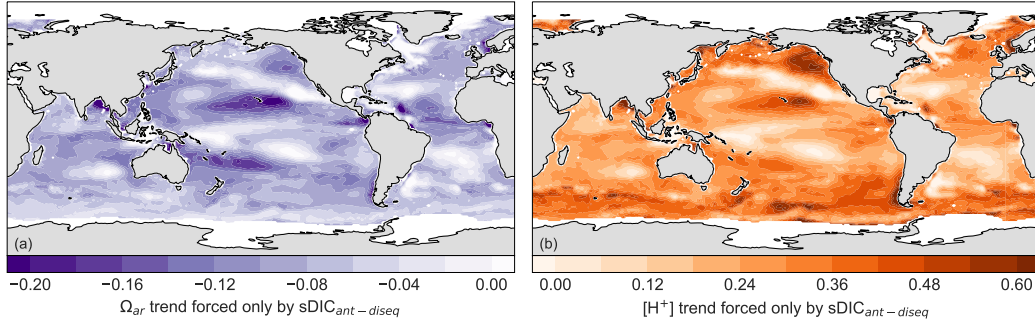


**Figure S1.** Maps of the slopes of salinity normalized DIC (sDIC) for various components. (a) sDIC trends from OceanSODA-ETHZ (Gregor & Gruber, 2021). (b) The disequilibrium term was calculated as the difference between oceanic and atmospheric  $p\text{CO}_2$  for a CESM-ETHZ model run with increasing  $\text{CO}_2$  and constant climate (run C). The  $p\text{CO}_2$  disequilibrium was added to OceanSODA-ETHZ  $p\text{CO}_2$  from which DIC was calculated with *PyCO2SYS*. (c) The expected increase in sDIC if  $p\text{CO}_2$  followed atmospheric  $p\text{CO}_2$  perfectly. (d) The increase in sDIC when accounting for the disequilibrium where uptake slows due to natural outgassing. (e) The slope of the natural sDIC component is calculated as the residual of (a) - (c). (f) The slope of the natural sDIC component including the disequilibrium term calculated as (a) - (d).



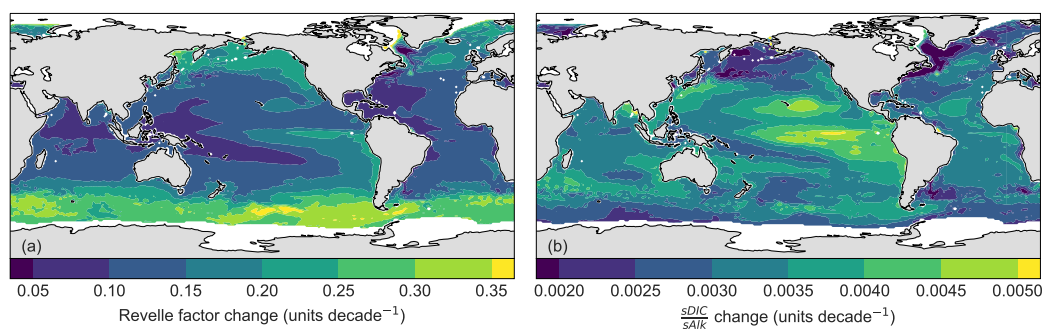
**Figure S2.** Expected increases in (a)  $\Omega_{ar}$  and (b)  $[\text{H}^+]$  due to an increase in sDIC<sub>ant + diseq</sub> (shown in Figure S1d).

**Table S1.** Trends for pH and  $\Omega_{ar}$  (units per decade) from various sources in the literature compared with OceanSODA-ETHZ for the same periods. Note that these trends may not compare well with OceanSODA-ETHZ results due to the different methods used to calculate the trends.

Station	Location	Period	pH (decade <sup>-1</sup> )		$\Omega_{ar}$ (decade <sup>-1</sup> )		Reference
			Trend	Literature	Trend	Literature	
BATS	32.0°N, 64.0°W	1983-2012	-0.014 ± 0.000	-0.017 ± 0.001	-0.066 ± 0.006	-0.095 ± 0.007	(Bates et al., 2014)
		1983-2020	-0.016 ± 0.000	-0.019 ± 0.001	-0.060 ± 0.004	-0.090 ± 0.010	(Bates & Johnson, 2020)
HOT	22.8°N, 158.0°W	1988-2007	-0.014 ± 0.001	-0.019 ± 0.002	-0.095 ± 0.013		(Dore et al., 2009)
		1988-2012	-0.015 ± 0.001	-0.016 ± 0.001	-0.098 ± 0.009	-0.084 ± 0.011	(Bates et al., 2014)
ESTOC	29.0°N, 15.5°W	1995-2012	-0.016 ± 0.001	-0.018 ± 0.002	-0.072 ± 0.014	-0.115 ± 0.023	(Bates et al., 2014)
Iceland Sea	68.0°N, 12.7°W	1983-2012	-0.019 ± 0.001	-0.014 ± 0.005	-0.056 ± 0.006	-0.018 ± 0.027	(Bates et al., 2014)
Irminger Sea	64.3°N, 28.0°W	1983-2012	-0.018 ± 0.000	-0.026 ± 0.006	-0.046 ± 0.007	-0.080 ± 0.040	(Bates et al., 2014)
Munida	45.7°S, 171.5°E	1998-2012	-0.019 ± 0.001	-0.013 ± 0.003	-0.122 ± 0.024	-0.085 ± 0.026	(Bates et al., 2014)
CARIACO	10.5°N, 64.7°W	1995-2012		-0.025 ± 0.004		-0.066 ± 0.028	(Bates et al., 2014)
137°E tropics	5-10°N, 137.0°E	1983-2017	-0.013 ± 0.000	-0.012 ± 0.008	-0.065 ± 0.005	-0.081 ± 0.050	(Ono et al., 2019)
137°E subtropics	20-22°N, 137.0°E	1983-2017	-0.016 ± 0.000	-0.017 ± 0.007	-0.080 ± 0.005	-0.113 ± 0.040	(Ono et al., 2019)
137°E Kuriosho	26-30°N, 137.0°E	1983-2017	-0.017 ± 0.001	-0.019 ± 0.008	-0.090 ± 0.006	-0.121 ± 0.050	(Ono et al., 2019)

**Table S2.** Comparison of trends (units per decade) for long-term observation stations. pH and  $\Omega_{ar}$  were calculated from DIC and Alk for the observation stations and OceanSODA-ETHZ was calculated from  $pCO_2$  and Alk. Uncertainties are the standard error associated with the slope coefficient and do not include other uncertainties associated with measurements or predictions.

Station	Location	Period	pH		$\Omega_{ar}$	
			Observations	OS-ETHZ	Observations	OS-ETHZ
BATS	31.5°N, 64.5°W	1992-2021	-0.0166 $\pm$ 0.001	-0.0174 $\pm$ 0.000	-0.0625 $\pm$ 0.010	-0.0605 $\pm$ 0.005
HOT	22.5°N, 158.5°W	1989-2021	-0.0180 $\pm$ 0.001	-0.0182 $\pm$ 0.001	-0.0888 $\pm$ 0.006	-0.0826 $\pm$ 0.005
ESTOC	29.5°N, 15.5°W	1995-2009	-0.0139 $\pm$ 0.003	-0.0155 $\pm$ 0.001	-0.1375 $\pm$ 0.013	-0.0676 $\pm$ 0.020



**Figure S3.** (a) Map showing the rates of Revelle factor change and (b) map showing the rates of DIC/Alk ratio change.

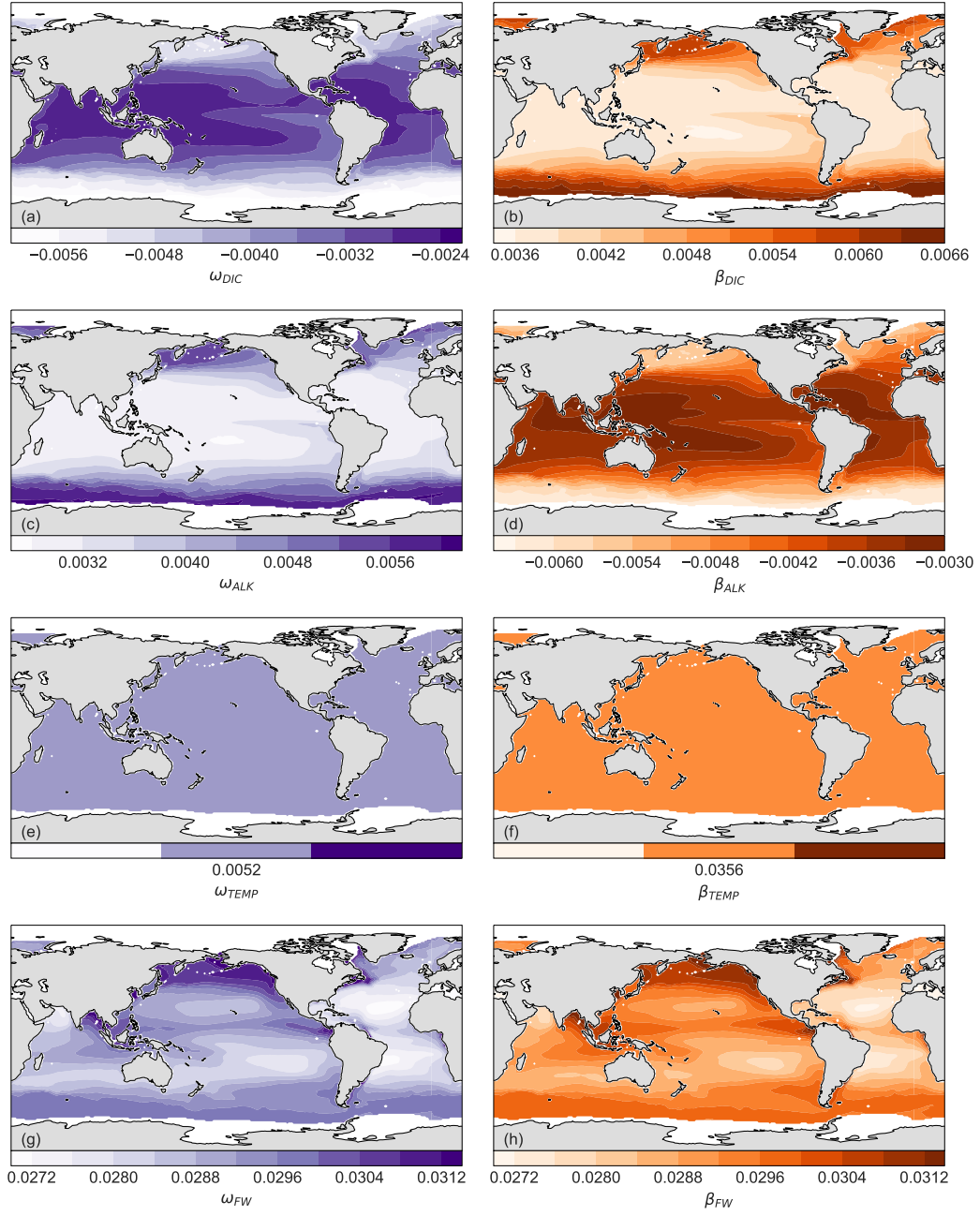


**Table S3.** Slopes of  $\Omega_{ar}$  (units per decade) decomposed into mechanisms (Mech.) for each driver. The regions are biomes from (Fay & McKinley, 2014), where the ice-covered biomes are not presented. We decompose sDIC into anthropogenic (sDIC<sup>ant</sup>) and natural (sDIC<sup>nat</sup>) components, where the latter is the residual of sDIC minus the anthropogenic component. We also show the actual trend for the variable (Trend) and the sum of the decomposed components ( $\Sigma$ ). The mechanisms are taken from Eq 2., where m.e. indicates the mass effect.

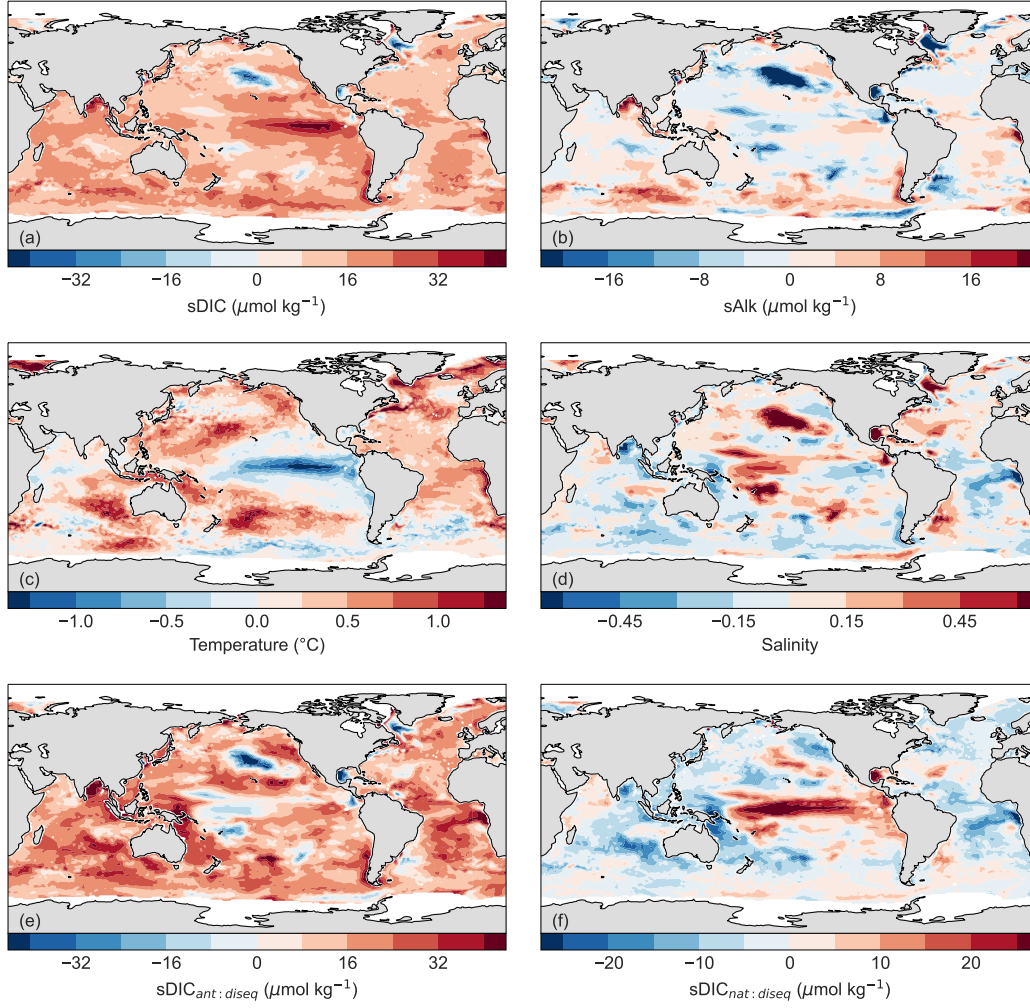
Region	Trend	$\Sigma$	Mech.	sDIC <sup>ant</sup>	sDIC <sup>nat</sup>	sDIC	sAlk	Temp	FW
GLOBAL	-0.0711	-0.0712	$\Delta X$	-0.0869	0.0158	-0.0711	-0.0051	0.0028	0.0020
			$\omega_X$	-0.0039	0.0005	-0.0034	0.0000	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0044	-0.0006	0.0038	-0.0001	-0.0001	0.0000
NP-SPSS	-0.0479	-0.0491	$\Delta X$	-0.0736	0.0269	-0.0467	-0.0054	0.0022	0.0005
			$\omega_X$	-0.0025	0.0005	-0.0019	-0.0003	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0032	-0.0008	0.0024	0.0002	-0.0001	-0.0000
NP-STSS	-0.0689	-0.0697	$\Delta X$	-0.0844	0.0243	-0.0601	-0.0167	0.0040	0.0032
			$\omega_X$	-0.0028	0.0006	-0.0022	-0.0012	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0031	-0.0007	0.0024	0.0012	-0.0001	-0.0002
NP-STPS	-0.0809	-0.0818	$\Delta X$	-0.1074	0.0272	-0.0801	-0.0055	0.0036	0.0003
			$\omega_X$	-0.0033	0.0003	-0.0030	-0.0007	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0035	-0.0004	0.0032	0.0007	-0.0001	-0.0002
PEQU-W	-0.0647	-0.0652	$\Delta X$	-0.0740	0.0092	-0.0648	-0.0115	0.0045	0.0069
			$\omega_X$	-0.0035	0.0006	-0.0029	0.0000	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0033	-0.0006	0.0027	-0.0000	-0.0001	-0.0000
PEQU-E	-0.0981	-0.0975	$\Delta X$	-0.1074	0.0019	-0.1055	0.0079	0.0012	-0.0024
			$\omega_X$	-0.0038	-0.0035	-0.0073	-0.0002	0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0045	0.0038	0.0083	0.0002	0.0004	-0.0001
SP-STPS	-0.0795	-0.0796	$\Delta X$	-0.0978	0.0180	-0.0798	-0.0039	0.0026	0.0014
			$\omega_X$	-0.0038	0.0003	-0.0035	-0.0002	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0042	-0.0004	0.0038	0.0002	-0.0001	-0.0001
NA-SPSS	-0.0452	-0.0457	$\Delta X$	-0.0635	0.0261	-0.0374	-0.0139	0.0032	0.0023
			$\omega_X$	-0.0027	0.0008	-0.0019	-0.0002	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0034	-0.0010	0.0024	-0.0001	-0.0001	-0.0000
NA-STSS	-0.065	-0.0652	$\Delta X$	-0.0873	0.0229	-0.0644	-0.0078	0.0037	0.0035
			$\omega_X$	-0.0033	0.0009	-0.0024	-0.0002	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0034	-0.0009	0.0025	0.0002	-0.0002	-0.0001
NA-STPS	-0.0676	-0.0673	$\Delta X$	-0.0842	0.0103	-0.0739	-0.0033	0.0044	0.0057
			$\omega_X$	-0.0034	0.0007	-0.0027	0.0000	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0034	-0.0007	0.0027	-0.0000	-0.0001	-0.0001
AEQU	-0.0774	-0.0783	$\Delta X$	-0.1254	0.0393	-0.0862	0.0074	0.0039	-0.0036
			$\omega_X$	-0.0055	0.0023	-0.0032	0.0006	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0059	-0.0024	0.0034	-0.0007	-0.0002	0.0003
SA-STPS	-0.0719	-0.071	$\Delta X$	-0.0621	-0.0018	-0.0640	-0.0172	0.0025	0.0076
			$\omega_X$	-0.0049	0.0015	-0.0034	0.0003	-0.0000	0.0000
			$\Omega_{ar}$ m.e.	0.0049	-0.0015	0.0034	-0.0003	-0.0001	0.0002
IND-STPS	-0.0734	-0.0737	$\Delta X$	-0.0940	0.0205	-0.0735	-0.0064	0.0035	0.0026
			$\omega_X$	-0.0047	0.0013	-0.0033	0.0002	-0.0000	0.0000
			$\Omega_{ar}$ m.e.	0.0048	-0.0013	0.0034	-0.0002	-0.0001	0.0001
SO-STSS	-0.0667	-0.0664	$\Delta X$	-0.0771	0.0155	-0.0616	-0.0095	0.0021	0.0021
			$\omega_X$	-0.0047	0.0008	-0.0039	0.0004	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0055	-0.0009	0.0046	-0.0006	-0.0001	0.0001
SO-SPSS	-0.0616	-0.0605	$\Delta X$	-0.0707	0.0033	-0.0674	0.0058	0.0004	-0.0006
			$\omega_X$	-0.0041	0.0002	-0.0040	0.0008	-0.0000	-0.0000
			$\Omega_{ar}$ m.e.	0.0058	-0.0002	0.0055	-0.0011	-0.0000	0.0001

**Table S4.** The same as Table S3 but for  $[\text{H}^+]$  in  $\text{nmol} \cdot \text{kg}^{-1} \cdot \text{decade}^{-1}$ .

Region	Trend	$\Sigma$	Mech.	sDIC <sup>ant</sup>	sDIC <sup>nat</sup>	sDIC	sAlk	Temp	FW
GLOBAL	0.2502	0.2853	$\Delta X$	0.2510	-0.0438	0.2073	0.0106	0.0381	0.0037
			$\beta_X$	0.0083	-0.0010	0.0073	-0.0003	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0195	-0.0025	0.0170	-0.0003	0.0019	0.0000
NP-SPSS	0.2777	0.314	$\Delta X$	0.3274	-0.1165	0.2109	0.0171	0.0561	0.0012
			$\beta_X$	0.0085	-0.0020	0.0066	0.0009	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0201	-0.0045	0.0156	0.0021	0.0033	0.0003
NP-STSS	0.2575	0.3006	$\Delta X$	0.2333	-0.0695	0.1638	0.0418	0.0601	0.0071
			$\beta_X$	0.0054	-0.0012	0.0042	0.0026	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0130	-0.0026	0.0104	0.0061	0.0036	0.0010
NP-STPS	0.2392	0.274	$\Delta X$	0.2543	-0.0634	0.1910	0.0135	0.0449	0.0011
			$\beta_X$	0.0050	-0.0004	0.0045	0.0013	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0126	-0.0011	0.0115	0.0029	0.0026	0.0008
PEQU-W	0.2124	0.2477	$\Delta X$	0.1673	-0.0208	0.1464	0.0217	0.0512	0.0114
			$\beta_X$	0.0049	-0.0008	0.0040	-0.0000	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0145	-0.0026	0.0119	-0.0001	0.0012	0.0001
PEQU-E	0.2852	0.3278	$\Delta X$	0.3000	-0.0031	0.2969	-0.0188	0.0159	-0.0055
			$\beta_X$	0.0080	0.0064	0.0144	0.0002	0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0169	0.0144	0.0313	0.0006	-0.0076	0.0004
SP-STPS	0.2427	0.2744	$\Delta X$	0.2540	-0.0460	0.2080	0.0063	0.0336	0.0021
			$\beta_X$	0.0070	-0.0006	0.0064	0.0002	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0164	-0.0016	0.0148	0.0006	0.0021	0.0003
NA-SPSS	0.2604	0.2973	$\Delta X$	0.2321	-0.0992	0.1329	0.0545	0.0738	0.0085
			$\beta_X$	0.0077	-0.0024	0.0053	0.0006	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0185	-0.0064	0.0121	0.0030	0.0062	0.0005
NA-STSS	0.229	0.2634	$\Delta X$	0.2281	-0.0602	0.1678	0.0172	0.0500	0.0067
			$\beta_X$	0.0059	-0.0016	0.0043	0.0005	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0151	-0.0042	0.0109	0.0011	0.0045	0.0004
NA-STPS	0.2155	0.2488	$\Delta X$	0.1870	-0.0229	0.1641	0.0064	0.0506	0.0095
			$\beta_X$	0.0048	-0.0009	0.0039	-0.0000	-0.0000	-0.0001
			$[\text{H}^+]$ m.e.	0.0137	-0.0025	0.0112	0.0000	0.0029	0.0003
AEQU	0.2187	0.2457	$\Delta X$	0.2923	-0.0906	0.2017	-0.0143	0.0470	-0.0062
			$\beta_X$	0.0082	-0.0034	0.0048	-0.0010	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0222	-0.0092	0.0129	-0.0021	0.0039	-0.0010
SA-STPS	0.2241	0.2563	$\Delta X$	0.1496	0.0041	0.1537	0.0366	0.0316	0.0141
			$\beta_X$	0.0080	-0.0024	0.0056	-0.0006	-0.0000	0.0000
			$[\text{H}^+]$ m.e.	0.0199	-0.0060	0.0139	-0.0011	0.0030	-0.0005
IND-STPS	0.2208	0.2527	$\Delta X$	0.2196	-0.0474	0.1723	0.0133	0.0426	0.0049
			$\beta_X$	0.0071	-0.0021	0.0051	-0.0003	-0.0000	0.0000
			$[\text{H}^+]$ m.e.	0.0191	-0.0055	0.0136	-0.0007	0.0023	-0.0004
SO-STSS	0.2654	0.303	$\Delta X$	0.2559	-0.0500	0.2060	0.0244	0.0380	0.0050
			$\beta_X$	0.0119	-0.0019	0.0100	-0.0014	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0258	-0.0041	0.0217	-0.0024	0.0021	-0.0004
SO-SPSS	0.325	0.3656	$\Delta X$	0.3584	-0.0165	0.3419	-0.0256	0.0121	-0.0023
			$\beta_X$	0.0168	-0.0007	0.0160	-0.0031	-0.0000	-0.0000
			$[\text{H}^+]$ m.e.	0.0345	-0.0018	0.0327	-0.0058	0.0004	-0.0007



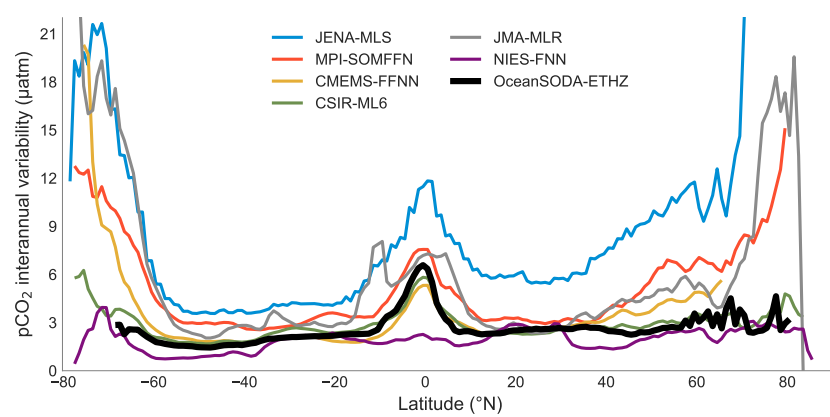
**Figure S4.** Maps depicting the average sensitivities of  $\Omega$  (left) and  $[H^+]$  (right) to changes in sDIC (a,b), sAlk (c,d), temperature (e,f), and freshwater input (g,h) in the period 1982-2020.



**Figure S5.** Maps showing the mean change ( $\Delta X$  in Eq 2 and 3) in (a) sDIC, (b) sAlk, (c) temperature, (d) freshwater input, (e)  $sDIC_{ant:diseq}$ , and (f)  $sDIC_{nat:diseq}$  from 1982 to 2021. Note that the mean change is calculated with  $\Delta X = \sum_{i=1}^N (X_i - X_0)/N$  where  $X$  is the driver,  $i$  is the time step (years),  $X_0$  is the first time step in  $X$  and  $N$  is the total number of years (40).

**Table S5.** Comparison of interannual variability among seven pCO<sub>2</sub> products from the lowest to highest relative to OceanSODA-ETHZ: OceanSODA-ETHZ (Gregor & Gruber, 2021), CSIR-ML6 version 2019a (Gregor et al., 2019), CMEMS-FFNN (Denvil-Sommer et al., 2019), NIES-FNN (Zeng et al., 2015), MPI-SOMFFN (Landschützer et al., 2016), JMA-MLR (Iida et al., 2021), and Jena-MLS (Rödenbeck et al., 2013). Interannual variability in other data sets is scaled proportionally to that of OceanSODA-ETHZ, which is set to 1 by default.

Product	Scaled interannual variability
OceanSODA-ETHZ	1.00
CSIR-ML6 version 2019a	1.10
CMEMS-FFNN	1.31
NIES-FNN	1.56
MPI-SOMFFN	2.14
JMA-MLR	2.36
Jena-MLS	3.21



**Figure S6.** Comparison of zonally averaged interannual variability of pCO<sub>2</sub> for the seven pCO<sub>2</sub> products shown in Table S5.

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