S3.1 North Atlantic

S3.1.1 North Atlantic carbon states from observations

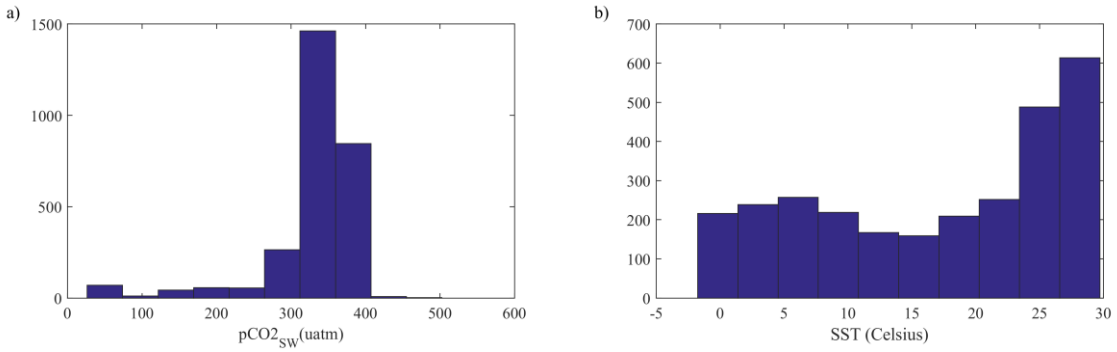


Fig. S1: Probability density distributions of North Atlantic observations data for (a) $pCO2_{SW}$ and (b) SST.

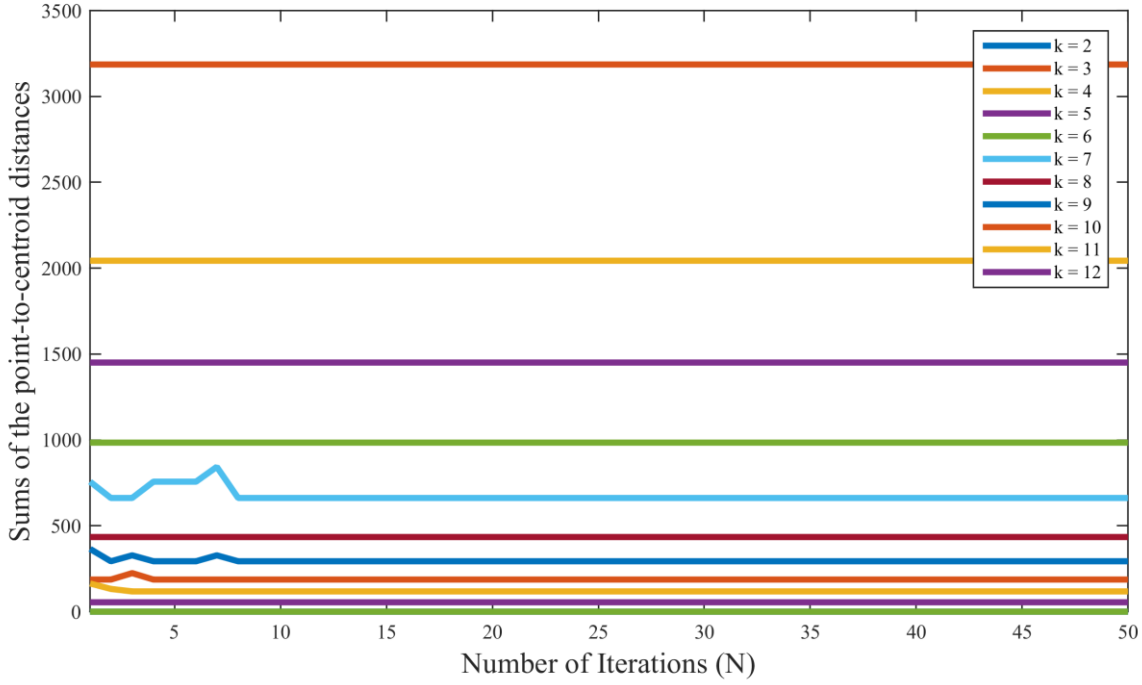


Fig. S2: Convergence test that shows the number of iterations needed so that clusters are unchanged. Each clustering analysis is performed for a different number of predetermined clusters (k) and is deemed as converged when the sum of the distances between each member 2D histogram from the cluster centroid is no longer changing. Convergence is tested for different number of predetermined clusters, k=1, k=2, …, k=12. We find that less than 10 iterations are needed to obtain convergence.

S3.1.2 North Atlantic carbon states in models

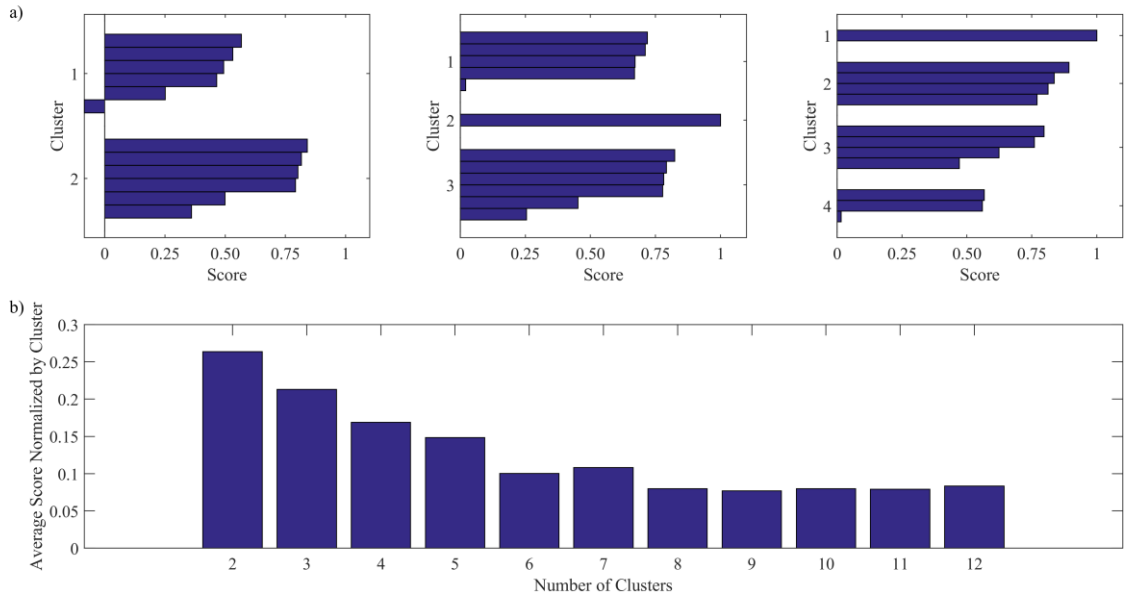


Fig. S3: a) Scores for each cluster analysis of model data in the North Atlantic for k = 2, k = 3, k = 4. b) Average scores of the clustering technique for each prechosen number of clusters.

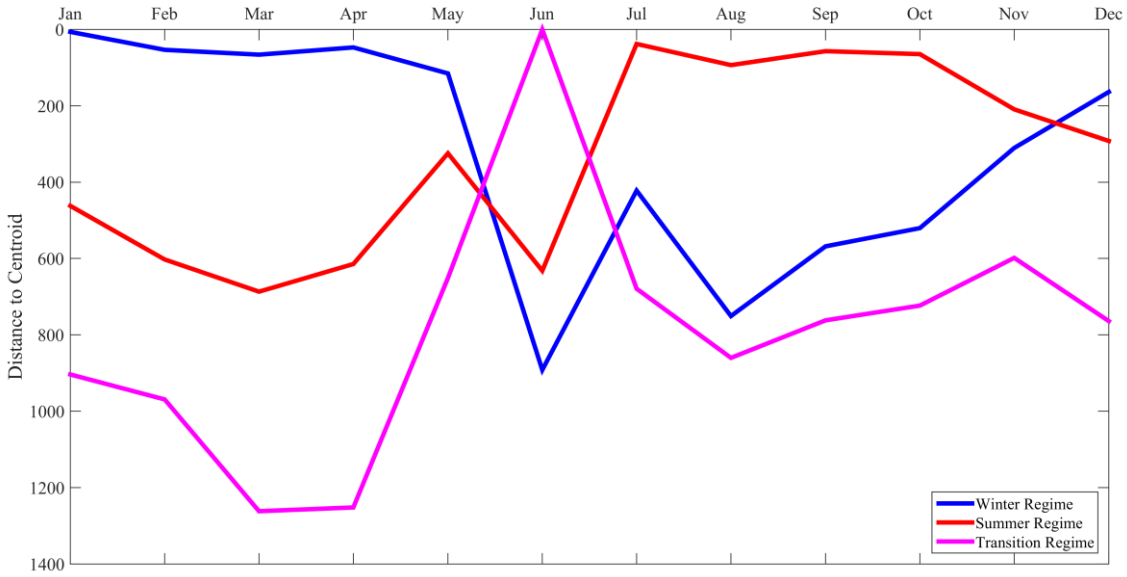


Fig. S4: Monthly attribution of each ocean carbon regime in the model dataset. Temporal attribution is based on the distance of each monthly 2D histogram to the centroid of each cluster.

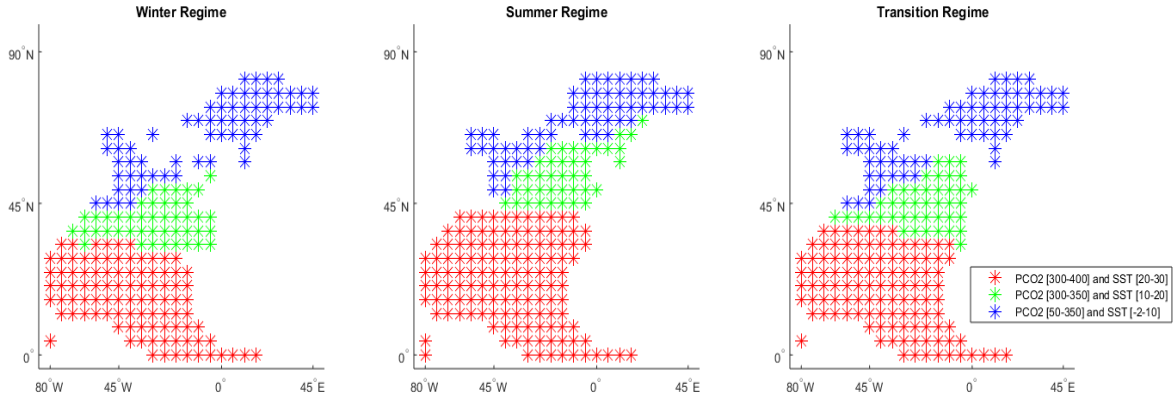
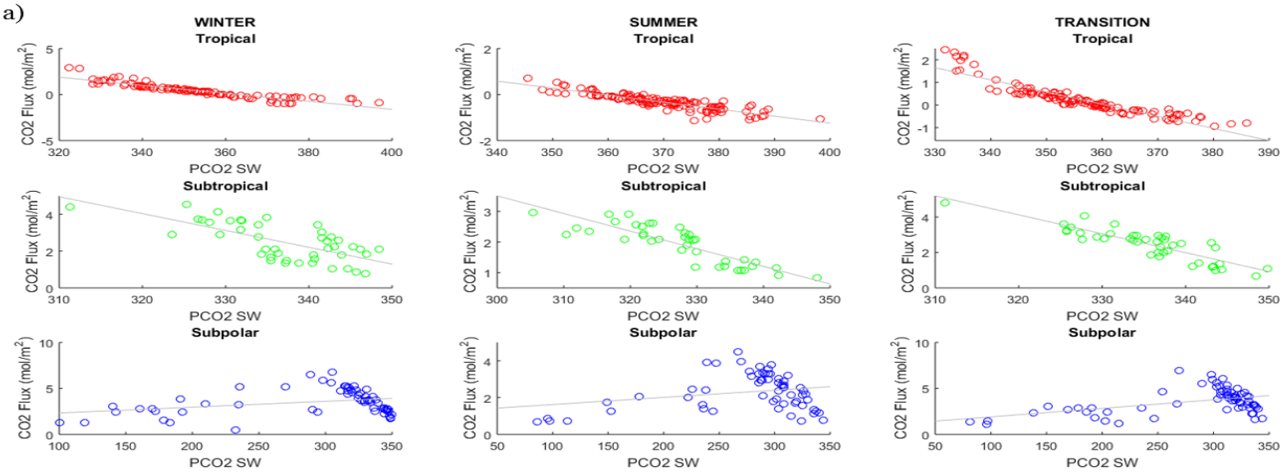


Fig. S5: Demarcated regions within the North Atlantic basin defined by ranges of pCO_{2SW} and SST values.

S3.1.3 Model air-sea flux of CO_2 error analysis and bias attribution

a)

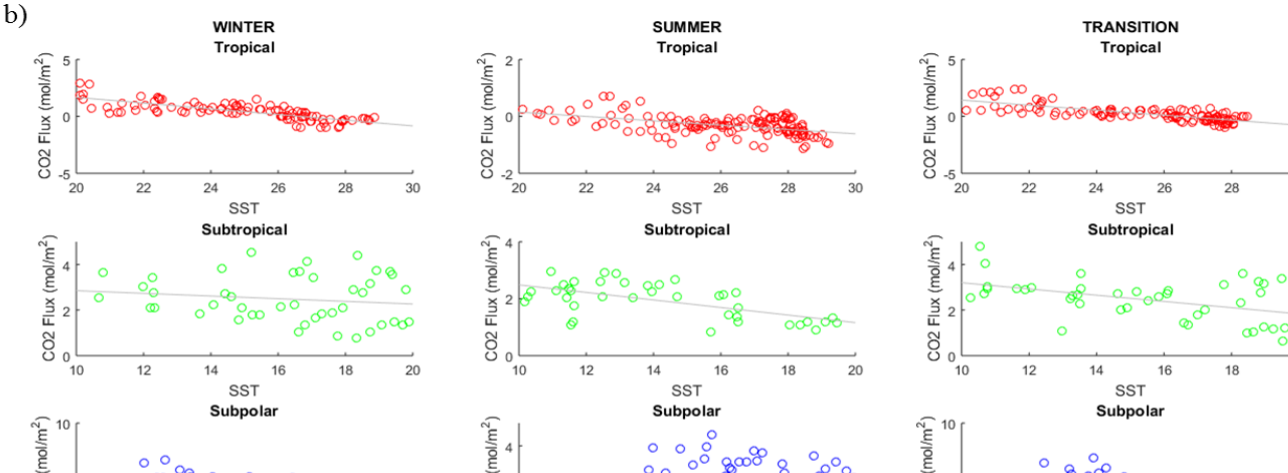


	RMSE	WEIGHT	Contribution
Tropical	37.123	-0.043694	-1.6221
Subtropical	18.353	-0.091541	-1.6801
Subpolar	117.85	0.0063676	0.75041

	RMSE	WEIGHT	Contribution
Tropical	35.568	-0.030469	-1.0837
Subtropical	18.843	-0.057559	-1.0846
Subpolar	74.048	0.0038785	0.2872

	RMSE	WEIGHT	Contribution
Tropical	37.407	-0.053648	-2.0068
Subtropical	18.45	-0.1068	-1.9705
Subpolar	96.923	0.0092523	0.89676

b)

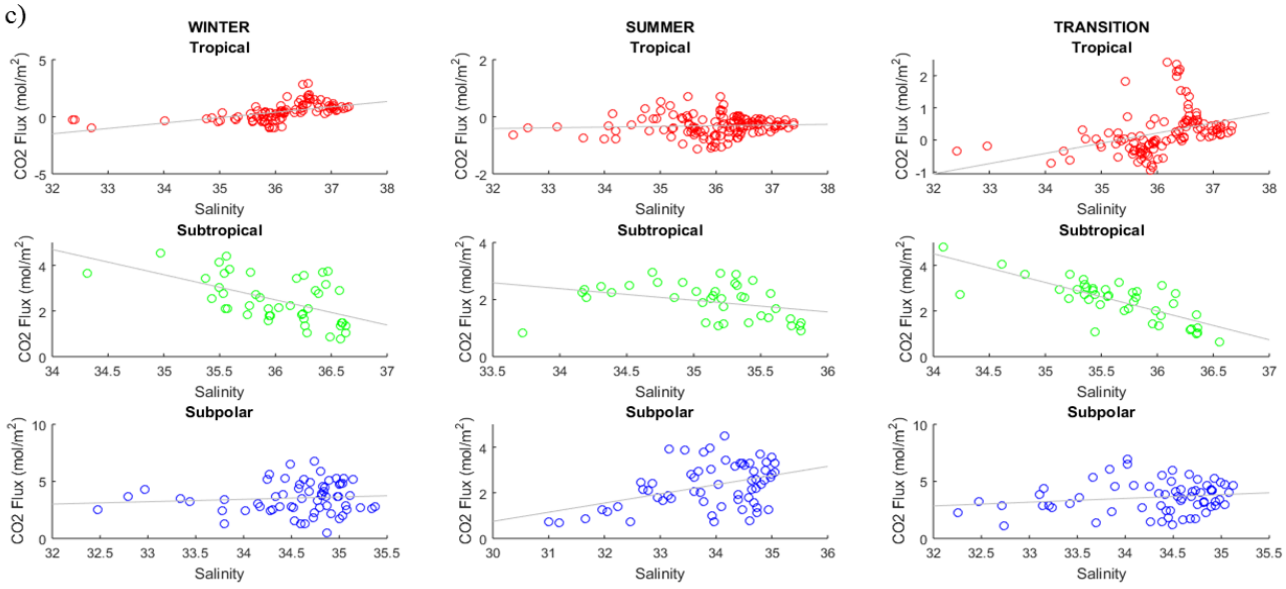


	RMSE	WEIGHT	Contribution
Tropical	1.0468	-0.25058	-0.26232
Subtropical	1.1045	-0.058524	-0.064639
Subpolar	2.8934	-0.030098	-0.087086

	RMSE	WEIGHT	Contribution
Tropical	1.4495	-0.076231	-0.1105
Subtropical	1.4735	-0.13263	-0.19542
Subpolar	3.4485	0.084806	0.29245

	RMSE	WEIGHT	Contribution
Tropical	1.0793	-0.22019	-0.23765
Subtropical	1.1242	-0.13694	-0.15395
Subpolar	2.9089	0.046045	0.13394

c)

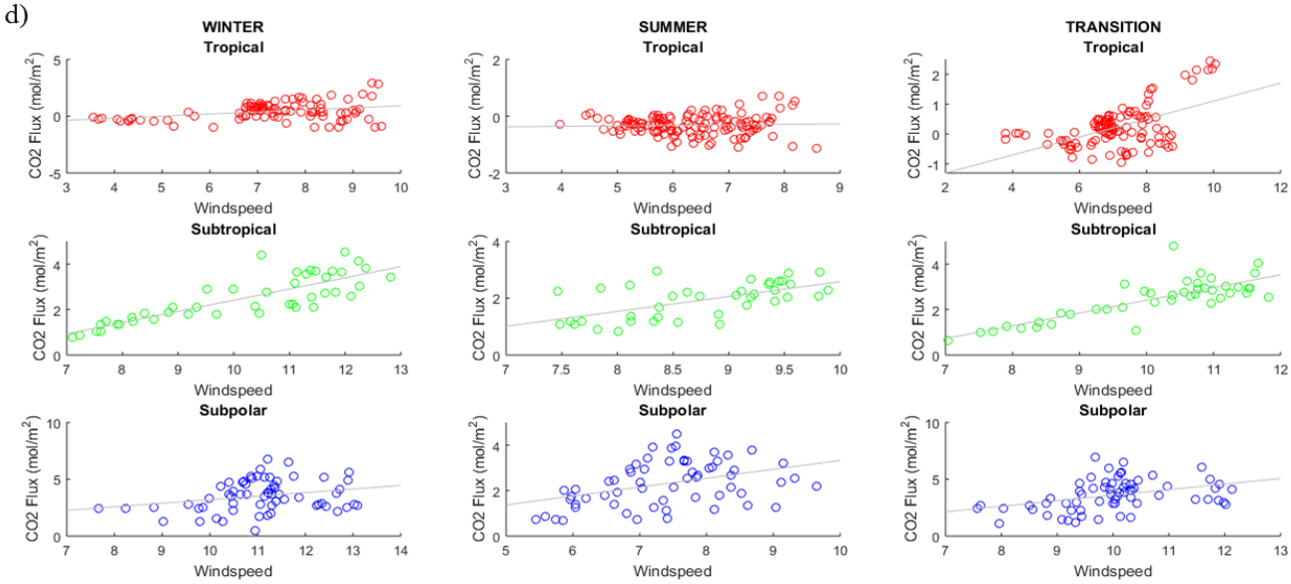


	RMSE	WEIGHT	Contribution
Tropical	0.5883	0.47181	0.27757
Subtropical	0.40244	-1.1026	-0.44372
Subpolar	1.3832	0.20802	0.28773

	RMSE	WEIGHT	Contribution
	0.59543	0.024209	0.014415
	0.44111	-0.40821	-0.18007
	1.9603	0.40142	0.78691

	RMSE	WEIGHT	Contribution
	0.49914	0.31722	0.15834
	0.49651	-1.2557	-0.62347
	1.5649	0.32974	0.51601

d)



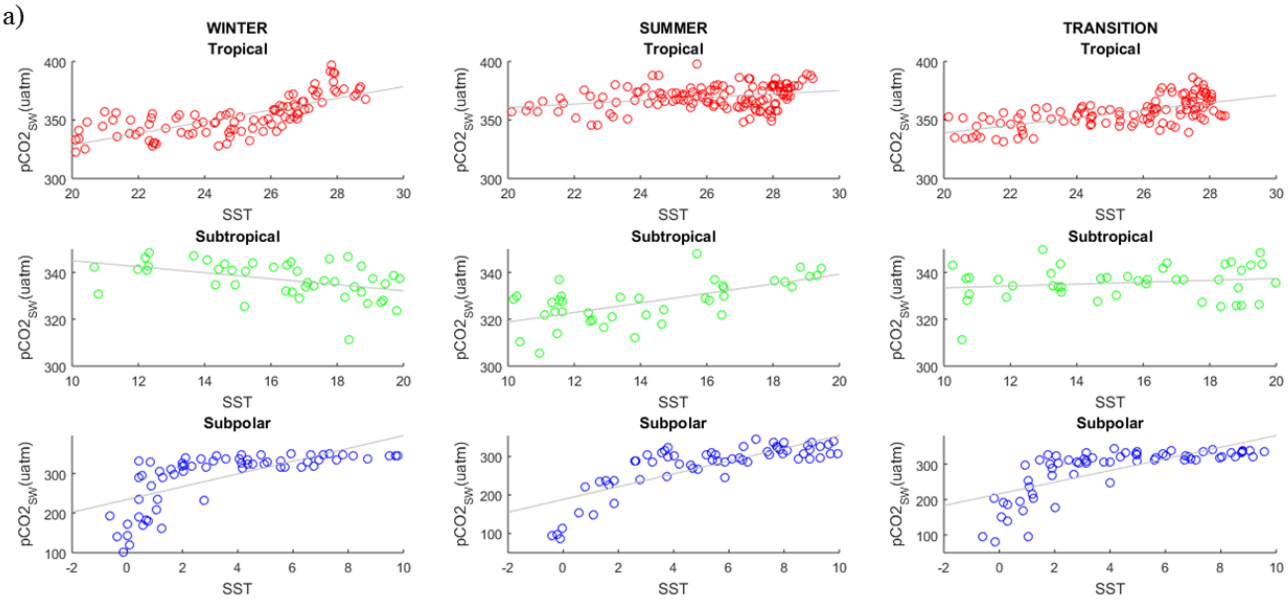
	RMSE	WEIGHT	Contribution
Tropical	1.1223	0.1827	0.20505
Subtropical	1.2357	0.49259	0.60867
Subpolar	2.4854	0.3137	0.77966

	RMSE	WEIGHT	Contribution
	1.1988	0.017796	0.021335
	1.3952	0.52245	0.72895
	1.077	0.39131	0.42143

	RMSE	WEIGHT	Contribution
	1.2952	0.30141	0.39039
	1.1105	0.56098	0.62298
	1.8207	0.48872	0.8898

Fig. S6: Scatter diagrams and linear fits of the air-sea flux of CO2 with a) $pCO2_{SW}$, b) SST, c) salinity, and d) wind speed in each of the North Atlantic regions that is represented in each regime. The RMSE terms are the bias terms denoted as Δq and the weight terms are the $\frac{\partial F}{\partial q}$ terms in Eq. (5). The contribution terms are the products of each bias*weight terms in Eq. (5).

a)

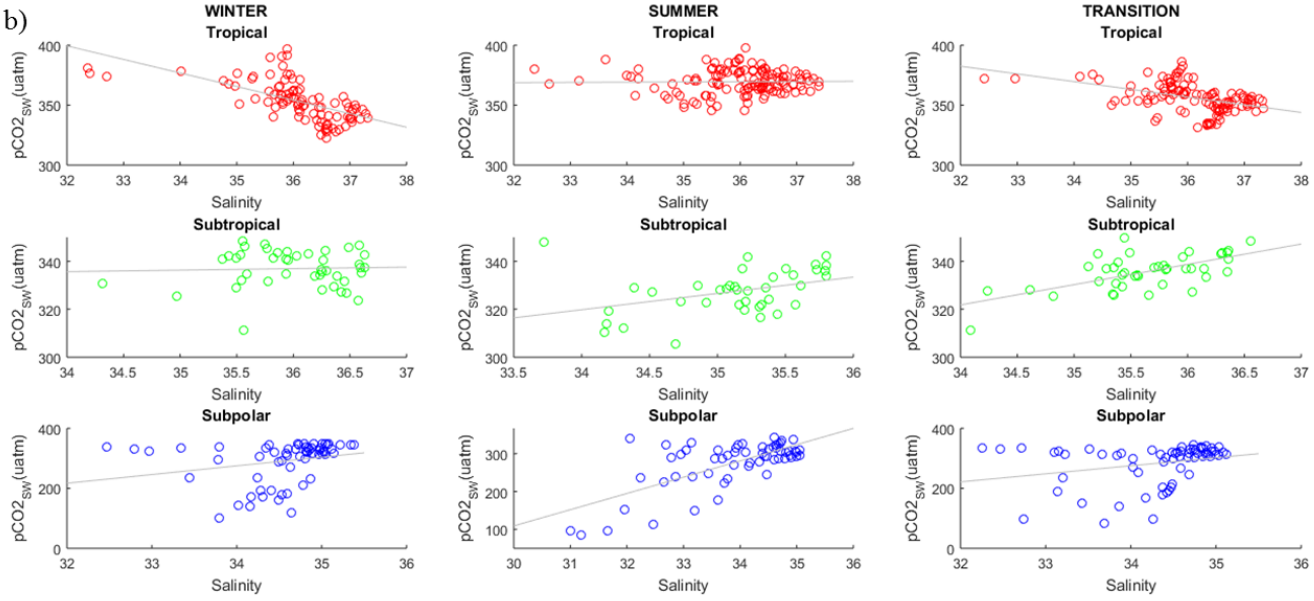


	RMSE	WEIGHT	Contribution
Tropical	1.0468	4.9894	5.223
Subtropical	1.1045	-1.2823	-1.4162
Subpolar	2.8934	16.13	46.67

	RMSE	WEIGHT	Contribution
	1.4495	1.4411	2.0889
	1.4735	2.0355	2.9992
	3.4485	16.581	57.18

	RMSE	WEIGHT	Contribution
	1.0793	3.2127	3.4675
	1.1242	0.39731	0.44666
	2.9089	16.442	47.828

b)

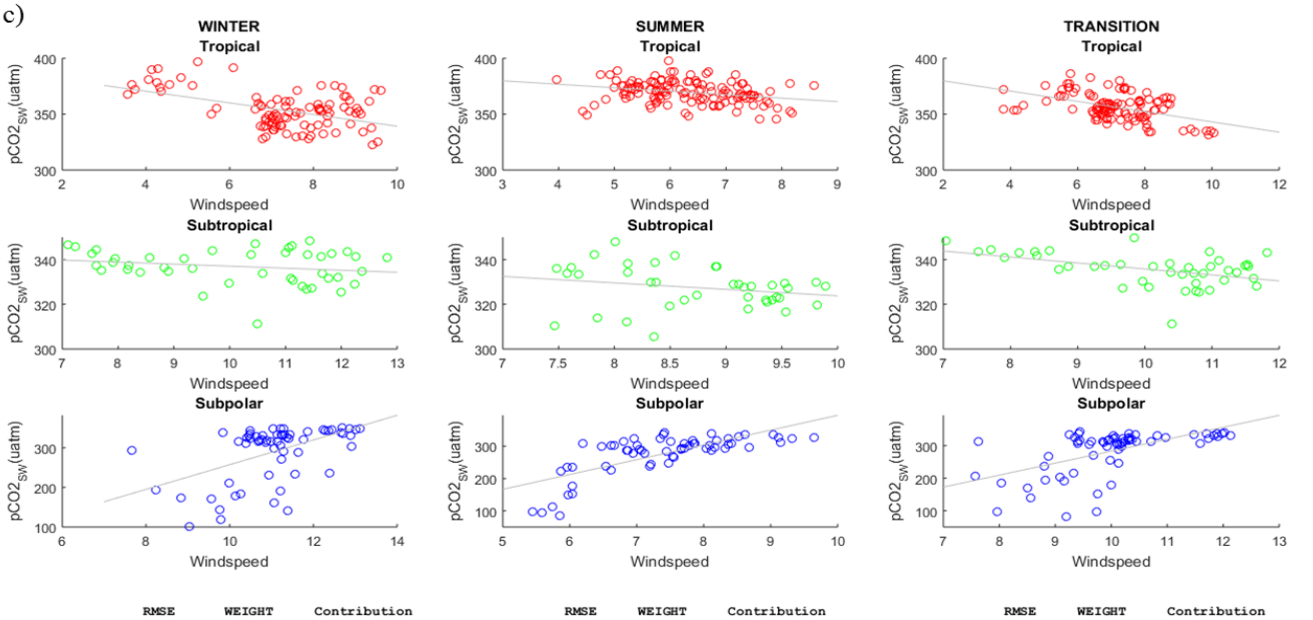


	RMSE	WEIGHT	Contribution
Tropical	0.5883	-11.311	-6.6541
Subtropical	0.40244	0.62911	0.25318
Subpolar	1.3832	28.93	40.015

	RMSE	WEIGHT	Contribution
	0.59543	0.17952	0.10689
	0.44111	6.7983	2.9988
	1.9603	43.005	84.304

	RMSE	WEIGHT	Contribution
	0.49914	-6.4285	-3.2088
	0.49651	8.4545	4.1977
	1.5649	26.782	41.911

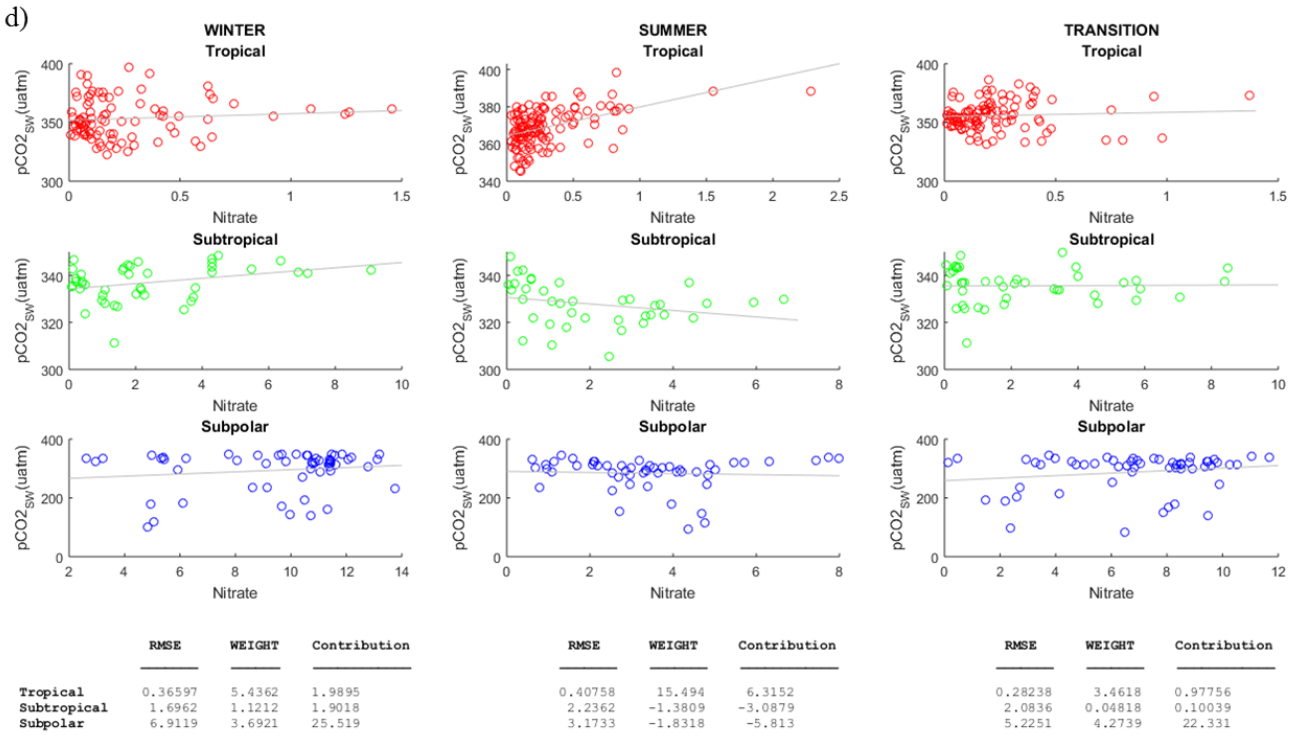
c)



	RMSE	WEIGHT	Contribution
Tropical	1.1223	-5.2078	-5.8449
Subtropical	1.2357	-0.91095	-1.1256
Subpolar	2.4854	31.143	77.404

	RMSE	WEIGHT	Contribution
	1.1988	-3.1069	-3.7247
	1.3952	-2.9038	-4.0516
	1.077	45.805	49.331

	RMSE	WEIGHT	Contribution
	1.2952	-4.5993	-5.957
	1.1105	-2.6724	-2.9678
	1.8207	36.761	66.929



	RMSE	WEIGHT	Contribution
Tropical	0.36597	5.4362	1.9895
Subtropical	1.6962	1.1212	1.9018
Subpolar	6.9119	3.6921	25.519

	RMSE	WEIGHT	Contribution
	0.40758	15.494	6.3152
	2.2362	-1.3809	-3.0879
	3.1733	-1.8318	-5.813

	RMSE	WEIGHT	Contribution
	0.28238	3.4618	0.97756
	2.0836	0.04818	0.10039
	5.2251	4.2739	22.331

Fig. S7: Scatter diagrams and linear fits of $pCO2_{SW}$ with a) SST, b) salinity, c) wind speed and d) nitrate in each of the North Atlantic regions of each regime.

S3.2 Southern Ocean

S3.2.1 Southern Ocean carbon states based on observations and model simulations

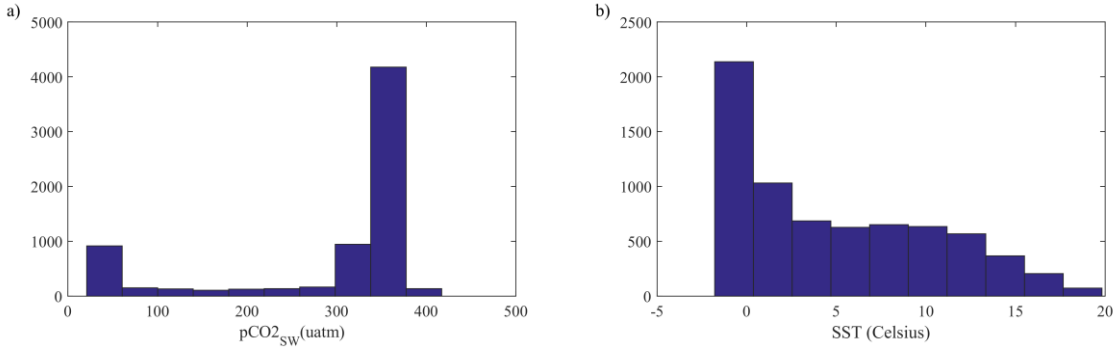


Fig. S8: Probability density distributions of Southern Ocean observations data for (a) $pCO2_{SW}$ and (b) SST.

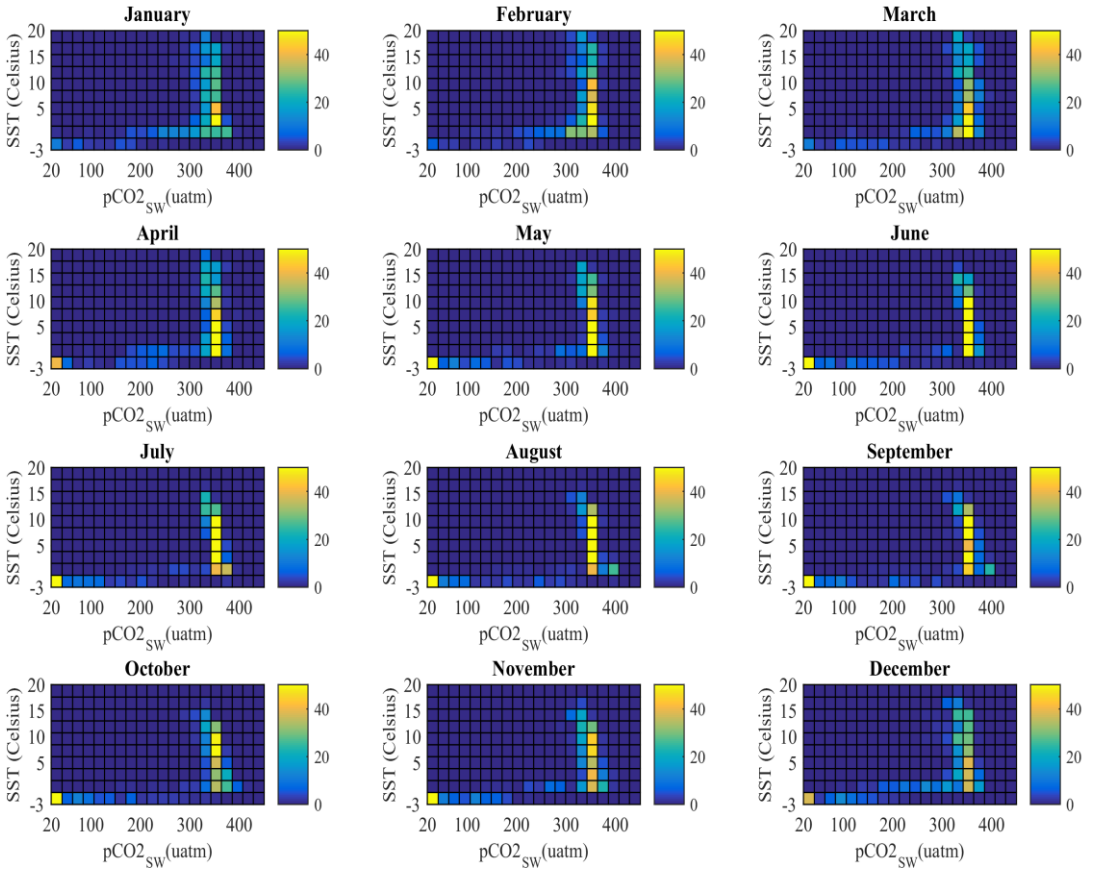


Fig. S9: Monthly 2D histograms of pCO2 of surface water (pCO2sw) and SST in the Southern Ocean (defined as 180°W to 180°E, 90°S to 40°S) from the Takahashi observational dataset.

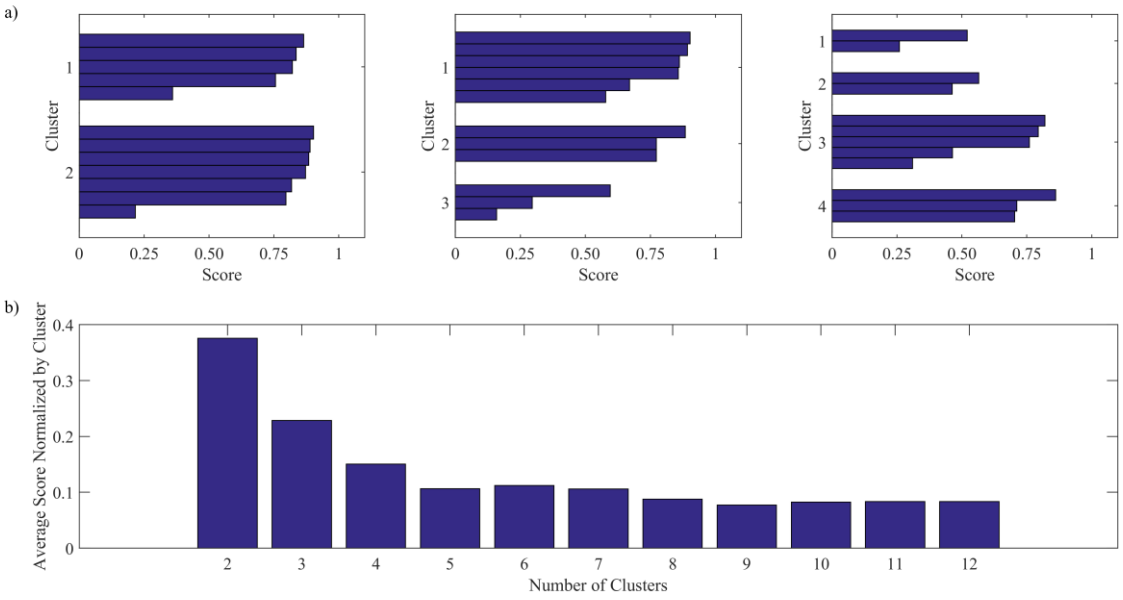


Fig. S10: a) Scores for each cluster analysis for k = 2, k = 3, k = 4 of model data in the Southern Ocean. b) Average scores of the clustering analysis for increasing k.

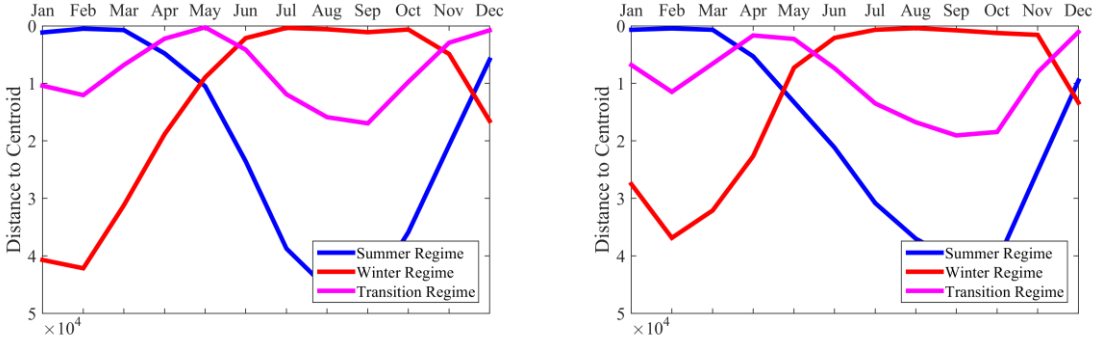


Fig. S11: Temporal attribution for the Southern Ocean (a) observation cluster run and (b) the model cluster run

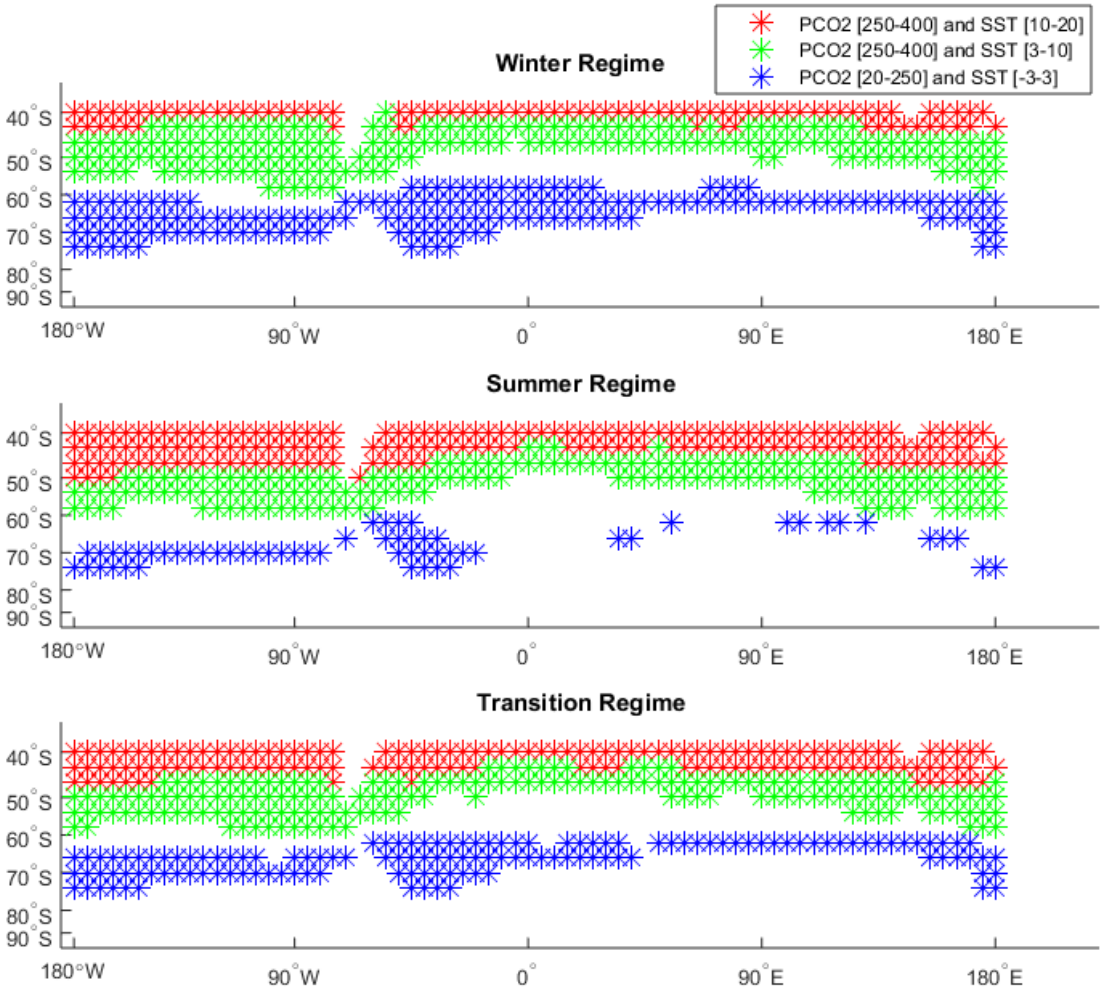


Fig. S12: Demarcated regions within the Southern Ocean basin defined by ranges of pCO_{2SW} and SST values.

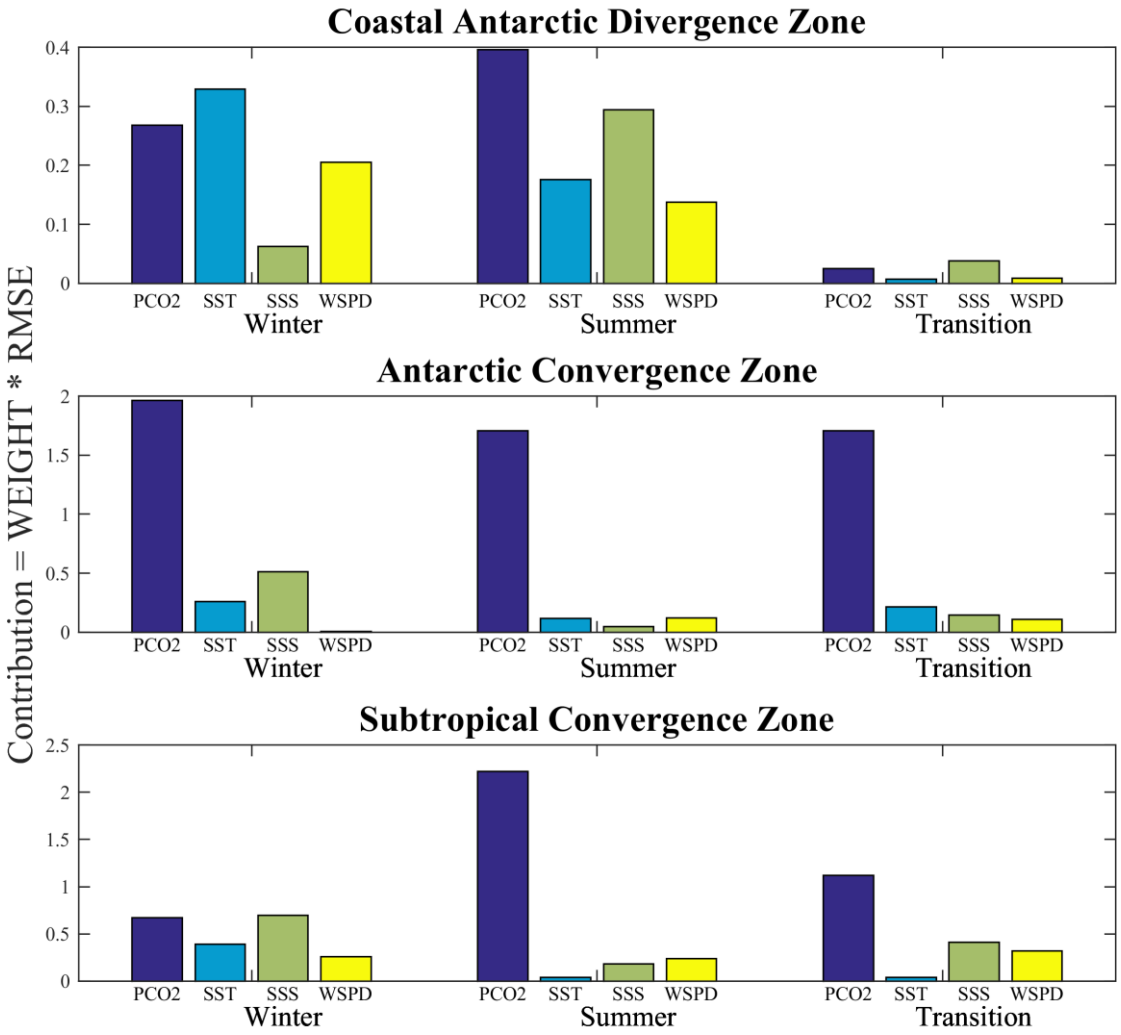
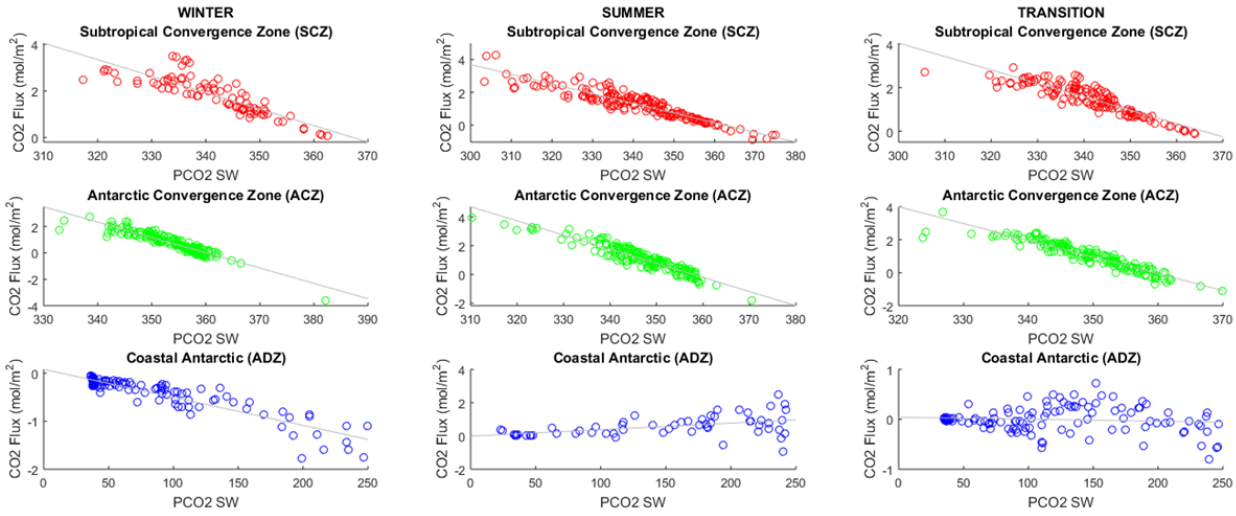


Fig. S13: Bias terms as computed in the Taylor expansion of the model bias for the air-sea flux of CO2.

a)

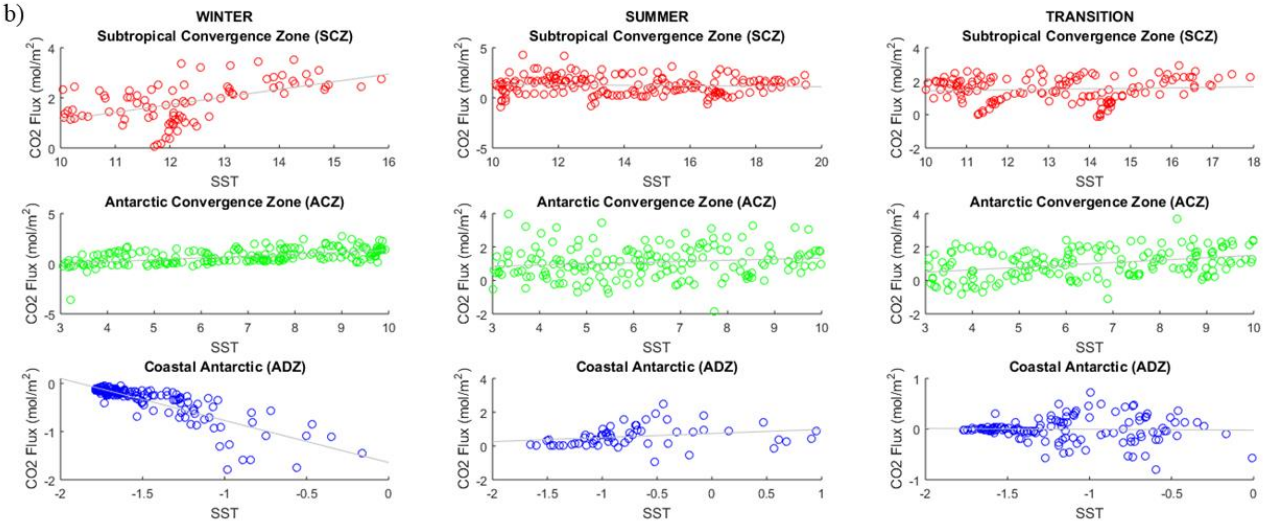


	RMSE	WEIGHT	Contribution
SCZ	9.4868	-0.070866	-0.67229
ACZ	16.926	-0.11606	-1.9645
ADZ	45.926	-0.0058327	-0.26787

	RMSE	WEIGHT	Contribution
SCZ	37.593	-0.059088	-2.2213
ACZ	17.335	-0.098416	-1.7061
ADZ	101.05	0.0039145	0.39556

	RMSE	WEIGHT	Contribution
SCZ	18.261	-0.061479	-1.1227
ACZ	16.81	-0.10166	-1.709
ADZ	64.404	-0.00038964	-0.025095

b)

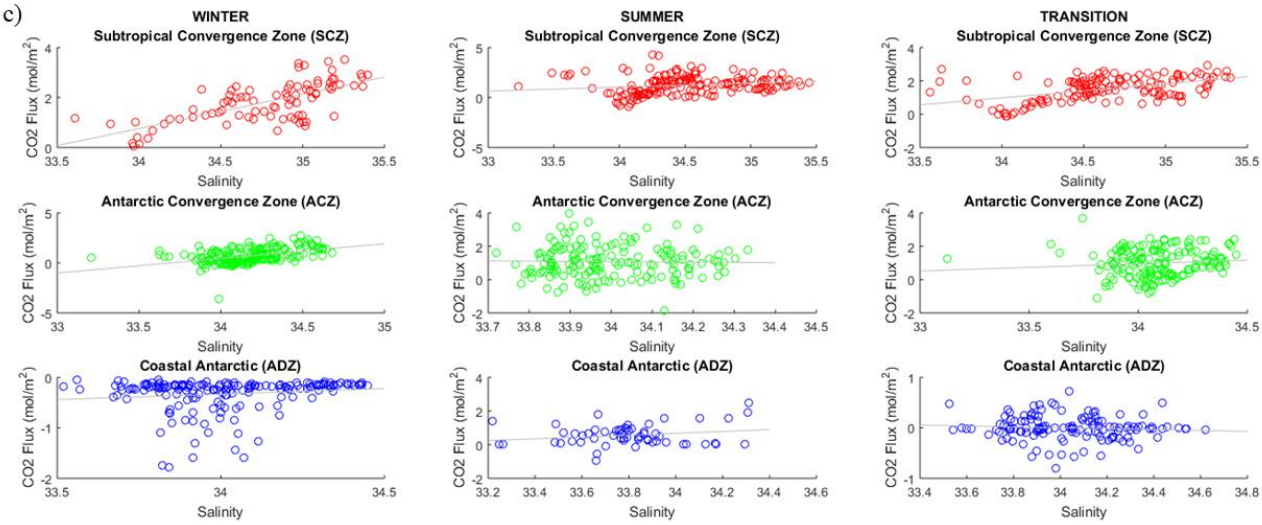


	RMSE	WEIGHT	Contribution
SCZ	1.2786	0.30523	0.39028
ACZ	1.3621	0.19047	0.25943
ADZ	0.3775	-0.87228	-0.32929

	RMSE	WEIGHT	Contribution
SCZ	1.6707	-0.022858	-0.038189
ACZ	1.6092	0.071622	0.11526
ADZ	0.72537	0.24193	0.17549

	RMSE	WEIGHT	Contribution
SCZ	1.3296	0.030282	0.040262
ACZ	1.4999	0.14442	0.21661
ADZ	0.42541	-0.016302	-0.0069349

c)

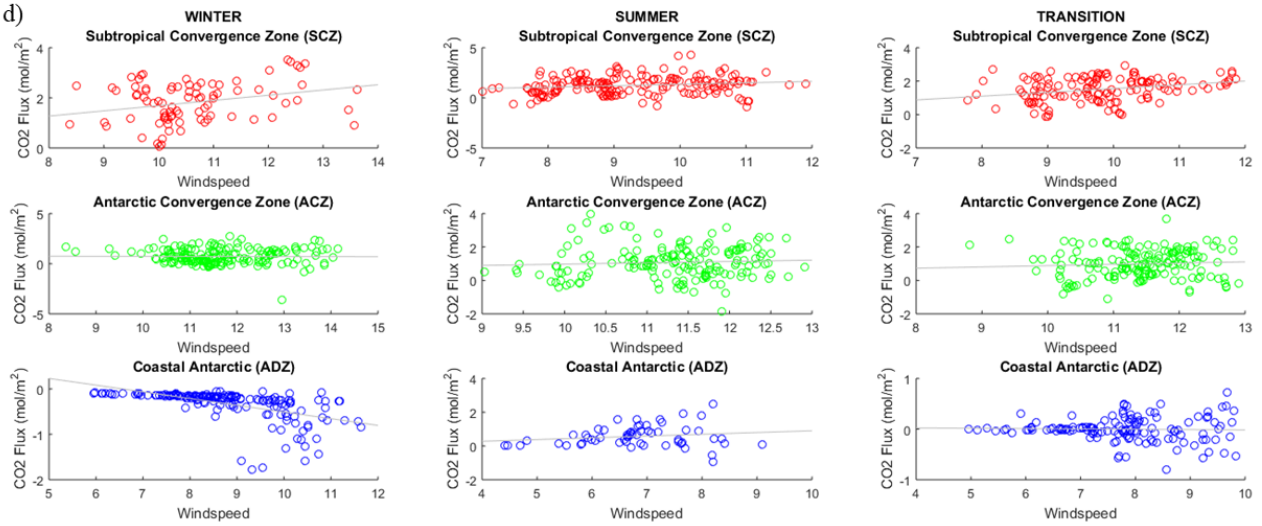


	RMSE	WEIGHT	Contribution
SCZ	0.50763	1.3685	0.6947
ACZ	0.35067	1.4666	0.5143
ADZ	0.28297	0.21993	0.062234

	RMSE	WEIGHT	Contribution
	0.45174	0.39999	0.18069
	0.30756	-0.15369	-0.047269
	0.53836	0.54606	0.29398

	RMSE	WEIGHT	Contribution
	0.48722	0.84231	0.41039
	0.33487	0.43413	0.14538
	0.42904	-0.089671	-0.038473

d)

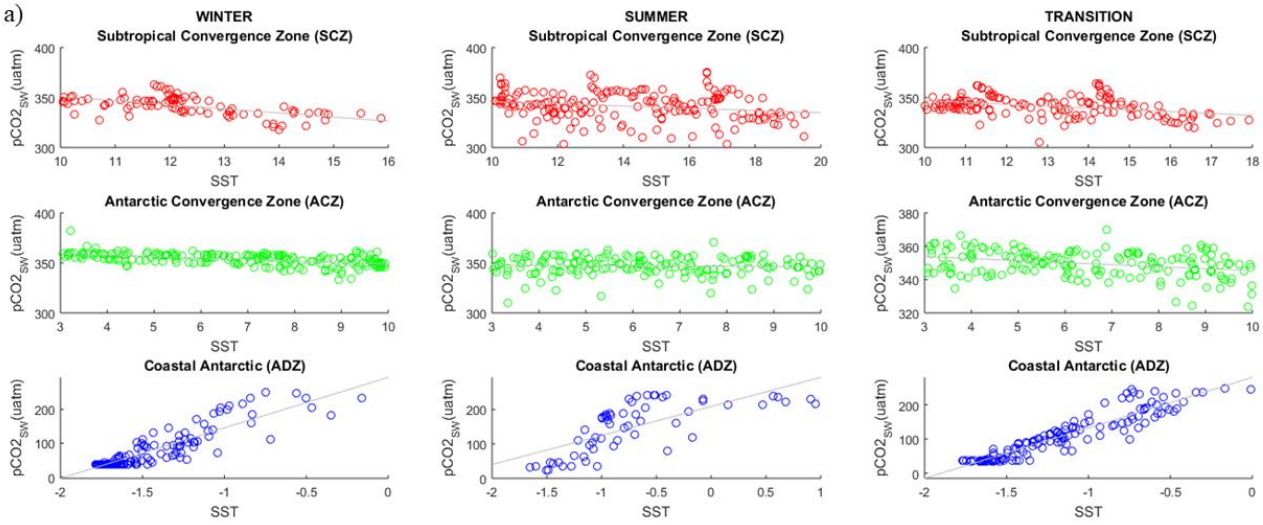


	RMSE	WEIGHT	Contribution
SCZ	1.2382	0.20805	0.2576
ACZ	1.5065	-0.0060822	-0.0091626
ADZ	1.3914	-0.14766	-0.20545

	RMSE	WEIGHT	Contribution
	1.6542	0.14321	0.23691
	1.5614	0.078969	0.1233
	1.3285	0.10377	0.13786

	RMSE	WEIGHT	Contribution
	1.4095	0.22587	0.31837
	1.4277	0.076115	0.10867
	1.3234	-0.0065508	-0.0086695

Fig. S14: Scatter diagrams and linear fits of the air-sea flux of CO_2 with a) $pCO2_{SW}$, b) SST, c) salinity, and d) wind speed in each of the Southern Ocean regions and for each regime.

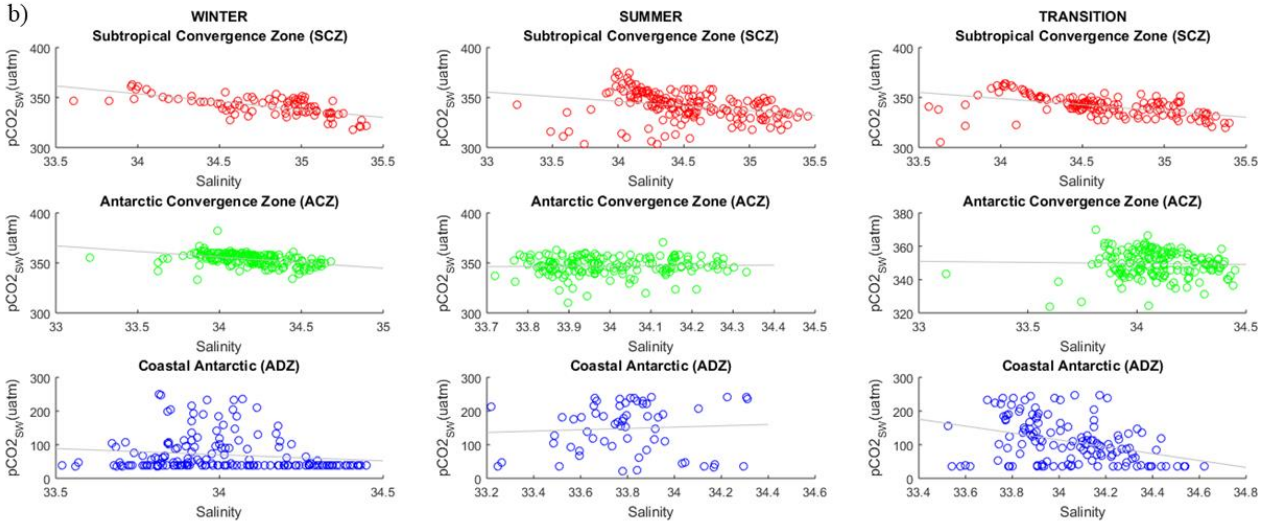


	RMSE	WEIGHT	Contribution
SCZ	1.2786	-4.1196	-5.2675
ACZ	1.3621	-1.5109	-2.058
ADZ	0.3775	148.92	56.217

	RMSE	WEIGHT	Contribution
SCZ	1.6707	-1.1189	-1.8693
ACZ	1.6092	-0.21114	-0.33977
ADZ	0.72537	84.277	61.132

	RMSE	WEIGHT	Contribution
SCZ	1.3296	-1.8385	-2.4444
ACZ	1.4999	-1.2111	-1.8165
ADZ	0.42541	147.62	62.799

b)

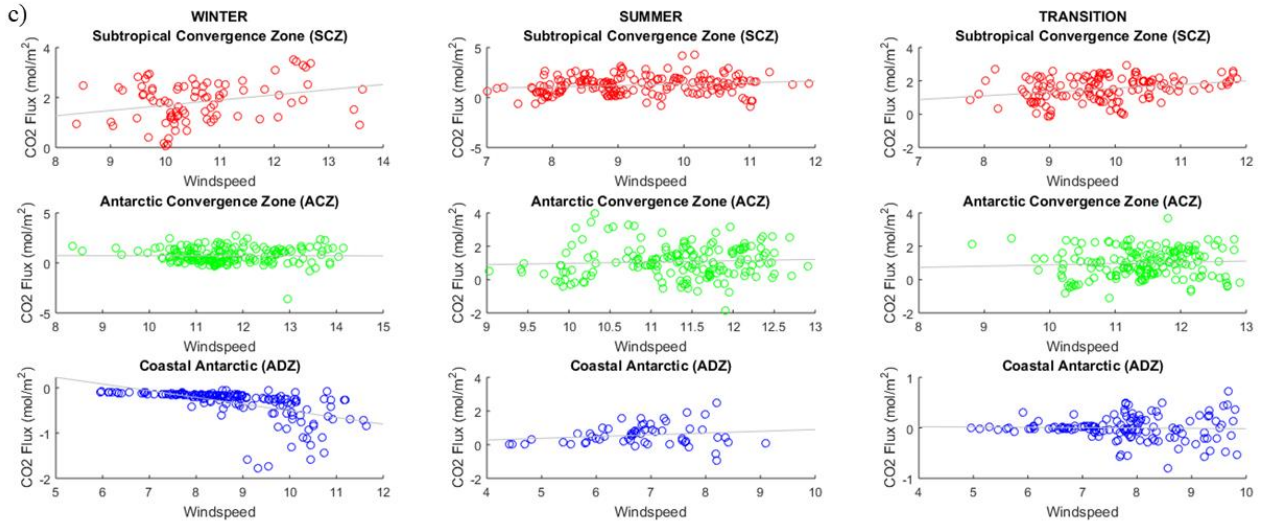


	RMSE	WEIGHT	Contribution
SCZ	0.50763	-15.768	-8.0045
ACZ	0.35067	-11.221	-3.935
ADZ	0.28297	-36.306	-10.273

	RMSE	WEIGHT	Contribution
	0.45174	-9.3632	-4.2298
	0.30756	2.3781	0.73142
	0.53836	19.523	10.51

	RMSE	WEIGHT	Contribution
	0.48722	-12.445	-6.0635
	0.33487	-1.2514	-0.41907
	0.42904	-102.26	-43.873

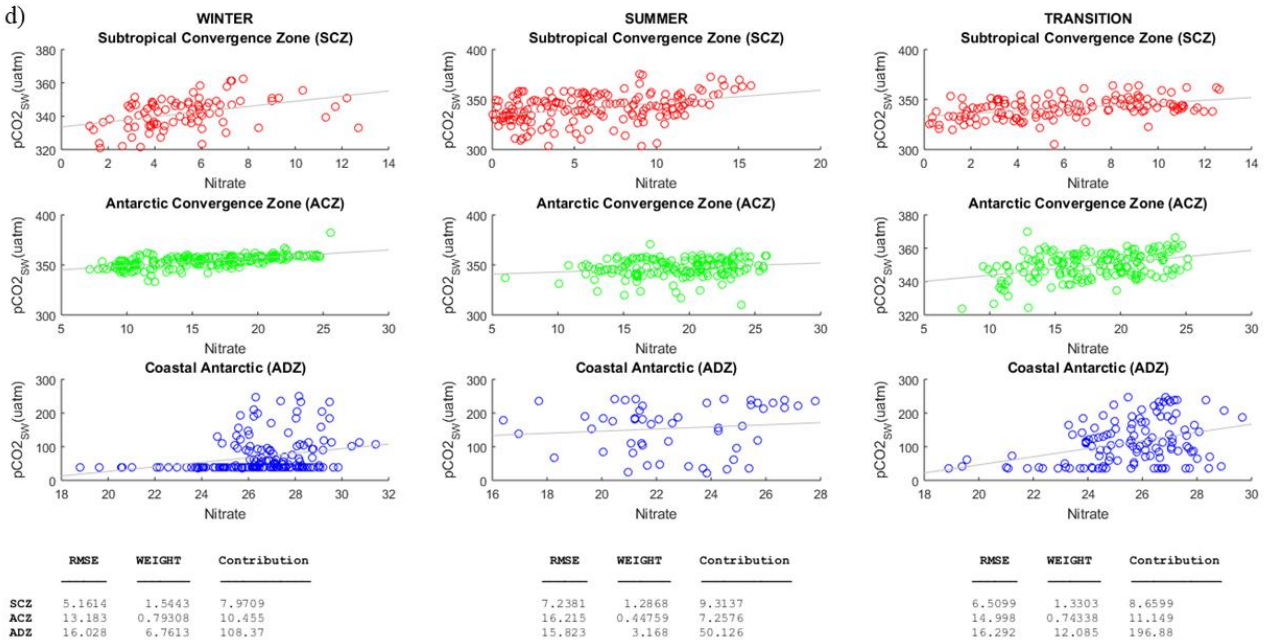
c)



	RMSE	WEIGHT	Contribution
SCZ	1.2382	0.20805	0.2576
ACZ	1.5065	-0.0060822	-0.0091626
ADZ	1.3914	-0.14766	-0.20545

	RMSE	WEIGHT	Contribution
	1.6542	0.14321	0.23691
	1.5614	0.078969	0.1233
	1.3285	0.10377	0.13786

	RMSE	WEIGHT	Contribution
	1.4095	0.22587	0.31837
	1.4277	0.076115	0.10867
	1.3234	-0.0065508	-0.0086695



	RMSE	WEIGHT	Contribution
SCZ	5.1614	1.5443	7.9709
ACZ	13.183	0.79308	10.455
ADZ	16.028	6.7613	108.37

	RMSE	WEIGHT	Contribution
SCZ	7.2381	1.2868	9.3137
ACZ	16.215	0.44759	7.2576
ADZ	15.823	3.168	50.126

	RMSE	WEIGHT	Contribution
SCZ	6.5099	1.3303	8.6599
ACZ	14.998	0.74338	11.149
ADZ	16.292	12.085	196.88

Fig. S15: Scatter diagrams and linear fits of $pCO2_{SW}$ with a) SST, b) salinity, c) wind speed and d) nitrate in each of the Southern Ocean regions and for each regime.