Sensitivity coefficients widely used for the decomposition of seas surface pCO_2 in the open ocean (Sarmiento and Gruber, 2006; Takahashi et al., 1993) :

$$\frac{\partial pCO_2}{\partial DIC} = \frac{\overline{pCO_2}}{\overline{DIC}} \frac{3\times\overline{ALK}\times\overline{DIC}-2\times\overline{DIC}^2}{(2\times\overline{DIC}-\overline{ALK})(\overline{ALK}-\overline{DIC})} \quad \textbf{(S1)}$$

$$\frac{\partial pCO_2}{\partial ALK} = -\frac{\overline{pCO_2}}{\overline{ALK}} \frac{\overline{ALK}^2}{(2\times\overline{DIC}-\overline{ALK})(\overline{ALK}-\overline{DIC})} \quad \textbf{(S2)}$$

$$\frac{\partial pCO_2}{\partial SST} = \overline{pCO_2} \times 0.0423 \quad \textbf{(S3)}$$

$$\frac{\partial pCO_2}{\partial SSS} = \frac{\overline{pCO_2}}{\overline{SSS}} \quad \textbf{(S4)}$$

, where the overbar represents the long-term mean value in the period of 1998-2015 at each point in space.

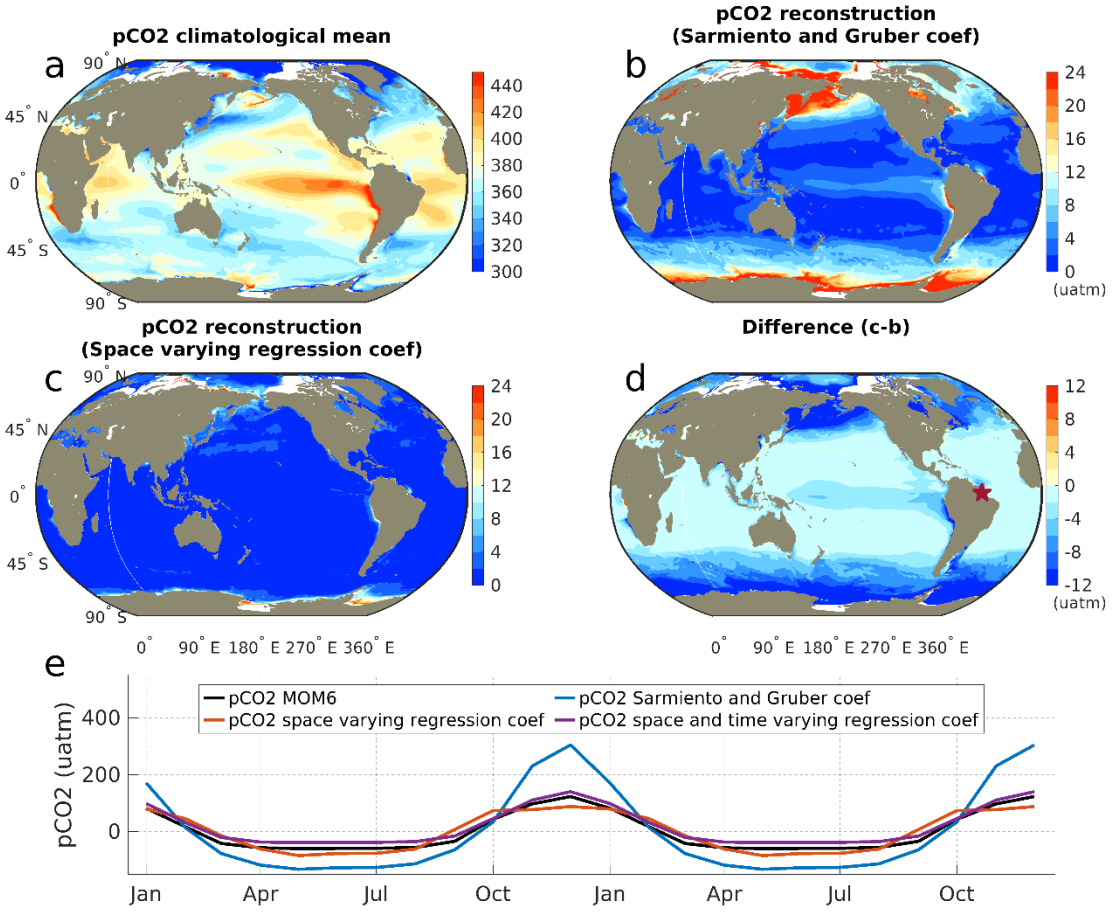

Figure S1: Evaluation of ocean pCO_2 reconstruction methods using the same SST, SSS, DIC and ALK fields but different methods to derive the pCO_2 sensitivity coefficients ($\frac{\partial pCO_2}{\partial DIC}$, $\frac{\partial pCO_2}{\partial ALK}$, $\frac{\partial pCO_2}{\partial SST}$ and $\frac{\partial pCO_2}{\partial SSS}$): (a) ocean pCO_2 simulated by MOM6-COBALT, (b) bias in reconstructed pCO_2 using the approach widely used in the open ocean to compute sensitivity coefficients (Sarmiento and Gruber, 2006; Takahashi et al., 1993) and (c) bias in reconstructed pCO_2 using the regression-based approach developed in this study to compute space varying sensitivity coefficients (using the CO2SYS program, see section 2.3 for details). (d) The difference in bias between the traditional and regression-based approaches shows a strong reduction in biases when using the regression-based method. Biases (in µatm) are quantified using the root mean square error (RMSE) between the pCO_2 simulated by the model and the pCO_2 reconstructed from simulated monthly SST, DIC, ALK and SSS (Eq. 2). (e) time series of seasonal pCO_2 anomaly at 310.25°E, 1°N (star on panel d) simulated by the MOM6 model (black), and reconstructed using the space varying coefficients of Sarmiento and Gruber 2006 (blue), using the space varying regression-based coefficients used in this study (red), and space and time varying regression-based coefficients (purple). See text in method section for further details.

Table S1: Spatial annual mean evaluation of MOM6-COBALT against observational data (SST, SSS, nutrients and pCO_2) for each MARCATS. A positive bias indicates higher values simulated by MOM6-COBALT. The observational SST and SSS fields are from the NOAA OI SST V2 (Reynolds et al., 2007) and the EN4 SSS (Good et al., 2013). The observational nutrients derived from the World Ocean Atlas version 2018 (Garcia et al., 2019). The bias between the pCO_2 simulated by MOM6-COBALT is also evaluated against SOCATv6 and the coastal-SOM-FFN. Absolute pCO_2 biases larger than 20 µatm are highlighted in red. Blank cells values represent regions for which no observation are available in the SOCATv6 database. The percentage of the MARCATS surface covered by SOCATv6 observations is also presented.

MARCATS number (Mx)	MARCATS name	MARCATS category	Evaluation against data (bias)							Evaluation against Coastal-SOM-FFN (bias)	Surface covered by SOCATv6 (%)
			SST (°C)	SSS (-)	Nutrients (µmol kg^{-1})			pCO_2 (µatm)		pCO_2 (µatm)	
					NO_3	PO_4	SiO_4	MOM6-COBALT vs SOCATv6	Coastal-SOM-FFN vs SOCATv6		
2	Californian Current	EBC	0.2	0.0	0.3	-0.2	2.5	20.7	-12.7	34.5	64
4	Peruvian upwelling Current	EBC	-0.2	-0.1	8.1	0.2	0.9	98.5	-17.7	106.4	18
19	Iberian upwelling	EBC	0.4	0.4	-0.5	-0.1	-0.2	5.8	-8.1	9.3	72
22	Moroccan upwelling	EBC	0.5	0.2	-0.5	-0.1	-0.4	4.4	-7.1	10.2	60
24	SW Africa	EBC	0.7	0.1	2.5	0.0	-0.4	10.2	-2.7	79.3	33
33	Leeuwin Current	EBC	-0.3	0.2	-0.2	-0.1	0.8	12.1	5.6	4.2	51
27	W Arabian Sea	Indian margins	0.3	0.4	-1.6	-0.4	-1.5	14.7	5.7	11.6	3
30	E Arabian Sea	Indian margins	-0.1	0.4	0.0	-0.3	-0.9			-8.3	0
31	Bay of Bengal	Indian margins	0.0	-0.6	3.4	-0.2	-1.2			-24.1	0
32	Tropical E Indian	Indian margins	0.1	-0.1	-0.1	-0.1	-0.5	-7.7	-7.1	0.3	8
9	Gulf of Mexico	Marginal sea	0.4	0.3	0.6	-0.2	-0.4	-2.0	3.8	-9.1	68
12	Hudson Bay	Marginal sea	-0.4	0.7	-1.2	-0.4	2.9			5.7	0
18	Baltic Sea	Marginal sea	-0.5	-5.8	2.5	1.5	-11.0	15.7	-5.1	21.4	51
20	Mediterranean Sea	Marginal sea	-0.1	-1.4	0.1	0.0	1.4	-4.2	-1.5	-11.9	33
21	Black Sea	Marginal sea	-0.2	7.6	3.0	3.4	74.5			25.2	0
28	Red Sea	Marginal sea	-0.2	-1.0	0.0	-0.2	0.9	13.6	0.2	-16.5	15
29	Persian Gulf	Marginal sea	-0.8	4.4	1.0	-0.2	1.3			-7.6	0

40	Sea of Japan	Marginal sea	0.1	-0.1	0.0	-0.2	-6.2	24.9	17.2	-9.3	28
41	Sea of Okhotsk	Marginal sea	-0.6	0.3	4.0	0.2	11.5	-22.0	-26.5	29.2	3
13	Canadian Archipelago	Polar	-0.5	2.5	1.1	-0.3	1.0	9.2	15.8	-53.1	12
14	N Greenland	Polar	-0.5	0.5	-0.5	-0.4	-2.4	-12.3	-10.0	-24.3	18
15	S Greenland	Polar	1.0	0.2	-0.7	-0.2	-0.9	6.6	0.0	1.3	47
16	Norwegian Basin	Polar	-0.5	0.1	-1.4	-0.2	-1.0	3.9	0.5	-0.7	77
43	Siberian Shelves	Polar	-0.1	1.2	1.8	-0.1	-0.2	14.7	21.6	-19.7	5
44	Barents and Kara seas	Polar	-0.5	-1.0	0.2	-0.1	-3.9	13.2	-0.9	-3.3	14
45	Antarctic Shelves	Polar	0.0	-0.2	0.0	0.0	-4.9	-25.8	-3.0	-17.6	39
1	N-E Pacific	Subpolar	-0.1	0.2	-1.6	-0.5	-4.2	-5.2	-24.9	16.8	61
5	Southern America	Subpolar	-0.1	0.0	-1.0	-0.1	5.3	7.9	1.3	14.0	49
11	Sea of Labrador	Subpolar	0.2	0.8	0.7	-0.2	0.6	7.6	-5.2	5.5	20
17	NE Atlantic	Subpolar	0.0	-0.1	-0.4	-0.1	0.0	-5.2	-3.1	-4.5	80
34	S Australia	Subpolar	-0.4	-0.2	-0.2	-0.1	0.0	5.2	-5.7	13.5	28
36	New Zealand	Subpolar	-0.2	-0.5	0.8	-0.1	0.9	13.7	-0.6	6.1	38
42	NW Pacific	Subpolar	-0.1	0.5	1.5	-0.1	5.8	46.8	-0.2	25.2	40
3	Tropical E Pacific	Tropical	-0.1	0.1	-0.8	-0.3	-0.9	7.9	-5.8	17.2	53
7	Tropical W Atlantic	Tropical	0.1	-2.0	1.4	-0.2	-2.1	11.4	-1.9	-19.8	20
8	Caribbean Sea	Tropical	0.0	0.3	-0.3	-0.1	-1.9	3.2	-2.8	-1.7	42
23	Tropical E Atlantic	Tropical	0.3	-0.1	0.3	-0.1	-1.4	27.8	6.5	15.9	12
26	Tropical W Indian	Tropical	-0.1	0.4	-0.1	-0.2	-0.6	1.0	-20.0	4.8	4
37	N Australia	Tropical	0.1	-0.2	0.0	-0.2	0.7	7.0	6.0	-4.0	11
38	SE Asia	Tropical	0.3	-0.8	0.6	-0.2	-2.9	0.1	10.9	0.6	5
6	Brazilian Current	WBC	0.2	-0.6	-0.6	-0.2	-1.6	10.9	-1.3	7.0	48
10	U.S. East Coast	WBC	1.3	1.0	0.5	-0.2	0.4	-7.6	-2.2	-9.6	83
25	Agulhas Current	WBC	0.1	0.4	-0.2	-0.2	-0.7	10.2	2.2	5.7	18
35	E Australian Current	WBC	-0.6	-0.3	-0.4	-0.2	-0.2	10.0	6.1	2.9	48
39	China Sea and Kuroshio	WBC	0.0	-0.8	1.5	-0.2	-2.4	2.7	5.5	-4.1	38

Table S2: Seasonal evaluation of MOM6-COBALT against observations (SST, SSS, nutrients and pCO_2) for each MARCATS. The observational SST and SSS fields are from the NOAA OI SST V2 (Reynolds et al., 2007) and the EN4 SSS (Good et al., 2013). The observational nutrients derived from the World Ocean Atlas version 2018 (Garcia et al., 2019).The evaluation is performed on their seasonal amplitude which is expressed as the bias between the RMS of their amplitude. A positive value indicates higher values simulated by MOM6-COBALT compared to observations. The seasonal evaluation is also performed on their seasonal cycles which is represented by the Pearson correlation coefficient. A Pearson correlation coefficient value of 1 indicates that both signals are perfectly in phase with one another while a value of -1 represents a complete phase shift. Pearson correlation coefficient < 0.5 for pCO_2 are highlighted in red. The seasonal pCO_2 simulated by MOM6-COBALT is evaluated against coastal-SOM-FFN and for some MARCATS against SOCATv6 (in bracket).

MARCATS number (Mx)	MARCATS name	MARCATS category	Evaluation against data										Evaluation against coastal-SOM-FFN (SOCATv6)	
			Bias RMS					Pearson correlation coefficient					Bias RMS	Pearson correlation coefficient
			SST (°C)	SSS (-)	Nutrients (µmol kg^{-1})			SST	SSS	Nutrients			pCO_2 (µatm)	
					NO_3	PO_4	SiO_4			NO_3	PO_4	SiO_4		
2	Californian Current	EBC	0.0	0.0	-0.1	0.0	-0.4	1.0	-0.3	-0.2	0.2	-0.1	16.2 (17.6)	1.0 (0.9)
4	Peruvian upwelling Current	EBC	0.1	0.0	0.0	0.0	-0.2	1.0	0.8	0.7	0.7	0.8	6.6	-0.4
19	Iberian upwelling	EBC	-0.2	0.0	-0.8	0.0	-0.7	1.0	0.7	1.0	0.9	0.6	15.6 (9.3)	0.8 (0.2)
22	Moroccan upwelling	EBC	-0.1	0.0	-0.3	0.0	-0.7	1.0	0.8	0.6	0.6	-0.1	8.7 (7.7)	0.9 (0.5)
24	SW Africa	EBC	0.1	0.0	-0.3	-0.1	-0.9	1.0	0.7	0.9	0.0	-0.4	4.2	0.9
33	Leeuwin Current	EBC	0.0	0.0	-0.1	0.0	-0.1	1.0	1.0	0.2	0.0	0.0	12.7	0.9
27	W Arabian Sea	Indian margins	-0.1	0.0	-1.0	-0.1	-1.1	1.0	0.7	0.9	0.8	0.5	3.6	0.3
30	E Arabian Sea	Indian margins	0.0	-0.1	0.1	0.0	-0.9	1.0	0.9	0.7	0.2	0.0	6.2	0.7
31	Bay of Bengal	Indian margins	0.1	0.5	2.0	-0.1	-1.8	1.0	0.9	0.5	0.7	-0.1	13.5	-0.2
32	Tropical E Indian	Indian margins	-0.1	0.0	-0.1	0.0	-0.9	1.0	0.8	-0.7	0.0	-0.5	5.4	0.9
9	Gulf of Mexico	Marginal sea	-0.3	0.0	-0.4	-0.1	-0.5	1.0	0.6	0.1	0.1	-0.7	12.9 (10)	1.0 (0.9)
12	Hudson Bay	Marginal sea	-0.2	0.0	0.9	0.0	0.9	1.0	0.1	0.0	-0.4	0.6	-46.4	0.4
18	Baltic Sea	Marginal sea	-0.5	-0.1	-0.1	0.0	-2.0	1.0	0.9	0.9	0.9	0.6	-44.4	0.9
20	Mediterranean Sea	Marginal sea	-0.1	0.0	0.1	0.0	-0.2	1.0	0.6	0.7	0.5	0.0	20.6	1.0
21	Black Sea	Marginal sea	-1.3	0.1	3.1	0.2	1.2	1.0	0.9	0.6	0.6	0.5	-116.9	-0.5

28	Red Sea	Marginal sea	0.0	-0.2	-0.1	0.0	-0.3	1.0	0.2	0.4	0.5	-0.1	-0.4	-0.9
29	Persian Gulf	Marginal sea	-0.2	0.0	0.1	0.0	-0.1	1.0	0.9	0.0	0.5	0.7	30.7	-0.9
40	Sea of Japan	Marginal sea	-0.6	0.0	-1.3	-0.1	-3.0	1.0	1.0	0.9	1.0	0.6	28.0	0.9
41	Sea of Okhotsk	Marginal sea	-0.4	0.1	-1.1	-0.1	-6.2	1.0	0.9	1.0	0.9	1.0	-6.5	0.7
13	Canadian Archipelago	Polar	-0.4	0.0	0.6	0.0	-0.9	1.0	0.8	0.9	0.7	0.9	-18.0	0.9
14	N Greenland	Polar	0.2	0.1	-0.3	0.0	-0.6	1.0	0.8	1.0	0.9	0.8	-9.0	0.8
15	S Greenland	Polar	0.1	0.0	0.1	0.0	-1.0	1.0	0.6	1.0	1.0	0.9	-8.5 (-8.8)	1.0 (1.0)
16	Norwegian Basin	Polar	0.0	0.0	-0.5	0.0	-0.4	1.0	0.9	1.0	1.0	0.9	-6.1 (-4.1)	0.9 (0.7)
43	Siberian Shelves	Polar	-0.4	-0.4	0.9	-0.1	-4.0	1.0	0.6	0.5	0.4	0.6	-15.7	0.9
44	Barents and Kara seas	Polar	-0.2	-0.3	0.1	0.0	-1.1	1.0	0.6	0.9	0.9	0.6	-7.4	0.7
45	Antarctic Shelves	Polar	0.2	0.1	2.0	0.1	1.3	1.0	0.9	1.0	0.9	0.6	13.3	1.0
1	N-E Pacific	Subpolar	-0.1	-0.1	-1.2	-0.2	-5.6	1.0	0.9	0.9	0.9	0.8	-4.5	0.8
5	Southern America	Subpolar	-0.1	0.0	-1.4	-0.1	-0.3	1.0	0.9	0.9	0.9	0.7	-6.4	0.8
11	Sea of Labrador	Subpolar	-0.2	-0.1	0.3	0.0	-0.5	1.0	1.0	0.9	0.9	0.9	0.8	0.2
17	NE Atlantic	Subpolar	0.0	0.1	-0.8	0.0	-0.7	1.0	1.0	1.0	1.0	1.0	-8.2 (-12.5)	0.6 (0.6)
34	S Australia	Subpolar	0.2	0.0	0.3	0.0	-0.2	1.0	0.6	0.9	0.7	0.2	12.8	0.9
36	New Zealand	Subpolar	0.0	0.0	0.4	0.0	-0.3	1.0	0.7	0.8	0.8	0.6	6.2 (2.8)	-0.5 (0.3)
42	NW Pacific	Subpolar	-0.2	0.0	-2.6	-0.3	-6.7	1.0	1.0	1.0	1.0	0.9	-19.2	1.0
3	Tropical E Pacific	Tropical	0.0	0.0	-0.5	0.0	-1.3	0.9	1.0	0.0	-0.4	0.3	3.1 (-3.3)	0.3 (0.4)
7	Tropical W Atlantic	Tropical	0.0	0.6	0.5	-0.1	-1.8	0.9	0.9	0.4	0.0	-0.4	9.6	1.0
8	Caribbean Sea	Tropical	-0.1	0.0	-0.2	0.0	-0.8	1.0	1.0	-0.2	-0.2	-0.8	2.2	1.0
23	Tropical E Atlantic	Tropical	-0.1	0.2	-0.5	0.0	-0.5	1.0	1.0	0.6	0.8	-0.5	1.5	0.6
26	Tropical W Indian	Tropical	0.0	0.1	0.0	0.0	-0.7	1.0	1.0	-0.6	0.5	0.3	5.6	0.9
37	N Australia	Tropical	0.0	0.0	0.0	0.0	-0.2	1.0	1.0	0.6	0.2	0.7	5.2	1.0
38	SE Asia	Tropical	-0.2	0.1	0.0	0.0	-1.0	1.0	1.0	-0.2	-0.3	-0.1	8.9	0.2
6	Brazilian Current	WBC	-0.1	0.0	-0.5	-0.1	-0.9	1.0	-0.2	0.8	0.7	-0.4	7.5	0.9
10	U.S. East Coast	WBC	-0.5	0.0	0.0	0.0	-0.3	1.0	1.0	0.9	0.9	0.8	12.4 (5.7)	0.9 (0.9)
25	Agulhas Current	WBC	0.0	0.1	-0.1	0.0	-1.0	1.0	1.0	0.4	0.1	0.3	8.1	1.0
35	E Australian Current	WBC	0.2	0.0	0.1	0.0	-0.3	1.0	0.6	1.0	0.9	0.6	7.4	1.0

39	China Sea and Kuroshio	WBC	-0.2	0.0	-0.6	0.0	-1.3	1.0	0.9	1.0	0.9	0.9	13.2 (9.1)	0.9 (0.4)