column headers, see Table 3).

This is confirmed by comparing this method with the approach of calculating the pH and then gridding and mapping as done in Lauvset et al. (2016) and Jiang et al. (2015). To examine the differences derived from using one or the other approach, both were compared for 2017 data. The pH and TA were calculated for every fCO_{2w} observation and binned into a (1˚ x 1˚ x mo) grid. A MLR from the calculated pH was created for 2017:

$$pH_T(\pm 0.005) = 0.0003194\ Lon - 0.00046744\ Lat - 0.00965183\ SST + 0.00019602\ MLD + 0.00069378\ SSS + 8.3240 \qquad r^2 = 0.89\ (n = 1244) \qquad (9)$$

This MLR was then applied to the independent variables for each grid box to determine $pH_T(MLR)$. This was compared to the approach used here of calculating the pH using the mapped fCO_{2wMLR} and TA-SSS relationships on (1˚ x 1˚ x mo) grids, called $pH_T(fCO_{2w},TA)$. The two approaches provided similar results with $pH_T(fCO_{2w},TA) - pH_T(MLR) = -0.0001 \pm 0.005$ for 2017. The small difference showed a pattern with SST (Fig. 6) but not with the other independent variables. The differences using either $pH_T(fCO_{2w},TA)$ or $pH_{T,MLR}$ are an order of magnitude smaller than the combined standard uncertainties such that the approach of using mapped fCO_{2w} and TA to determine $pH_T(MLR)$ yielded precise and consistent gridded pH_T.

Calculated air-sea CO_2 fluxes have a significant uncertainty as they are driven by relatively small air-water concentration differences. Using the uncertainty in fCO_{2wMLR} of 6.4 µatm and an uncertainty fCO_{2a} of 1 µatm, and uncertainties in k of 20 % (Wanninkhof, 2014) and K_0 of 0.002 (Weiss, 1974), the corresponding combined standard uncertainty is 21 % for the flux. For Flux calculations the systematic error, or bias, is a big issue. The near-surface temperature gradient and skin temperatures will have an impact on the fCO_{2w} and K_0. The magnitude, along with its applicability to the bulk flux formulation is under debate (McGillis and Wanninkhof, 2006; Woolf et al., 2016). For the mapped product, the OISST is used. The OISST uses a variety of temperature data, including remote sensing of the skin temperature and the product is adjusted to buoy temperatures nominally at 1-m (Reynolds et al., 2007) such that implicitly a common reference depth is used for fCO_{2wMLR}. As shown in Wanninkhof et al. (2019b) the OISST was on average 0.25 ˚C lower than SST, and using the SST instead of the OISST would change the flux from -0.87 to -0.63 mol m^{-2} yr^{-1} a 27 % decrease. If OISST is used for bulk temperature and using a canonical value for difference in bulk and skin temperature of 0.17 ˚C, it would increase the uptake of CO_2 by 18 % from -0.87 to -1.04 mol m^{-2} yr^{-1}. Based on current knowledge, we believe that using the OISST and the resulting calculated fCO_{2wMLR} as was done here yields the appropriate fluxes.

Table 2. Estimated uncertainties in measured, gridded, and mapped variables

4 Datasets and data products

Several different data products are provided in conjunction with this paper. The methodology to create the products is described above, and here the file format and column headers is presented with a brief description when warranted.

4.1 Underway pCO_2 data

The quality controlled cruise data are posted at different locations. The individual cruise files with metadata can be found at https://www.aoml.noaa.gov/ocd/ocdweb/occ.html. Data can be found as part of the SOCAT holdings (Bakker et al. 2016) using an interactive graphical user interface https://ferret.pmel.noaa.gov/socat/las/. In addition, cruise files of the three ships are provided in annual directories at the National Center for Environmental Information (NCEI) (https://www.nodc.noaa.gov/ocads/oceans/VOS_Program/explorer.html). The data file structures are from the MATLAB data reduction program of Pierrot (pers. comm.).The primary identifier for the cruises is the EXPO code which is the International Council for the Exploration of the Sea (ICES) ship code and the day the ship starts the cruise. Examples are as follows: For a cruise of the *EoS* starting March 6, 2002, the EXPO code is 33KF20020306; for an *ALoS* cruise starting November 25, 2018, the EXPO code is BHAF20181125, and for the *Eqnx* cruise departing her homeport on February 10, 2018, it is MLCE20180210. The individual cruise files at the sites above sometimes include data outside the study region.

4.2 Gridded data

The gridded datasets are the binned and averaged fCO_{2w}, SST, and SSS observations on a (1˚ by 1˚ by mo) grid. The files include the auxiliary data obtained from remote sensing and interpolated data (OISST), data assimilation of remotely sensed winds (CCMP-2) ,and from the HYCOM model (SSS_{HYCOM}). Calculated TA, pH_T and Ω_{Ar} using procedures outlined above are provided in the file. The calculated fCO_{2w} using the annual MLRs (Table 1) are provided as well. This gridded data set has spatial and temporal gaps as the ships did not transit through each pixel, and coverage is uneven. The number of observations differ for each grid cell and are listed in the gridded data set. For the auxiliary data the number of data points are fixed by the resolution of the data products except where part of the grid includes land which is masked. The column headers are provided in Table 3 and include units and descriptions when warranted.

Table 3. Column headers for the monthly 1-degree gridded observational product (1˚ by 1˚ by mo)

4.3 Mapped product

The mapped product provides the data on a homogeneous (1˚ by 1˚ by mo) grid boxes utilizing the annual MLRs of fCO_{2w} as a function of Lat, Lon, OISST, SSS_{HYCOM}, and MLD_{HYCOM} for the region from 15˚ N to 28˚ N and -62˚ to -88˚ (= 62˚ W to 88˚ W). The modeled and remotely sensed products OISST, SSS_{HYCOM}, and MLD_{HYCOM} and position were used in the MLR (Eq. 4) as the independent parameters. The mapped product includes the air-sea CO_2 fluxes in the region as a specific flux (mol m^{-2} yr^{-1}) for each grid box. The column headers are provided in Table 4 including units and descriptions when warranted.

Table 4. Column headers for the monthly 1-degree mapped product (1˚ by 1˚ by mo) for the whole region

4.4 Monthly and Annual estimates for the Caribbean 2002-2018

Summary files of monthly and annual data and products covering the whole region from 15º N to 28˚ N and 88º W and 62˚ W are provided based on averaging or summing the data in the mapped products. The column headers for the monthly and annual products are similar (Tables 5 and 6). For the monthly files the average of each parameter for each cell are area weighted based on the area of the cell as provided in the mapped product (Table 4) according to: area cell/(total area/#of cells). The total CO_2 mass flux (CO_2_$Flux_{Total}$) is the integral of the monthly area weighted CO_2 fluxes (mol m^{-2} yr^{-1}) expressed in teragrams of carbon (= 10^{12} g C) per month or per year.

Table 5. Column headers for the monthly averaged mapped product

Table 6. Column headers for the annual averaged mapped product

5 Data availability

The observations are available at three locations in slightly different formats but all files are stored by ship and cruise. The primary source is the website at the Atlantic Oceanographic and Meteorological Laboratory (AOML) (https://www.aoml.noaa.gov/ocd/ocdweb/occ.html). These data are submitted to SOCAT at least once a year such that they can be posted in the annual updates of SOCAT (https://socat.info). The permanent depository of the data is at NCEI where the data are stored per cruise in directories listed per year (https://www.nodc.noaa.gov/ocads/oceans/VOS_Program/explorer.html). The gridded observations and mapped products described herein are posted at directories at AOML and NCEI. The dataset, and derived quantities are provided on a 1-degree monthly grid at http://accession.nodc.noaa.gov/0207749, DOI:10.25921/2swk-9w56 (Wanninkhof et al., 2019a). The products cover the years 2002 through 2018 and will be updated annually.

6 Conclusions

The datasets from the cruise ships sailing the Caribbean Sea are a rich resource for studying the trends and patterns of inorganic carbon cycling and ocean acidification in the region. The scales of variability and data density are such that the (1˚ by 1˚ by mo) monthly gridding captures the magnitudes and trends of fCO_{2w} and derived inorganic carbon products on seasonal to interannual scales. Using annual MLRs to interpolate fCO_{2w} with position, SST, SSS, and MLD as independent variables yielded accurate monthly products (Wanninkhof et al., 2019a). A comprehensive investigation of the changes in decadal trends based on the dataset and products was presented in Wanninkhof et al. (2019b). The combined standard uncertainties and systematic offsets of gridding and mapping were estimated from comparing fCO_{2w} observations with gridded and mapped products including TA, pH_T and Ω_{Ar}. The MLRs capture the spatial and temporal variability in fCO_{2w} and calculated pH_T and Ω_{Ar} well in the region. The datasets and products are invaluable for model initiation and validation, and serve as boundary conditions for near-shore fine scale models.

Team list

This work was done on Royal Caribbean Cruise Lines (RCCL) ships who provided access, and personnel and infrastructure resources for the measurement campaign coordinated through the Rosenstiel School of Marine and Atmospheric Sciences (RSMAS) of the University of Miami (U. Miami). Peter Ortner, Elizabeth Williams, Don Cucchiara and Chip Maxwell of the Marine Technical group at RSMAS/U. Miami have been instrumental in maintaining the science operations. Denis Pierrot, Kevin Sullivan, Leticia Barbero, Robert Castle (ret.), and Betty Huss of NOAA/AOML have led the gathering, maintenance, data processing and posting of fCO_2 data. In addition to fCO_2 measurements; skin temperature (MAERI), led by P. Minnet and M. Izaguirre, RSMAS; TSG with instruments, supplied by G. Goni and F. Bringas of AOML; optics; and ADCP, with data processed at the University of Hawaii Currents center (E. Firing and J. Hummon), operations take place on the RCCL ships.

Author contributions

All authors contributed to writing and editing the documents. JT performed most of the gridding and binning, and provided the model and remotely sensed data from the sources listed in the text. KF and DP performed maintenance and data reduction, and liaised with all parties involved in the operations.

Competing interests

The authors of this manuscript have no competing interest involving this work.

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Figure captions

Figure 1. Map with the cruise tracks of the *EoS*, *Eqnx*, and *ALoS* from 2002 though 2018. The grayscales show bathymetry
695 with white being less than 200-m. The green rectangle depicts the region where the data and products are compared in Figure 4.

Figure 2. Histogram of number of cruises per year used in this work.

Figure 3. The difference between fCO_{2wMLR} for subsequent months plotted versus time. The solid squares are the differences between December and January where different MLRs are used.

700 Figure 4. Zonal section of gridded and mapped products between 23˚ N and 24˚ N for January 2004 (Black), January 2011 (Blue), and January 2017 (Red). a. fCO_{2w}; b. SST; c. SSS. The lines with small solid circles are the mapped product, the larger solid circles are the gridded data with standard deviation of data in the box shown as error bars.

Figure 5. Relationships of surface water TA with salinity. The relationship of Cai et al. (2010), with standard error shown as error bars, is used to calculate pH and Ω_{Ar} from fCO_{2w}.

705 Figure 6. pH_T calculated from gridded fCO_{2w} and TA estimated from SSS, $pH_T(fCO_2,TA)$, as done in this work, minus pH_T calculated from observed fCO_{2w} and TA from SSS, $pH_T(MLR)$ plotted against temperature for the 2017 data.

Appendix A

A 17-year dataset of surface water fugacity of CO_2, along with calculated pH, Aragonite saturation state, and air-sea CO_2 fluxes in the Caribbean Sea

Rik Wanninkhof, Denis Pierrot, Kevin Sullivan, Leticia Barbero, and Joaquin Triñanes

Multi-linear regressions (MLRs) for fCO_{2w} were applied to the data on a (1˚ by 1˚ by mo) grid using sequentially fewer independent parameters and the increase in residual was determined. This analysis was performed as two of the independent parameters used, sea surface salinity, SSS and mixed layer depth, MLD, are modeled and their accuracy is not readily known. The functional form for the multi-linear regression, MLR fit is:

$$fCO_{2w,MLR} = a\ Lon + b\ Lat + c\ SST + [d\ MLD] + [e\ SSS] + f \quad (A1)$$

Lon is longitude; Lat is latitude, and SST is sea surface temperature. The MLR with the full number of independent parameters are used in the data products described the main text and the resulting data products are in Wanninkhof et al. (2019a). The terms in brackets indicate the parameters omitted in the estimates here. The coefficients for the MLRs and their standard errors for each year without MLD, and the MLRs without MLD and SSS, are provided in Tables A1 and A2. SST and location are the strongest predictors of fCO_{2w} levels in the region. The increase in the average root mean square of the residual/error (RMSE) excluding MLD and SSS in the annual MLRs is shown in Tables A1 and A2. The average RMSE of the calculated $fCO_{2w,MLR}$ increases by 8 ± 5.5 % by excluding MLD, and increases by 11 ± 5.4 % when MLD and SSS are omitted with the annual differences provided in the last column of Tables A1 and A2.

For the entire record, from 2002-2018, single regressions of fCO_{2w} with position, SST, SSS, and MLD showed larger standard errors and coefficients of determination (r^2) as they do not capture the increase in fCO_{2w} over time due to fCO_{2w} increases in response to increasing atmospheric CO_2 levels. Regressing against ΔfCO_2 which, in principle should not have a trend over time if surface water CO_2 levels keep up with atmospheric CO_2 increases, did not improve the correlation with the independent parameters. This was attributed to the relatively small magnitude of ΔfCO_2 and the observed multi-year changes in trends of fCO_{2w} (Wanninkhof et al., 2019b). However, using the year as a variable in the regression provides a reasonable means of extrapolating over the entire time/space domain with a single regression from 3/2002 through 2/2018:

$$fCO_{2w,MLR}\ (\pm 7.8) = 23.3\ (\pm 2.0) + 1.45\ (\pm 0.02)\ (YR-2002) + 10.23\ (\pm 0.06)\ SST + 1.19\ (\pm 0.03)\ Lat - 0.50\ (\pm 0.01)\ Lon, \quad r^2 = 0.84 \quad (A2)$$

where YR is the integer calendar year. Thus (YR-2002) is the year since the start of the record. The coefficient for the (YR-2002) term of 1.45 reflects the annual increase in surface water fCO_{2w} due to atmospheric fCO_{2a} increase, which averages 2.1 µatm yr^{-1} over the 2002-2018 time period.

The standard error in the $fCO_{2w,MLR}$ of 7.8 µatm in Eq. A2 is generally greater than the standard error in the annual algorithms used to fill the gaps for the annual estimates (Eq. A1) that range from 4.7 to 9.9 µatm depending on the year with an average of 6.4 µatm (Table 1 in main text).

The importance of the different independent variables for the $fCO_{2w,MLR}$ can, in part, be discerned from the standard error in the coefficients but since variables are cross-correlated, other means are investigated such as creating a MLR with either a subset or substitution of variables. Physical parameters were correlated with location (Lat, Lon) in the region. In particular, salinity showed broad correspondence with position. Therefore, substituting SSS for Lat and Lon provided similar magnitude and standard error in the coefficients of the independent variable, but a 10 % greater RMS in the estimated fCO_{2w}:

$$fCO_{2w,MLR}\ (\pm 8.8) = -238\ (\pm 9.8) + 1.36\ (\pm 0.02)\ (YR-2002) + 10.11\ (\pm 0.06)\ SST + 9.07\ (\pm 0.25)\ SSS,\ r^2 = 0.79 \quad (A3)$$

Finally, a simple two parameter linear fit with YR and SST had reasonable predictive capability:

$$fCO_{2w,MLR}\ (\pm 9.4) = 107.2\ (\pm 1.7) + 1.38\ (\pm 0.02)\ (YR-2002) + 9.50\ (\pm 0.06)\ SST \qquad r^2 = 0.77 \qquad (A4)$$

Eq. A4 which used only SST and time showed an increase in the standard error of the derived independent variable fCO_{2w} compared to the other permutations of the MLR. This simple relationship did show some biases with location (not shown), and for gap filling to create uniform monthly fields of fCO_{2w} the full annual regressions (Table 1) using all independent parameters, SST, SSS, MLD and location was the best option.

Table A1. Coefficients for the MLR, $fCO_{2w,MLR} = a\ Lon + b\ Lat + c\ SST + e\ SSS + f$

Table A2. Coefficients for the MLR, $fCO_{2w,MLR} = a\ Lon + b\ Lat + c\ SST + f$

References Appendix A

Wanninkhof, R., Pierrot, D., Sullivan, K.F., Barbero, L., Triñanes, J. A. A 17-year dataset of surface water fugacity of carbon dioxide (fCO_2), along with calculated pH, aragonite saturation state, and air-sea CO_2 fluxes in the Caribbean Sea, Gulf of Mexico and North-East Atlantic Ocean covering the timespan from 2003-03-01 to 2018-12-31, (NCEI Accession 0207749).

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Table 1. Coefficients for the MLR for each year*

	a (LON)	b (LAT)	c (SST)	d (MLD)	e (SSS)	f (Icept)	Standard error $fCO_{2w,MLR}$	r^2	#points
2002	-0.32	0.45	10.29	-0.04	0.77	24.4	4.72	0.90	537
	0.04	*0.11*	*0.18*	*0.02*	*0.72*	*25.8*			
2003	-0.56	0.32	9.24	-0.09	1.11	32.6	5.21	0.86	731
	0.03	*0.09*	*0.17*	*0.02*	*0.66*	*24.6*			
2004	-0.42	0.82	10.34	-0.20	2.78	-55.4	5.21	0.92	740
	0.03	*0.09*	*0.17*	*0.01*	*0.63*	*23.6*			
2005	-0.43	0.49	8.71	-0.07	7.30	-172.0	6.58	0.85	664
	0.04	*0.12*	*0.18*	*0.02*	*0.85*	*32.1*			
2006	-0.31	1.13	9.60	-0.19	2.56	-26.6	4.99	0.89	670
	0.03	*0.08*	*0.16*	*0.02*	*0.70*	*26.1*			
2007	-0.62	1.12	10.56	-0.36	2.75	-77.8	7.04	0.79	483
	0.05	*0.15*	*0.32*	*0.03*	*1.33*	*49.0*			
2008	-0.12	1.12	10.58	-0.30	3.16	-55.8	5.66	0.95	107
	0.43	*0.31*	*0.45*	*0.05*	*1.39*	*59.8*			
2009	-0.66	0.30	7.18	-0.47	0.11	131.9	6.91	0.84	125
	0.22	*0.24*	*0.53*	*0.07*	*1.49*	*58.8*			
2010	-0.53	2.02	8.48	-0.23	2.12	-10.5	9.31	0.85	323
	0.17	*0.26*	*0.41*	*0.05*	*1.48*	*51.7*			
2011	-0.32	0.98	6.65	-0.28	3.06	46.8	7.19	0.76	305
	0.13	*0.19*	*0.34*	*0.04*	*1.51*	*59.7*			
2012	-0.13	1.66	10.81	-0.33	5.56	-154.1	7.29	0.91	358
	0.10	*0.16*	*0.28*	*0.04*	*1.36*	*50.9*			
2013	-0.43	1.30	11.45	-0.47	4.45	-137.5	8.16	0.83	219
	0.18	*0.23*	*0.48*	*0.05*	*1.63*	*55.5*			
2014	-0.60	0.62	9.48	-0.18	3.26	-45.2	7.02	0.78	362
	0.08	*0.15*	*0.29*	*0.03*	*1.06*	*37.7*			
2015	-0.84	0.14	7.76	-0.18	1.20	70.0	6.01	0.82	455
	0.04	*0.11*	*0.27*	*0.02*	*0.90*	*34.5*			

2016	-0.79	0.37	8.72	-0.14	4.17	-62.7	6.66	0.82	1001
	0.03	*0.08*	*0.20*	*0.01*	*0.45*	*17.1*			
2017	-0.42	0.56	9.49	-0.22	5.65	-112.6	6.17	0.87	974
	0.03	*0.08*	*0.15*	*0.02*	*0.58*	*17.1*			
2018	-0.34	-0.09	10.85	-0.13	6.24	-151.2	5.08	0.90	1162
	0.02	*0.07*	*0.13*	*0.01*	*0.39*	*14.7*			

* These annual regressions were used to create the mapped fCO_{2wMLR} fields using the 1˚ by 1˚ by month gridded data product.

fCO_{2wMLR} = a Longitude + b Latitude + c OISST + d MLD_{Hycom} + e SSS_{Hycom} + f

The second row (in italics) for each annual entry is the error of the coefficient.

Table 2. Estimated uncertainties in measured, gridded, and mapped variables

Parameter	Method*	Combined Standard Uncertainty	Systematic Uncertainty**
fCO_{2w}	Measured	2 µatm	
SSS	Measured	0.1 per mil	
SST	Measured	0.02 ˚C	
fCO_{2w}	Gridded	3.4 µatm	
SSS	Gridded	0.10 per mil	
SST	Gridded	0.22 ˚C	
TA	Gridded/Calculated	8 µmol kg^{-1}	
pH_T	Gridded/Calculated	0.0075	
Ω_{Ar}	Gridded/Calculated	0.2	
fCO_{2wMLR}	Mapped/MLR	6.4 µatm	Gridded fCO_{2w}-fCO_{2wMLR}= 1.5 µatm
SSS_{Hycom}	Model	0.2	Gridded SSS-SSS_{Hycom}= -0.1
OISST	Interpolated/remote sensing	0.2 ˚C	Gridded SST- OISST= 0.24˚C
TA	Mapped/Calculated	7.9 µmol kg^{-1}	TA_{SSS}-$TA_{SSSHycom}$=-5.25 µmol kg^{-1}
pH_T	Mapped/Calculated	.009	pH(TA,fCO_{2w}) – pH_T(MLR)= 0.002
Ω_{Ar}	Mapped/Calculated	0.21	0.004
Flux CO_2	Mapped/Calculated	0.18 mol m^{-2} yr^{-1}	$Flux_{SST}$- Flux OISST =0.24 mol m^{-2} yr^{-1}

*Method refers to either the individual data point (measured); the values averaged on a 1˚ by1˚ by mo (gridded); or the interpolated products (mapped).

**The systematic uncertainty is based on the difference between the different methods and products as indicated.

Note, the combined standard uncertainties and systematic uncertainties are based on average conditions

Table 3. Column headers for the monthly 1-degree gridded observational product (1˚ by 1˚ by mo)

Parameter	Unit	Description
Year		
Month		1 (January) through 12 (December)
Latitude (Lat)	Degrees	North is positive. Location is the center point of the grid cell. That is, 15.5˚ N is the grid box spanning 15˚ N to 16˚ N
Longitude (Lon)	Degrees	East is positive. All values in the Caribbean are negative. Location is center point of the grid cell. That is, -87.5 is the grid box spanning 87˚ W to 88˚ W
Area	Km^2	Area of grid box excluding land where appropriate
#_Obs		Number of fCO_{2w} observations in the particular grid box for the particular month
SST_OBS	˚C	Sea surface temperature measured at the intake (average of the grid box)
SST_STDEV	˚C	Standard deviation of SST
SSS_OBS	permil	Sea surface salinity measured by thermosalinograph (average of the grid box)
SSS_STDEV	permil	Standard deviation of SSS
fCO_{2w}_OBS	µatm	Fugacity of CO_2 in seawater (average of the grid box)
fCO_{2w}_STDEV	µatm	Standard deviation of fCO_{2w} observations in the grid box
TA	µmol kg^{-1}	Total alkalinity calculated from a relationship salinity TA = 57.3SSS_OBS+ 296.4 (Cai et al., 2010) using the measured SSS
pH_T		pH on the total scale at SST calculated from fCO_{2w}_OBS and TA using the CO2SYS program of Pierrot et al. (2006) with pH Scale: Total scale (mol/kg-SW) at OISST; CO_2 Constants: K1, K2 from Lueker et al. (2000); KSO_4^- for Dickson (1990); KF from Perez and Fraga (1987) and Total Boron from Uppström (1974)
Ω_{Ar}		Aragonite saturation state calculated using CO2SYS with fCO_{2w}_OBS and TA as input parameters and the same dissociation constants as used for pH_T
OISST	˚C	Optimal interpolated sea surface temperature (Reynolds et al., 2007) for the particular grid box
SSS_{HYCOM}	permil	Sea surface salinity from the HYCOM model
fCO_{2wMLR}	µatm	Fugacity of CO_2 in seawater determined from annual MLRs (see Table 1) with Lat, Lon, SST, OISST, SSS_{HYCOM}, and MLD_{HYCOM}.

800 **Table 4.** Column headers for the monthly 1-degree mapped product (1˚ by 1˚ by mo) for the whole region

Parameter	Unit	Description
Year		
Month		1 (January) through 12 (December)
Latitude (Lat)	Degrees	North is positive. Location is center point of the grid cell. For example, 15.5˚ N is the grid box spanning 15˚ N to 16˚ N
Longitude (Lon)	Degrees	East is positive. All values in Caribbean are negative. Location is center point of the grid cell. For example, -87.5 is the grid box spanning 87˚ W to 88˚ W
Area	Km^2	Area of grid box excluding land where appropriate
OISST	˚C	Optimal interpolated sea surface temperature (Reynolds et al., 2007)
SSS_{HYCOM}	permil	Sea surface salinity Sea surface salinity from HYCOM
MLD_{HYCOM}	m	Mixed layer depth from the HYCOM model
fCO_{2wMLR}	µatm	Fugacity of CO_2 in seawater determined from annual MLRs with Lat, Lon, SST, SSS, and MLD (see Table 1)
fCO_{2a}	µatm	Fugacity of CO_2 in air using the average value between atmospheric sampling station KEY and RBP
ΔfCO_2	µatm	Air water fugacity difference, fCO_{2w} - fCO_{2a}
TA	µmol kg^{-1}	Total alkalinity calculated from a relationship salinity TA = $57.3SSS_{HYCOM}$+ 296.4 (Cai et al., 2010)
pH_T		pH calculated from fCO_{2w} and TA with the CO2SYS program of Pierrot et al. (2006) with pH Scale: Total scale (mol/kg-SW) at OISST; CO_2 Constants: K1, K2 from Lueker et al. (2000); KSO_4^- for Dickson (1990); KF from Perez and Fraga (1987) and Total Boron from Uppström (1974)
Ω_{Ar}		Aragonite saturation state calculated using CO2SYS with fCO_{2wMLR}, TA, OISST, and SSS_{HYCOM} as input parameters with same dissociation constants as used for pH_T
$<u^2>$	m^2 s^{-2}	Second moment of the wind based on ¼˚ 6-h CCMP-2 product (Atlas et al., 2011)
CO_2_Flux	mol m^{-2} yr^{-1}	Monthly air-sea CO_2 flux calculated according to Eqs. 5 and 6, with fCO_{2wMLR} @ OISST

Table 5. Column headers for the monthly averaged mapped product

Parameter	Unit	Description
Year		
Month		1 (January) through 12 (December)
Area	km^2	Total area of Caribbean region excluding land (15˚ N to 28˚ N and 62˚ W to 88˚ W)
OISST	˚C	Optimal interpolated sea surface temperature (Reynolds et al., 2007)
SSS_{HYCOM}	permil	Sea surface salinity Sea surface salinity from HYCOM
MLD_{HYCOM}	m	Mixed layer depth from the HYCOM model
fCO_{2wMLR}	µatm	Fugacity of CO_2 in seawater determined from annual MLRs with Lat, Lon, SST, SSS, and MLD (see Table 1)
fCO_{2a}	µatm	Fugacity of CO_2 in air using the average value between atmospheric sampling station KEY and RBP
ΔfCO_2	µatm	Air-water fugacity difference, fCO_{2w} - fCO_{2a}
TA	µmol kg^{-1}	Total alkalinity calculated from a relationship salinity TA = $57.3SSS_{HYCOM}$+ 296.4 (Cai et al., 2010)
pH_T		pH calculated from fCO_{2w} and TA with the CO2SYS program of Pierrot et al. (2006) with pH Scale: Total scale at OISST; CO_2 Constants: K1, K2 from Lueker et al. (2000); KSO_4^- for Dickson (1990); KF from Perez and Fraga (1987) and Total Boron from Uppström (1974)
Ω_{Ar}		Aragonite saturation state calculated using CO2SYS with fCO_{2wMLR}, TA, OISST, and SSS_{HYCOM} as input parameters with same dissociation constants as used for pH_T
$<u^2>$	m^2 s^{-2}	Second moment of the wind based on ¼˚ 6-h CCMP-2 product (Atlas et al., 2011)
CO_2_Flux	mol m^{-2} mo^{-1}	Monthly air-sea CO_2 flux calculated according to Eqs. 5 and 6
CO_2_$Flux_{Total}$	Tg C mo^{-1}	Total monthly air-sea CO_2 flux calculated according to Eqs. 5 and 6 in Teragram carbon

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Table 6. Column headers for the annual averaged mapped product

Year		
Area	km^2	Total area of Caribbean region, excluding land, from 15˚ N to 28˚ N and 62˚ W to 88˚ W
OISST	˚C	Optimal interpolated sea surface temperature (Reynolds et al., 2007)
SSS_{HYCOM}	permil	Sea surface salinity Sea surface salinity from HYCOM
MLD_{HYCOM}	m	Mixed layer depth from the HYCOM model
Area	Km^2	Area of grid box excluding the surface area of land where appropriate
fCO_{2wMLR}	µatm	Fugacity of CO_2 in seawater determined from annual MLRs with Lat, Lon, SST, SSS, and MLD (see Table 1)
fCO_{2a}	µatm	Fugacity of CO_2 in air using the average value between atmospheric sampling station KEY and RBP
ΔfCO_2	µatm	Air water fugacity difference, fCO_{2w} - fCO_{2a}
TA	µmol kg^{-1}	Total alkalinity calculated from a relationship salinity TA = $57.3SSS_{HYCOM}$+ 296.4 (Cai et al., 2010)
pH_T		pH calculated from fCO_{2w} and TA with the CO2SYS program of Pierrot et al. (2006) with pH Scale: Total scale at OISST; CO_2 Constants: K1, K2 from Lueker et al. (2000); KSO_4^- for Dickson (1990); KF from Perez and Fraga (1987) and Total Boron from Uppström (1974)
Ω_{Ar}		Aragonite saturation state calculated using CO2SYS with fCO_{2wMLR}, TA, OISST, and SSS_{HYCOM} as input parameters with same dissociation constants as used for pH_T
$<u^2>$	m^2 s^{-2}	Second moment of the wind based on ¼˚ 6-h CCMP-2 product (Atlas et al., 2011)
CO_2_Flux	mol m^{-2} yr^{-1}	Annual air-sea CO_2 flux calculated according to Eqs. 5 and 6
CO_2_$Flux_{Total}$	Tg C y^{-1}	Total annual air-sea CO_2 flux calculated according to Eqs. 5 and 6 in Teragram carbon

Table A1. Coefficients for the MLR, $fCO_{2w,MLR}$ = a Lon + b Lat + c SST + e SSS + f

	a (Lon)	b (Lat)	c (SST)	f (Icept)	RMSE $fCO_{2w,MLR}$	r^2	#points	% increase RMSE**
2002	-0.30	0.48	10.42	16.6	4.74	0.90	537	0.4
*	*0.04*	*0.11*	*0.17*	*25.7*				
2003	-0.53	0.35	9.60	26.7	5.32	0.86	731	2.1
	0.03	*0.09*	*0.16*	*25.1*				
2004	-0.35	0.89	11.52	-118.3	5.79	0.90	740	11.1
	0.03	*0.10*	*0.16*	*25.7*				
2005	-0.41	0.53	9.08	-198.3	6.65	0.85	664	1.1
	0.04	*0.12*	*0.15*	*31.8*				
2006	-0.20	1.26	10.23	-98.5	5.48	0.87	670	9.8
	0.03	*0.09*	*0.16*	*27.9*				
2007	-0.50	1.33	12.05	-246.0	7.96	0.74	483	13.1
	0.05	*0.17*	*0.33*	*53.0*				
2008	0.26	1.77	12.50	-138.3	6.43	0.94	107	13.6
	0.49	*0.33*	*0.32*	*65.9*				
2009	-0.38	0.24	9.26	-7.8	7.95	0.79	125	15.1
	0.25	*0.28*	*0.48*	*62.8*				
2010	-0.45	2.43	10.11	-61.4	9.66	0.84	323	3.8
	0.18	*0.26*	*0.25*	*52.6*				
2011	-0.49	1.03	8.11	-157.1	7.83	0.71	305	8.9
	0.14	*0.20*	*0.31*	*58.0*				
2012	-0.19	1.95	12.27	-253.6	8.01	0.89	358	9.9
	0.11	*0.17*	*0.24*	*54.5*				
2013	-0.13	1.89	13.04	-256.5	9.66	0.76	219	18.4
	0.21	*0.27*	*0.53*	*63.9*				
2014	-0.31	0.96	9.89	-20.1	7.40	0.76	362	5.4
	0.07	*0.15*	*0.30*	*39.5*				
2015	-0.86	0.09	7.96	23.8	6.65	0.78	455	10.6
	0.04	*0.12*	*0.30*	*37.8*				

2016	-0.75	0.32	9.57	-121.2	6.95	0.80	1001	4.4
	0.03	*0.09*	*0.18*	*16.6*				
2017	-0.42	0.56	9.49	-112	6.16	0.87	969	0
	0.03	*0.09*	*0.15*	*21.3*				
2018	-0.34	0.009	11.57	-173.2	5.35	0.89	1164	5.3
	0.02	*0.07*	*0.12*	*15.43*				

*The second row (in italics) for each annual entry is the error of the coefficient.

** The increase in root mean square error (RMSE) of $fCO_{2w,MLR}$ compared to Table 1 that includes MLD as an input

815

Table A2*. Coefficients for the MLR, $fCO_{2w,MLR}$ = a Lon + b Lat + c SST + f

	a (Lon)	b (Lat)	c (SST)	f (Icept)	RSME $fCO_{2w,MLR}$	r^2	#points	% increase RMSE**
2002	-0.28	0.53	10.45	47.1	4.74	0.90	537	0.4
***	*0.04*	*0.10*	*0.17*	*5.3*				
2003	-0.54	0.40	9.56	60.9	5.33	0.86	731	2.3
	0.03	*0.09*	*0.16*	*5.2*				
2004	-0.37	1.14	11.30	9.4	5.89	0.90	740	13.1
	0.03	*0.09*	*0.15*	*5.0*				
2005	-0.35	1.08	8.68	84.0	7.05	0.83	664	7.1
	0.04	*0.11*	*0.16*	*5.9*				
2006	-0.18	1.56	10.06	47.8	5.59	0.86	670	12.0
	0.03	*0.07*	*0.16*	*6.4*				
2007	-0.52	1.77	11.87	-31.8	8.10	0.73	483	15.1
	0.05	*0.13*	*0.34*	*12.8*				
2008	0.46	2.17	12.42	11.9	6.50	0.93	107	14.8
	0.49	*0.30*	*0.33*	*28.4*				
2009	-0.42	0.44	9.04	82.0	7.99	0.78	125	15.6
	0.25	*0.24*	*0.46*	*20.3*				
2010	-0.45	2.67	10.11	3.8	9.67	0.84	323	3.9
	0.18	*0.18*	*0.25*	*12.4*				
2011	-0.37	1.65	7.76	101.1	8.09	0.69	305	12.5
	0.15	*0.16*	*0.31*	*14.9*				
2012	-0.18	2.42	11.95	-13.8	8.22	0.88	358	12.8
	0.12	*0.14*	*0.23*	*11.0*				
2013	-0.34	2.39	13.27	-60.7	9.88	0.75	219	21.1
	0.20	*0.22*	*0.53*	*23.5*				
2014	-0.32	1.17	9.90	62.9	7.44	0.75	362	6.0
	0.07	*0.12*	*0.30*	*11.6*				
2015	-0.86	0.22	7.83	100.7	6.68	0.77	455	11.1

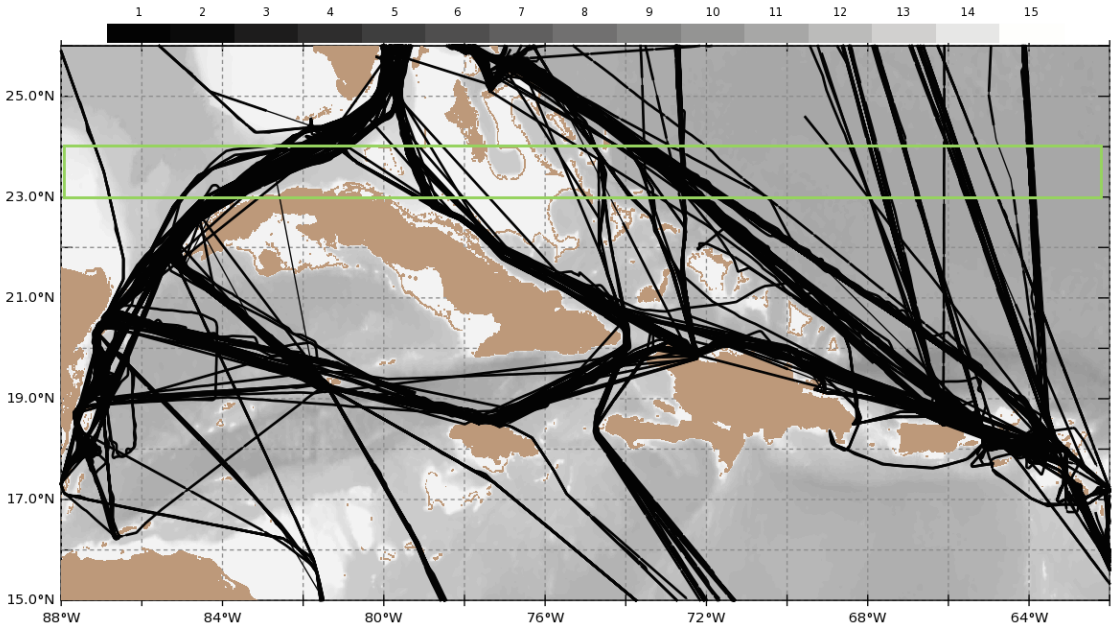
	0.04	*0.11*	*0.29*	*9.7*				
2016	-0.70	0.67	9.84	52.3	7.37	0.77	1001	10.7
	0.04	*0.08*	*0.19*	*6.3*				
2017	-0.40	1.07	10.33	52.8	7.02	0.83	967	13.8
	0.03	*0.08*	*0.16*	*5.32*				
2018	*-0.37*	*0.48*	*11.22*	*44.94*	5.82	0.87	1165	14.6
	0.026	*0.07*	*0.13*	*4.23*				

820 * This table is the same as Table A1 except that MLD in addition to SSS are omitted as independent variables.

** The increase in root mean square error (RMSE) of $fCO_{2w,MLR}$ compared to Table 1 that included MLD and SSS as an independent variable.

*** The second row (in italics) for each annual entry is the error of the coefficient.

825


1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
25.0°N
23.0°N
21.0°N
19.0°N
17.0°N
15.0°N
88°W
84°W
80°W
76°W
72°W
68°W
64°W

830

Fig 1.

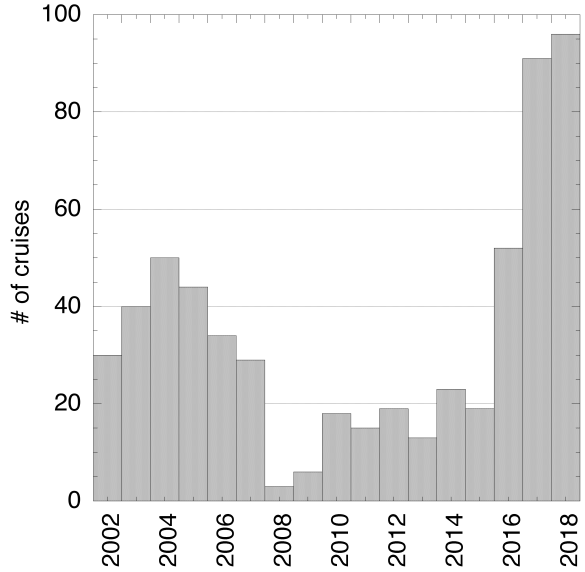

100
80
60
40
20
0
# of cruises
2002
2004
2006
2008
2010
2012
2014
2016
2018

Fig 2.

845

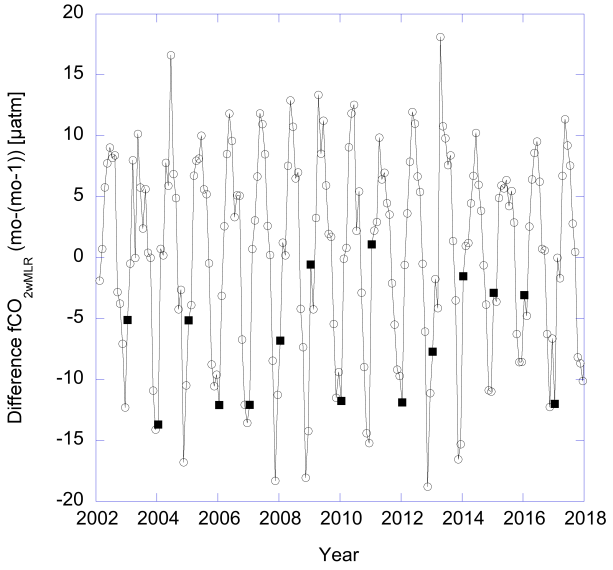
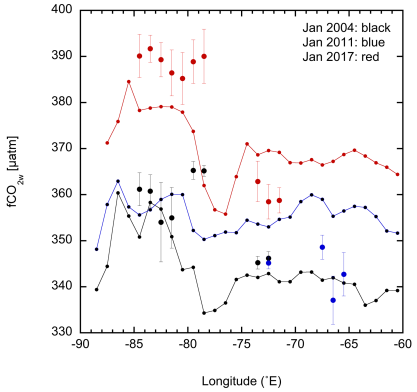

Difference fCO2wMLR (mo-(mo-1)) [µatm]
20
15
10
5
0
-5
-10
-15
-20
2002
2004
2006
2008
2010
2012
2014
2016
2018
Year

Fig. 3



850

Fig. 4a

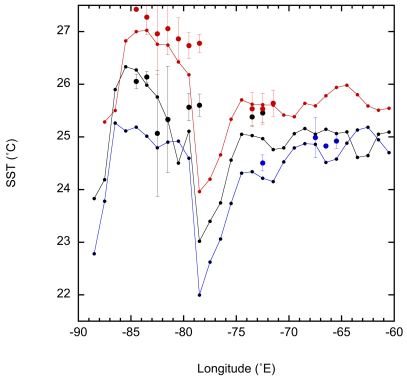


Fig. 4b

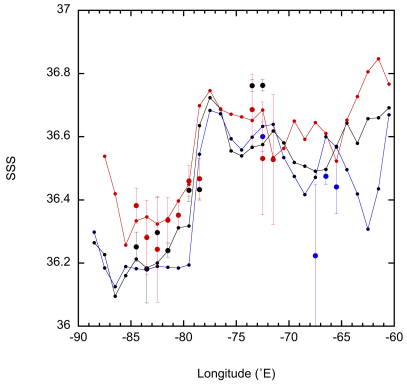


Fig. 4c

855

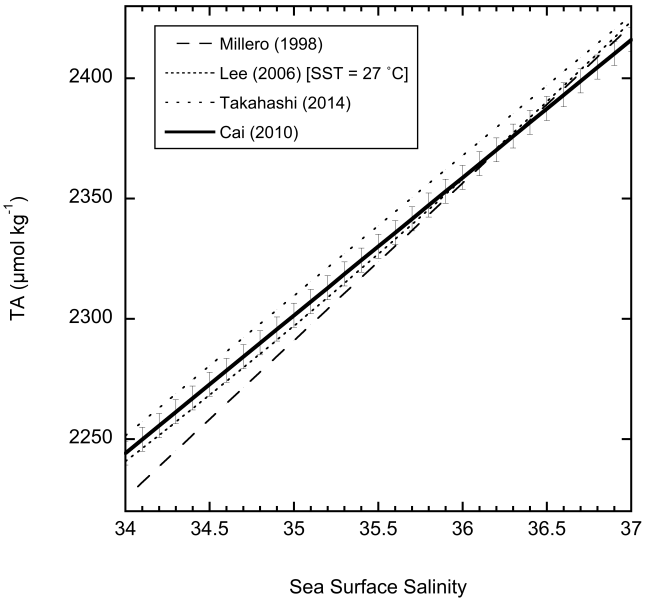
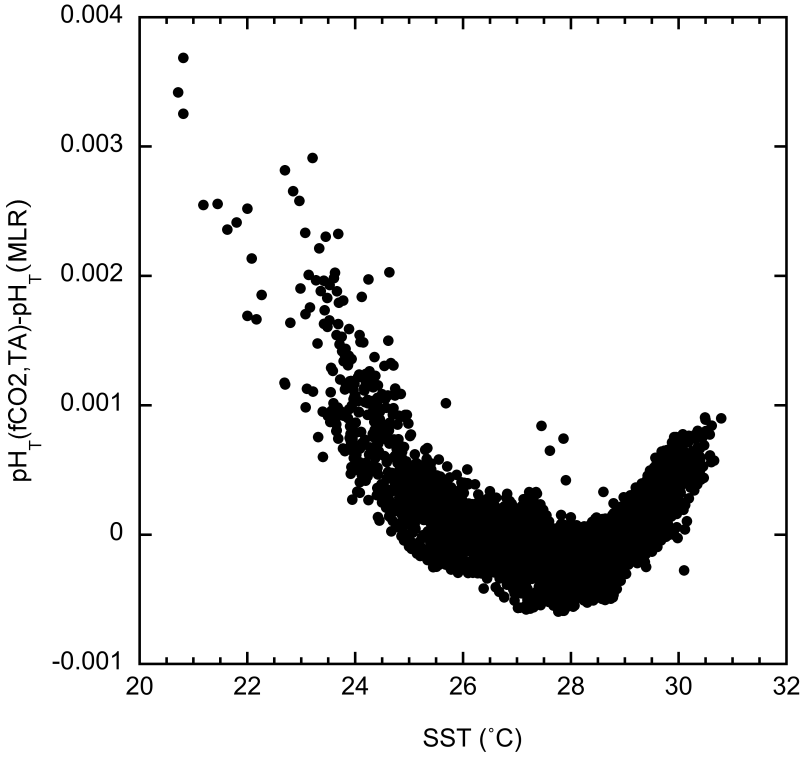

Millero (1998)
Lee (2006) [SST = 27 °C]
Takahashi (2014)
Cai (2010)
TA (μmol kg-1)
2400
2350
2300
2250
34
34.5
35
35.5
36
36.5
37
Sea Surface Salinity

Fig. 5


0.004
0.003
0.002
0.001
0
-0.001
20
22
24
26
28
30
32
pH_T(fCO2,TA)-pH_T(MLR)
SST (˚C)

860 Fig. 6

865