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6 7	Yuan-Yuan Xu <sup>1,2</sup> , Rik Wanninkhof <sup>2</sup> , Emily Osborne <sup>2</sup> , Molly Baringer <sup>2</sup> , Leticia Barbero <sup>1,2</sup> , Wei-Jun Cai <sup>3</sup> , James Hooper <sup>1,2</sup>					
8 9 10	<sup>1</sup> Now at Cooperative Institute for Marine and Atmospheric Studies, University of Miami, Miami, FL <sup>2</sup> NOAA Atlantic Oceanographic and Meteorological Laboratory, Miami, FL <sup>3</sup> School of Marine Science and Policy, University of Delaware, Newark, DE					
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21 22	The following supporting information consists two tables, nine figures, and one reference in support of the statements in the main text.					
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- **Table S1.** List of Western Boundary Time Series cruises used in this study that collected
- 33 hydrographic and oxygen data aboard R/V Walton Smith in the Florida Straits at 27°N. Note that
- the cruises in 2001 and 2003 were excluded due to the lack of usable bottle data for dissolved
- 35 oxygen calibration (NOAA Data Reports; https://doi.org/10.7289/V5/DR-AOML-72;

36 https://doi.org/10.7289/DR-AOML-70).

Cruise Name	Year	Month
ws_2002_03_09	2002	3
ws_2002_06_01	2002	6
ws_2002_08_20	2002	8
ws_2004_01_09	2004	1
ws_2004_05_07	2004	5
ws_2004_06_05	2004	6
ws_2004_08_28	2004	8
ws_2004_12_03	2004	12
ws_2005_06_03	2005	6
ws_2005_07_11	2005	7
ws_2005_11_23	2005	11
ws_2005_12_15	2005	12
ws_2006_01_30	2006	1
ws_2006_06_27	2006	6
ws_2006_09_18	2006	9
ws_2006_12_14	2006	12
ws_2007_06_29	2007	6
ws_2007_10_04	2007	10
ws_2007_12_20	2007	12
ws_2008_07_08	2008	7
ws_2008_12_12	2008	12
ws_2009_02_24	2009	2
ws_2009_06_16	2009	6
ws_2009_09_11	2009	9
ws_2009_11_24	2009	11
ws_2010_02_26	2010	2
ws_2010_05_19	2010	5
ws_2010_08_24	2010	8
ws_2010_12_01	2010	12
ws_2011_03_09	2011	3
ws_2011_06_15	2011	6
ws_2011_09_22	2011	9
ws_2011_12_07	2011	12
ws_2012_05_30	2012	5
ws_2012_07_14	2012	7
ws_2012_10_02	2012	10
ws_2013_06_04	2013	6

ws_2013_08_06	2013	8
ws_2013_10_18	2013	10
ws_2013_12_20	2013	12
ws_2014_04_24	2014	4
ws_2014_07_22	2014	7
ws_2014_09_17	2014	9
ws_2014_12_11	2014	12
ws_2015_01_12	2015	1
ws_2015_04_10	2015	4
ws_2015_05_26	2015	5
ws_2015_07_14	2015	7
ws_2015_09_08	2015	9
ws_2015_11_11	2015	11
ws_2016_03_24	2016	3
ws_2016_07_13	2016	7
ws_2016_09_15	2016	9
ws_2016_12_12	2016	12
ws_2017_02_07	2017	2
ws_2017_06_15	2017	6
ws_2017_07_20	2017	7
ws_2017_10_16	2017	10
ws_2017_12_20	2017	12
ws_2018_04_25	2018	4
ws_2018_06_27	2018	6
ws_2018_09_05	2018	9
ws_2018_10_23	2018	10

- **Table S2.** Coefficients (±standard errors) and statistic information for individual Multiple Linear
- 59 Regression (MLR) fit developed for A05, GOMECC and ECOA cruises.  $DIC = b_0 + b_0$

 $b_1$ ×Temperature +  $b_2$ ×Salinity +  $b_3$ ×DO.

Cruise	Year	bo	<b>b</b> 1	<b>b</b> 2	b3	<b>R</b> <sup>2</sup>
A05	1998	1131.85	-10.4788	35.4297	-0.624679	0.998
		±74.63	±0.4276	±2.1356	±0.054212	
GOMECC-1	2007	1416.36	-8.0810	26.4306	-0.561056	0.998
		±100.48	±0.4754	±2.8498	$\pm 0.048495$	
GOMECC-2	2012	1137.43	-7.8712	35.6629	-0.940353	0.991
		±55.96	±0.2146	±1.6237	±0.049113	
ECOA-1	2015	1173.60	-8.1396	34.0504	-0.744593	0.994
		±63.46	±0.2829	±1.8102	$\pm 0.044886$	
ECOA-2	2018	818.56	-9.8668	43.4810	-0.391386	0.999
		±70.78	±0.2922	±1.9795	±0.041710	



**Figure S1.** (a) The variation pattern of  $\Delta DIC_{ant}$  with depth (y-axis) and longitude (color) in

subsurface water of the Florida Straits at  $27^{\circ}$ N using GOMECC-1 cruise data. (b)  $\Delta$ DIC<sub>ant</sub>

calculated for GOMECC-1 using the eMLR method versus the estimated  $\Delta DIC_{ant}$  using date,

89 longitude, and depth. The black dashed 1:1 line represents where estimation equals observation.

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94 Figure S2. Relationship between salinity and TA in subsurface water of the Florida Straits at

- 95 27°N. The black line represents the best linear fit line for subsurface salinity and TA (TA =
- 96  $48.966(\pm 0.559) \times \text{Salinity} + 600.03(\pm 20.14), n=261, R^2=0.967, p<0.01, RMSE=5.97 \mu mol kg^{-1}).$ 97 The TA values that are associated with salinities less than 35 and fall above the black best linear
- 97 The TA values that are associated with salinities less than 35 and fall above the black best line98 fit line are data from AAIW. The blue line represents the linear regression used for the
- 98 In time are data from AATw. The blue line represents the linear regression used for the 99 estimation of TA in surface waters with the relationship of Cai et al. (2010) where TA =
- $100 57.3 \times \text{Salinity} + 296.4.$



Figure S3. The change of (a) DIC with depth, (b) DO with depth, (c) salinity with depth, and (d) the relationship between density and DIC in subsurface water of the Florida Straits at 27°N and -

79.62°W. 





**Figure S4.** The change of (a) salinity and (b) DIC from 2002 to 2018 in SW of the Florida Straits

110 at 27°N. The black open circles represent the measured salinity and estimated DIC using a nearly

seasonally resolved ship-based time series data, which were sampled at a 10-m vertical depth interval across the transect at 9 evenly distributed stations. The mean values were used to

represent the cross-sectional average for each cruise. The blue dashed lines represent salinity or

DIC trends from 2002 to 2012 while the red dashed lines represent salinity or DIC trends from

- 115 2012 to 2018.







- (f) DIC transport of SW across the Florida Straits at 27°N. The dashed lines represent linear 134 trends for DO and DIC.
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**Figure S6.** Temporal changes in (a) temperature, (b) salinity, (c) DO, (d) DIC, (e) volume

transport, and (f) DIC transport of AAIW across the Florida Straits at 27°N. The dashed lines
represent linear trends for DO and DIC.



Figure S7. The relationships between volume transport (left axis) and right axis: (a) DO in SW,
(b) DIC in SW, (c) DO in NACW, and (d) DIC in NACW in the Florida Straits at 27°N.





Figure S8. ΔDIC<sub>ant</sub> calculated with the eMLR method using bottle data. The black dots represent
bottle data for individual cruises during 1998-2018. The black dashed line represents the best
linear fit for data during 1998-2018. The blue crosses and the blue dashed line represent bottle
data and the best linear fit for the period 2002-2014.



Figure S9. The change of calculated TA from 2002 to 2018 in the surface mixed layer using data
from Wanninkhof et al. (2020). The blue line with crosses represents data for the entire northern
Caribbean Sea while the red line with triangles represents data for the Florida Straits at 27°N.

## **Reference**

- Wanninkhof, R., Pierrot, D., Sullivan, L., Barbero, L., and Triñanes, J. (2020), A 17-year dataset of
  surface water fugacity of CO<sub>2</sub> along with calculated pH, aragonite saturation state and air-sea
  CO<sub>2</sub> fluxes in the northern Caribbean Sea, *Earth System Science Data*, *12*(3), 1489-1509.