

S3. Methodology

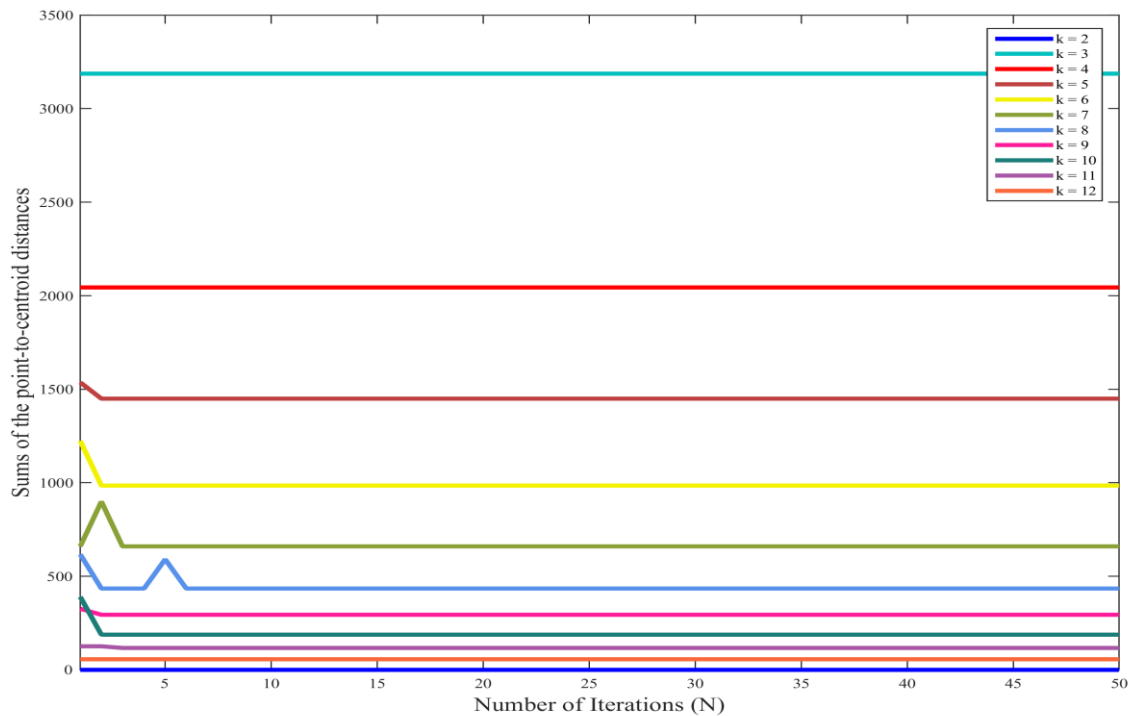


Fig. S1: Convergence test that shows the number of iterations needed so that clusters are unchanged. Each clustering analysis is performed for a different number of predetermined clusters (k) and is deemed as converging when the sum of the distances between each member 2D histogram from the cluster centroid 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.

S4. Results

S4.1 The North Atlantic Ocean Carbon States

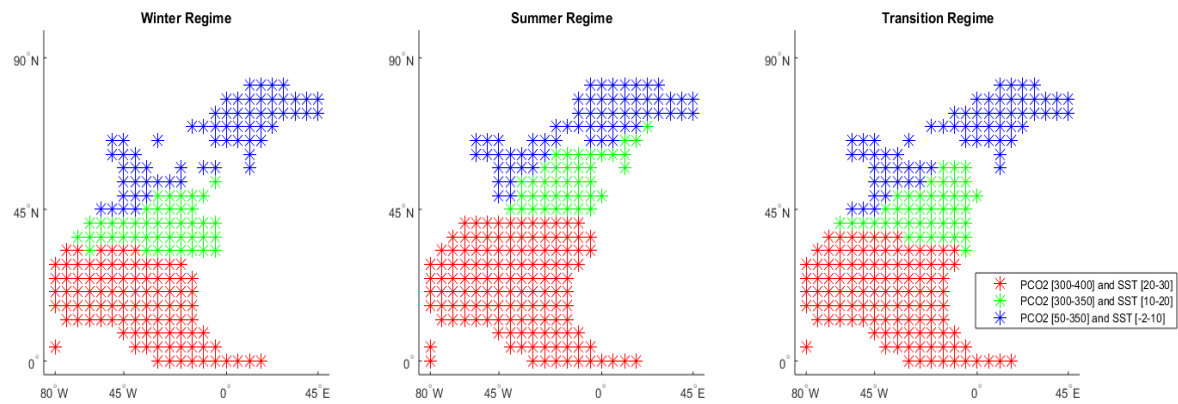
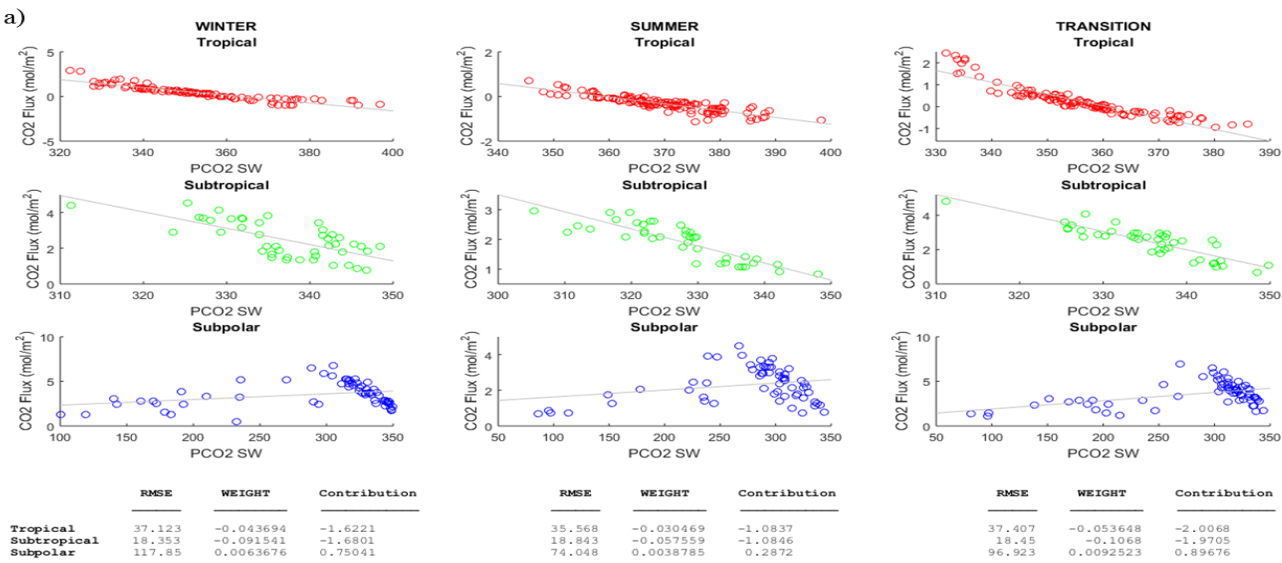
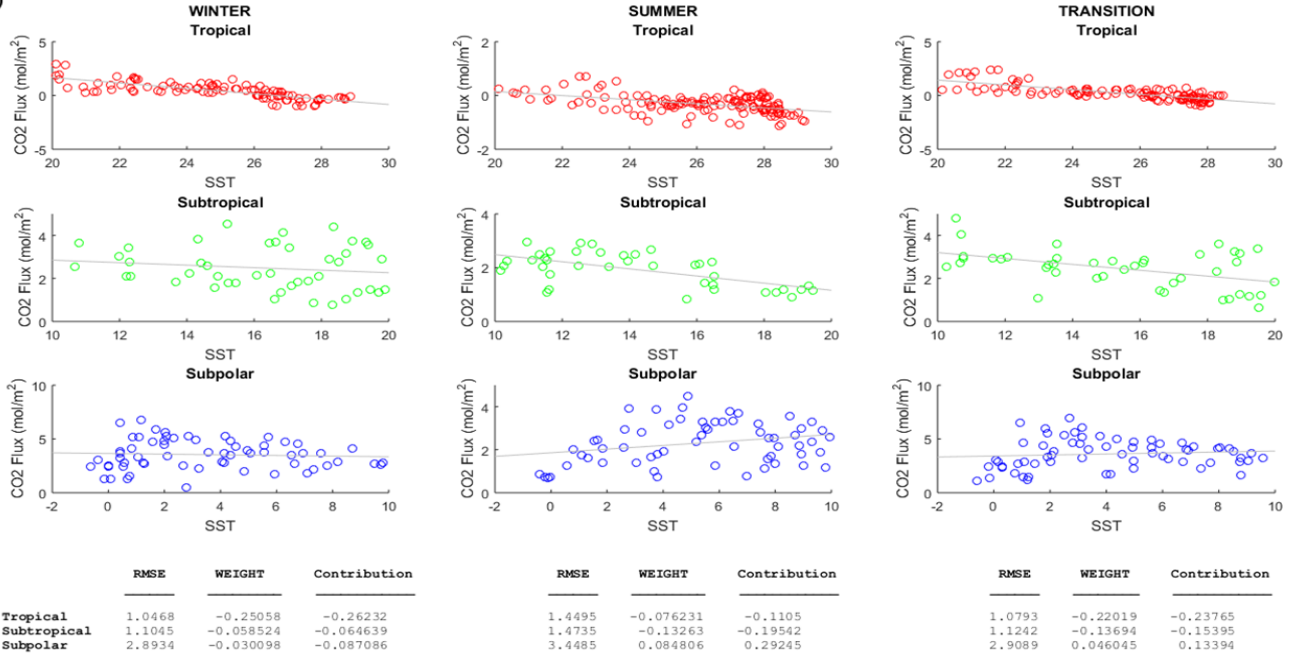


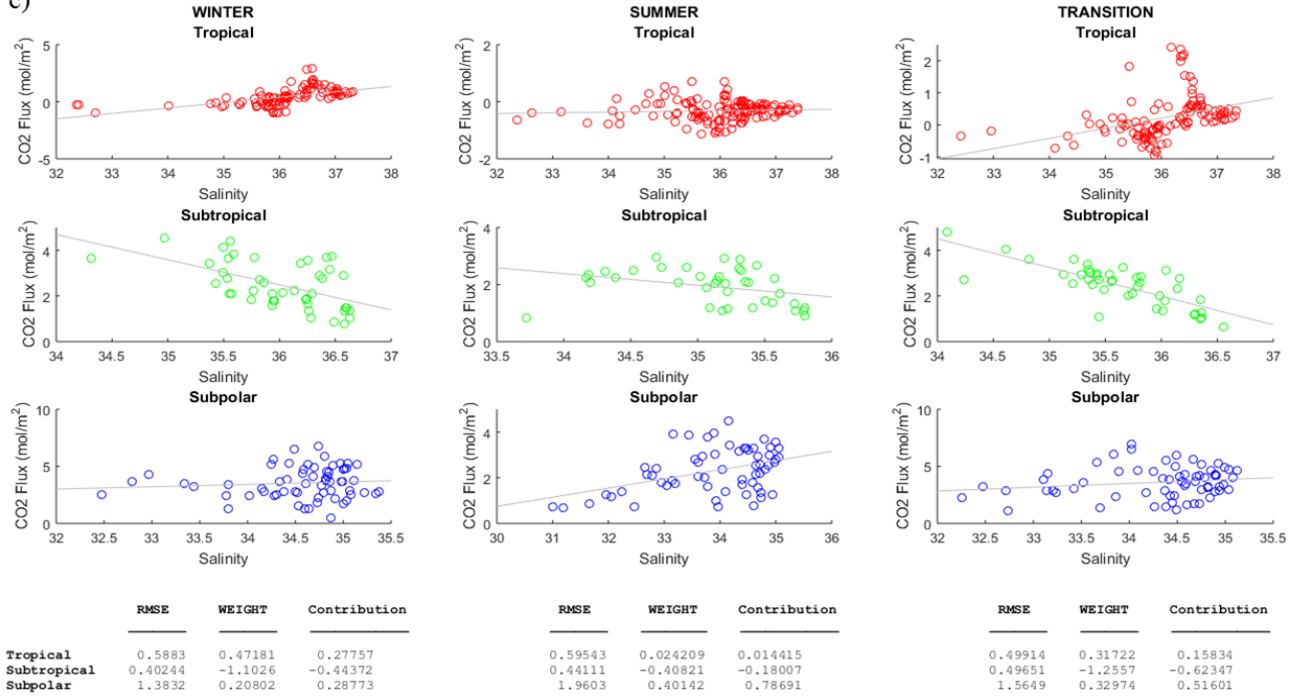
Fig. S2: Demarcated regions within the North Atlantic basin defined by ranges of  $\text{pCO}_{2\text{SW}}$  and SST values in the observed ocean carbon states.



b)



c)



d)

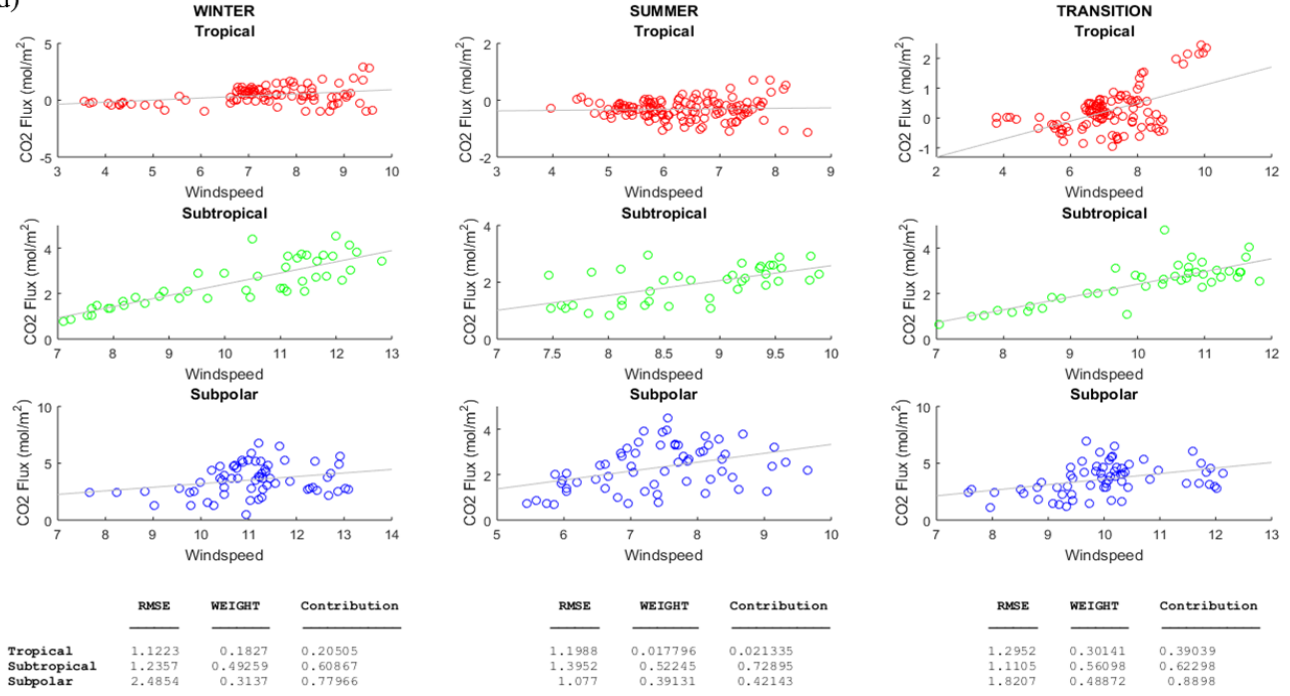
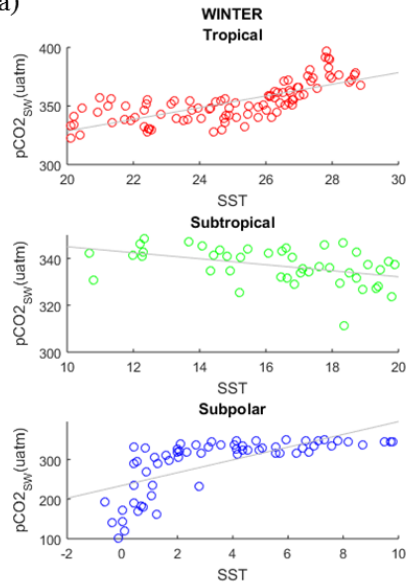
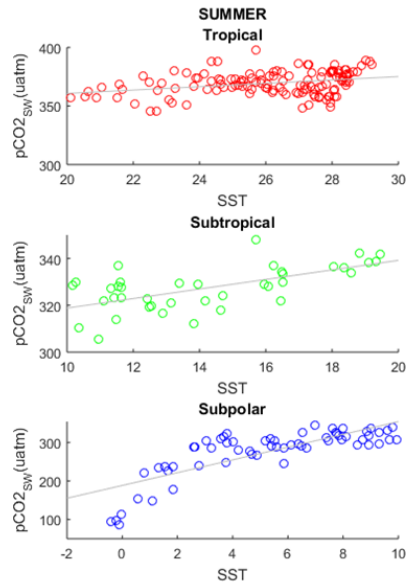


Fig. S3: Scatter diagrams and linear fits of the air-sea flux of CO<sub>2</sub> with a) pCO<sub>2sw</sub>, b) SST, c) salinity, and d) wind speed in each of the North Atlantic regions that is represented in each regime. The regions are also shown in Fig. S2. The RMSE terms are the bias terms denoted as  $\Delta q$  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).

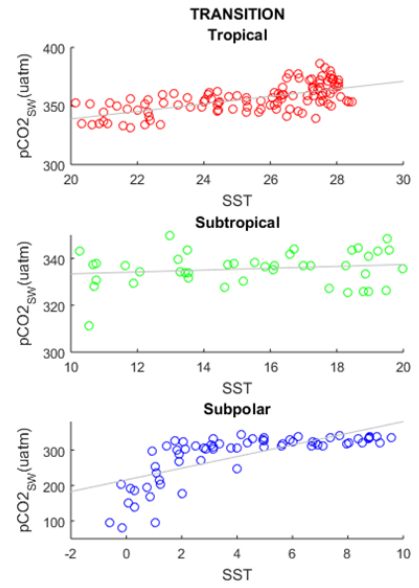
a)



|                    | RMSE   | WEIGHT  | Contribution |
|--------------------|--------|---------|--------------|
| <b>Tropical</b>    | 1.0468 | 4.9894  | 5.223        |
| <b>Subtropical</b> | 1.1045 | -1.2823 | -1.4162      |
| <b>Subpolar</b>    | 2.8934 | 16.13   | 46.67        |

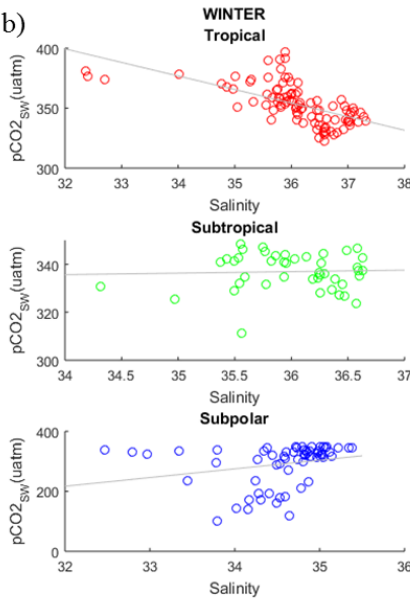


|                    | RMSE   | WEIGHT | Contribution |
|--------------------|--------|--------|--------------|
| <b>Tropical</b>    | 1.4495 | 1.4411 | 2.0889       |
| <b>Subtropical</b> | 1.4735 | 2.0355 | 2.9992       |
| <b>Subpolar</b>    | 3.4485 | 16.581 | 57.18        |

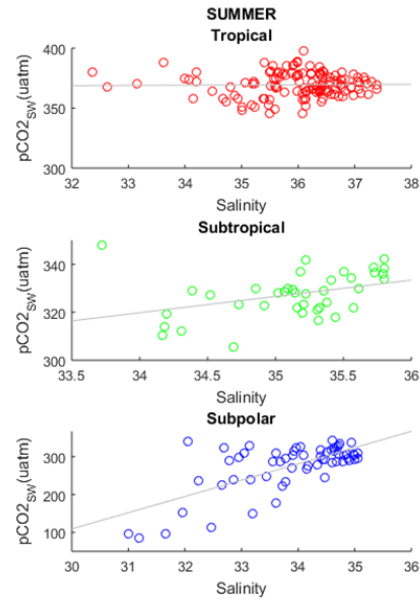


|                    | RMSE   | WEIGHT  | Contribution |
|--------------------|--------|---------|--------------|
| <b>Tropical</b>    | 1.0793 | 3.2127  | 3.4675       |
| <b>Subtropical</b> | 1.1242 | 0.39731 | 0.44666      |
| <b>Subpolar</b>    | 2.9089 | 16.442  | 47.828       |

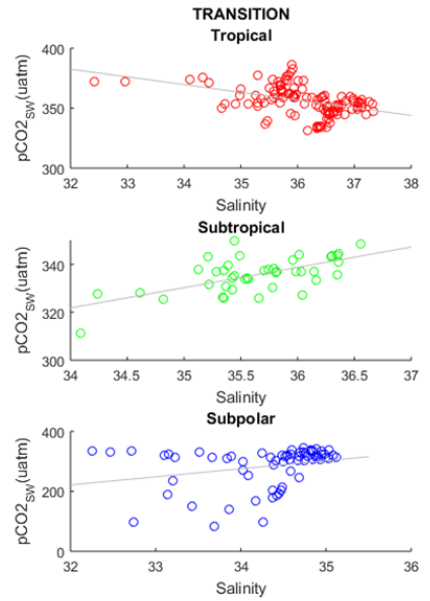
b)



|                    | RMSE    | WEIGHT  | Contribution |
|--------------------|---------|---------|--------------|
| <b>Tropical</b>    | 0.5883  | -11.311 | -6.6541      |
| <b>Subtropical</b> | 0.40244 | 0.62911 | 0.25318      |
| <b>Subpolar</b>    | 1.3832  | 28.93   | 40.015       |



|                    | RMSE    | WEIGHT  | Contribution |
|--------------------|---------|---------|--------------|
| <b>Tropical</b>    | 0.59543 | 0.17952 | 0.10689      |
| <b>Subtropical</b> | 0.44111 | 6.7983  | 2.9988       |
| <b>Subpolar</b>    | 1.9603  | 43.005  | 84.304       |



|                    | RMSE    | WEIGHT  | Contribution |
|--------------------|---------|---------|--------------|
| <b>Tropical</b>    | 0.49914 | -6.4285 | -3.2088      |
| <b>Subtropical</b> | 0.49651 | 8.4545  | 4.1977       |
| <b>Subpolar</b>    | 1.5649  | 26.782  | 41.911       |

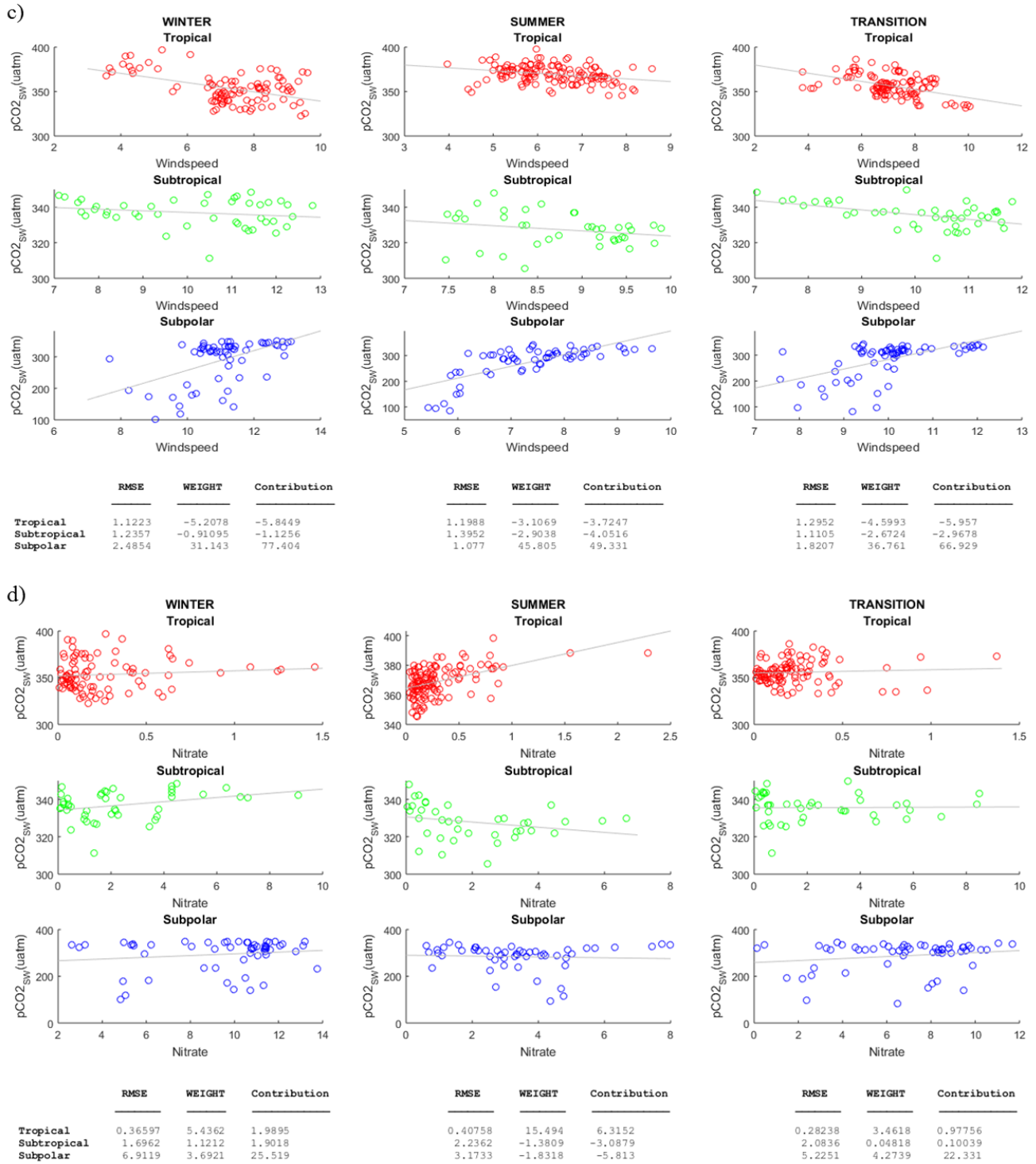
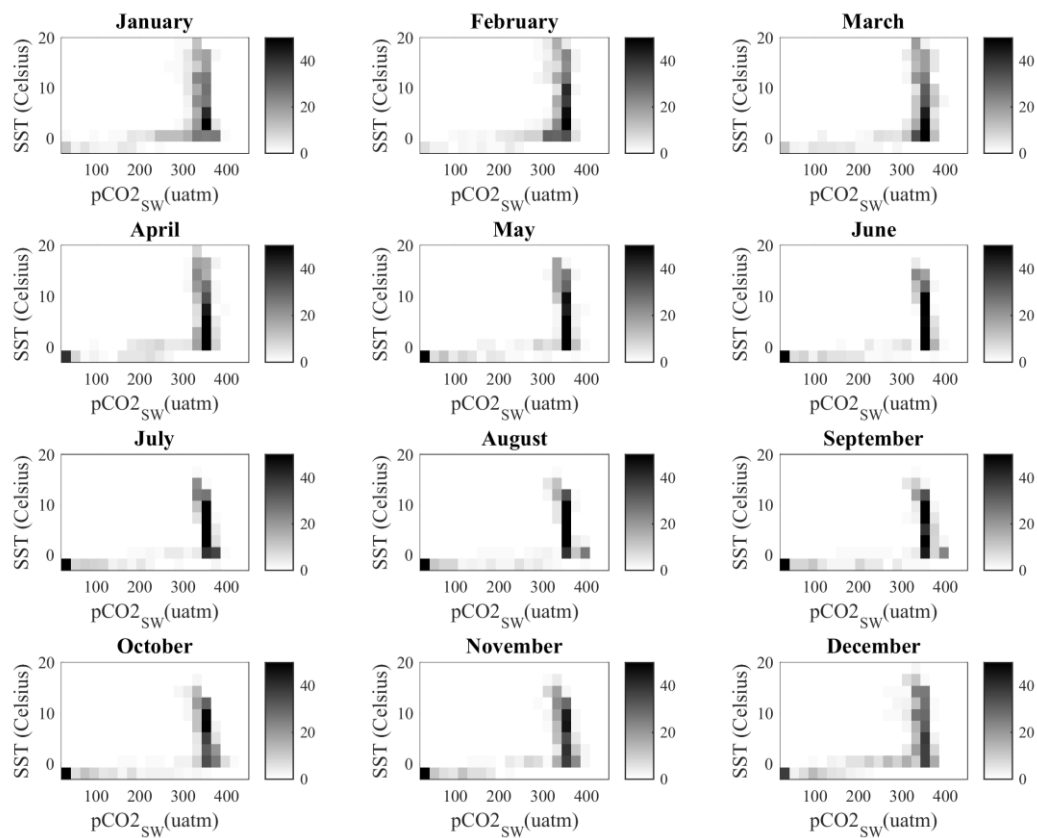
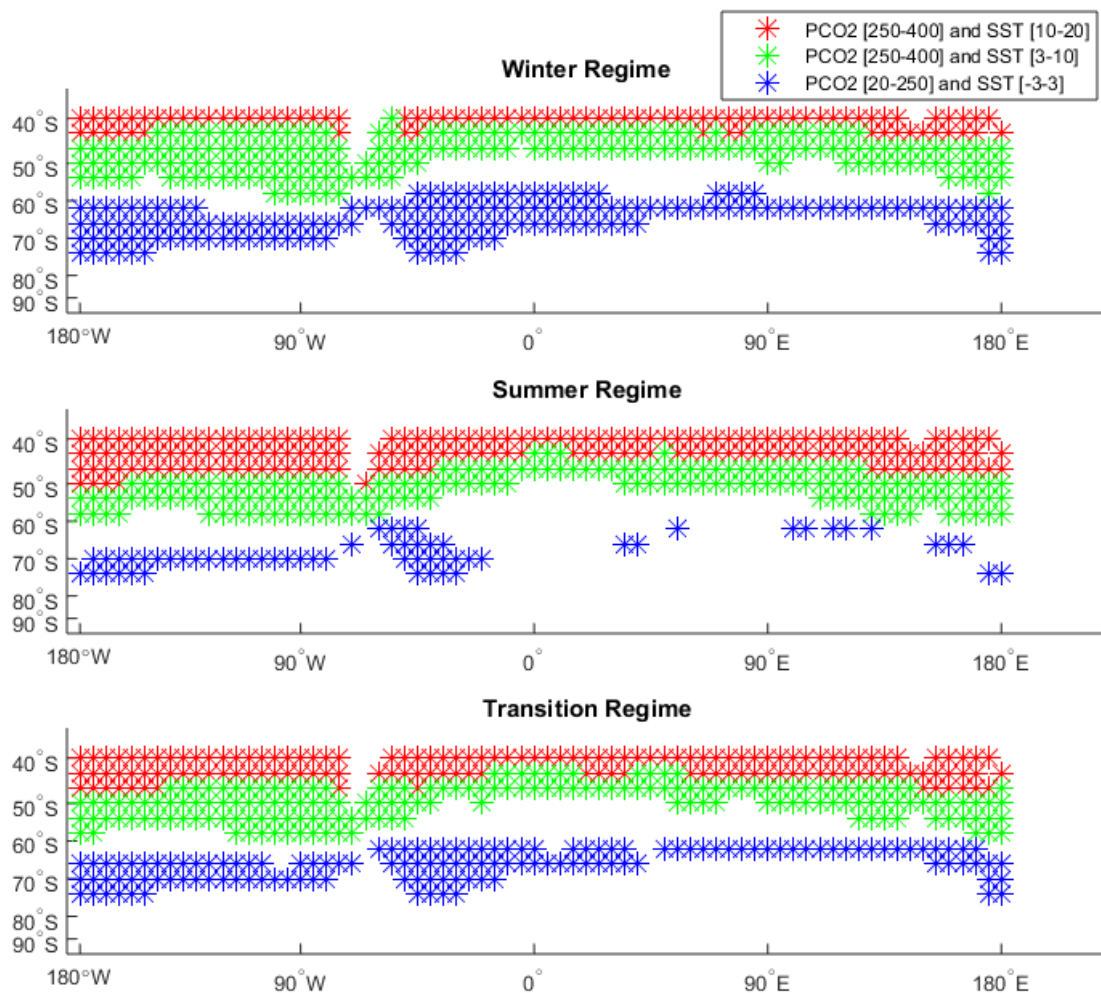


Fig. S4: Scatter diagrams and linear fits of  $pCO_{2sw}$  with a) SST, b) salinity, c) wind speed and d) nitrate in each of the North Atlantic regions of each regime.

## S4.2 The Southern Ocean Carbon States

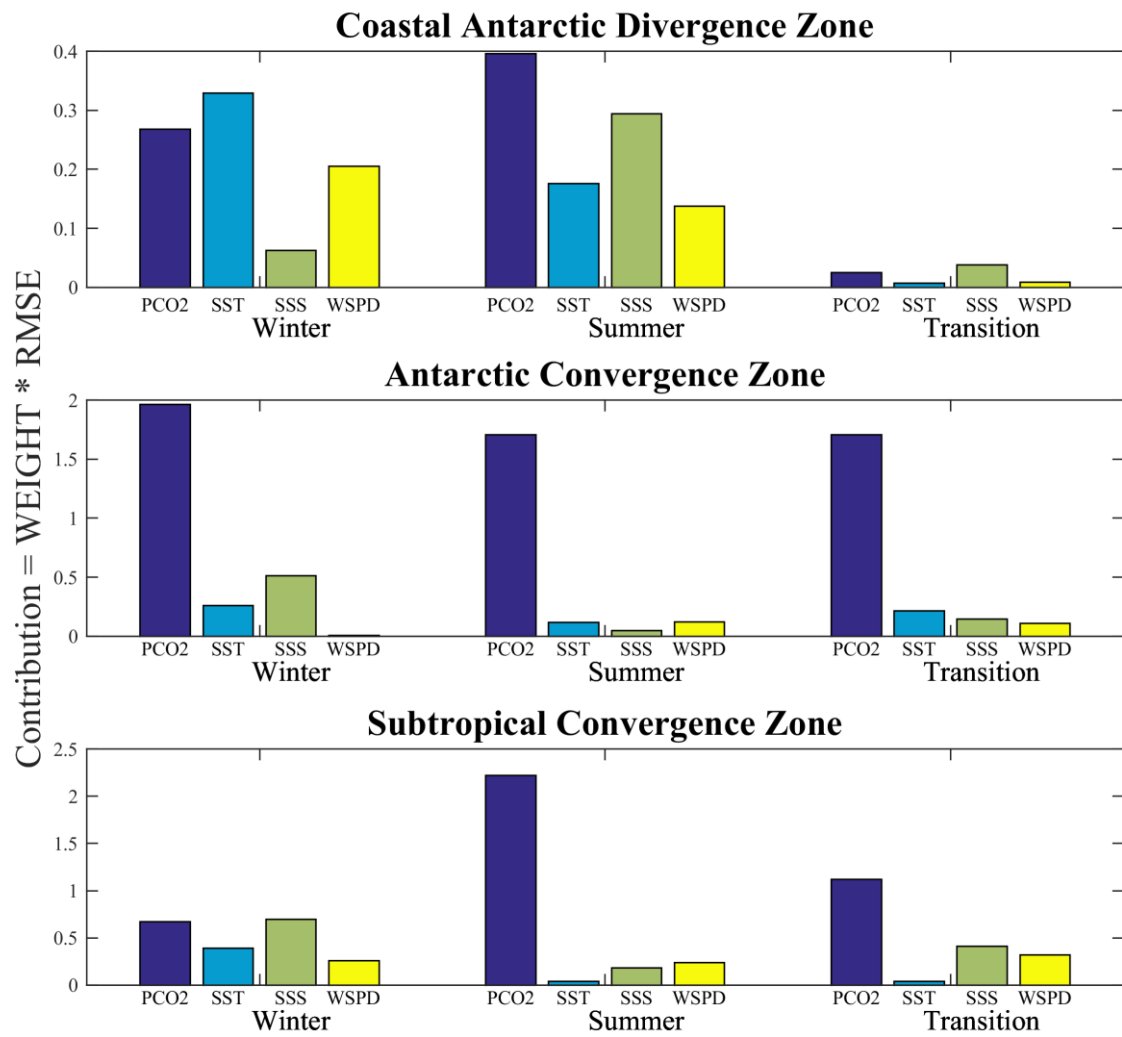


**Fig. S5: Monthly 2D histograms of pCO<sub>2</sub> of surface water (pCO<sub>2</sub>sw) and SST in the Southern Ocean (defined as 180°W to 180°E, 90°S to 40°S) from the Takahashi observational dataset.**

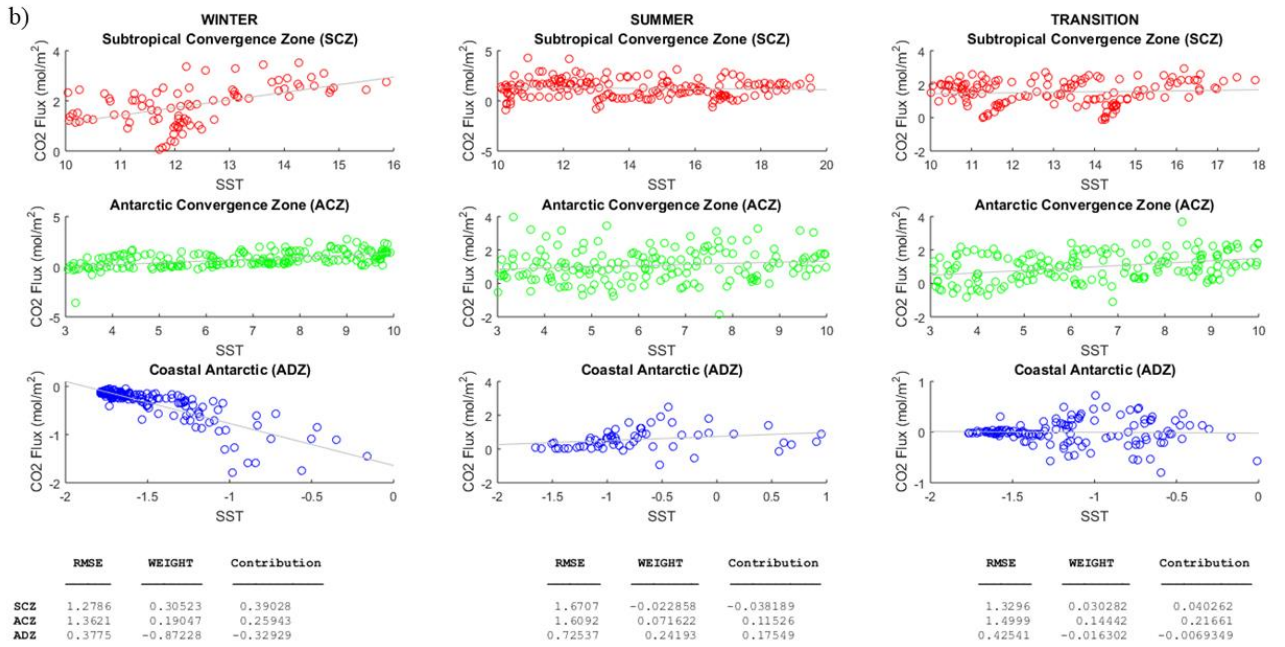
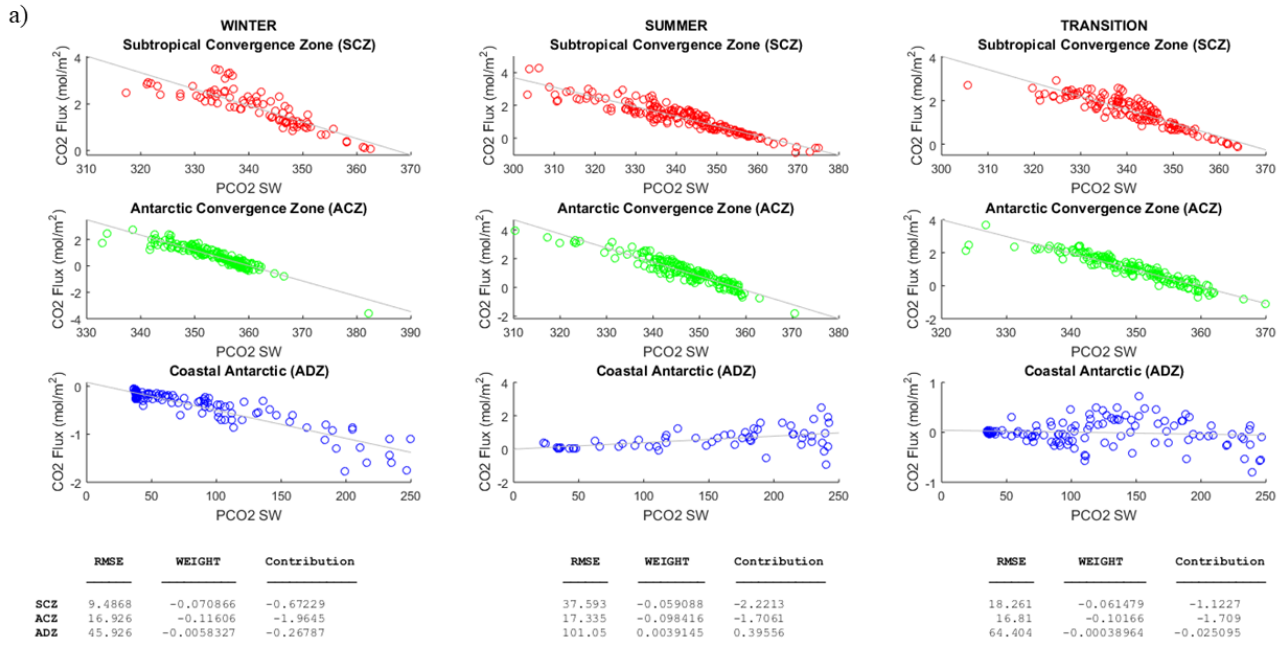


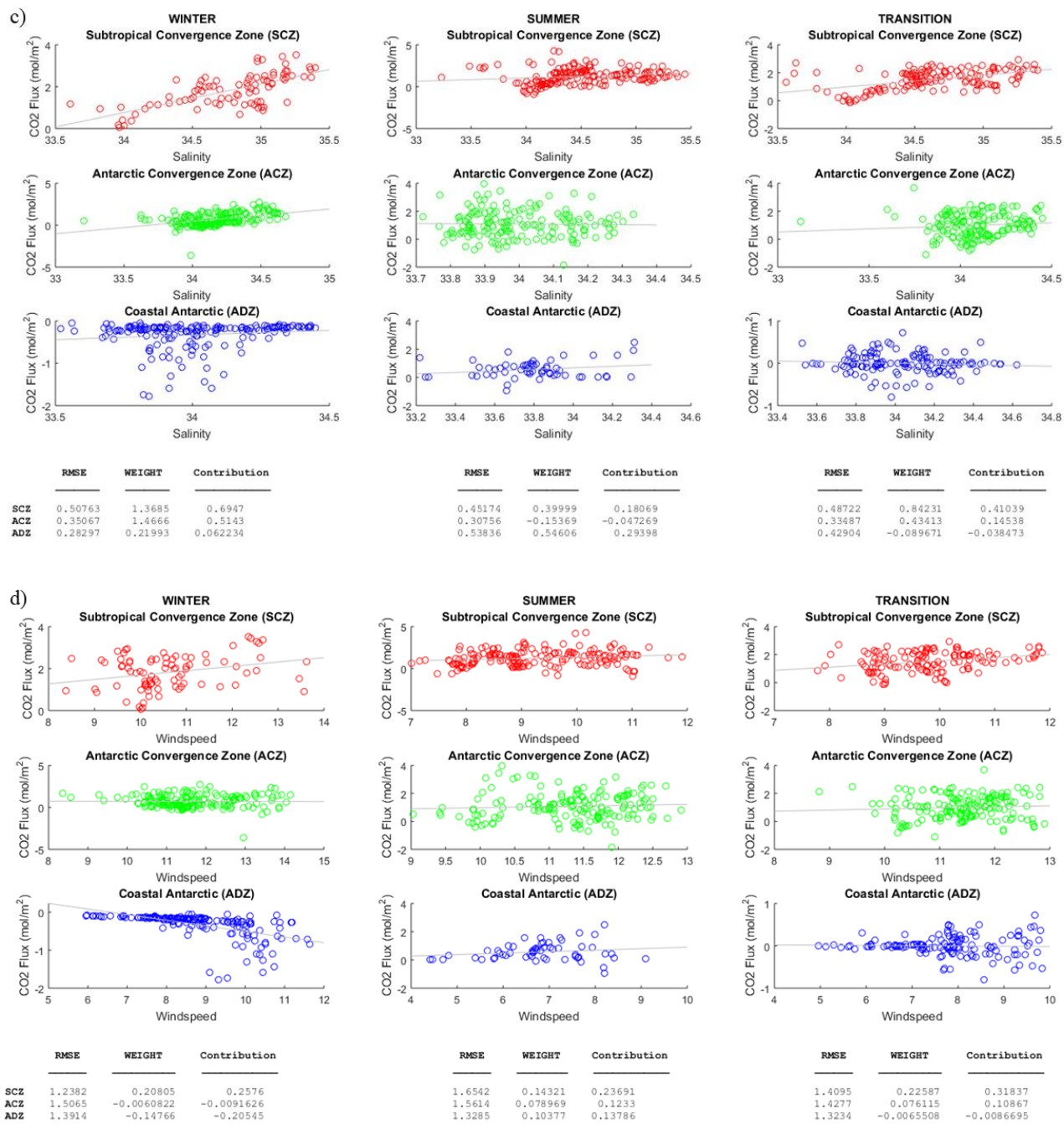
**Fig. S6: Demarcated regions within the Southern Ocean basin defined by ranges of pCO<sub>2</sub><sub>sw</sub> and SST values. The choice of the regions is based on the dominant bins in each ocean carbon state.**



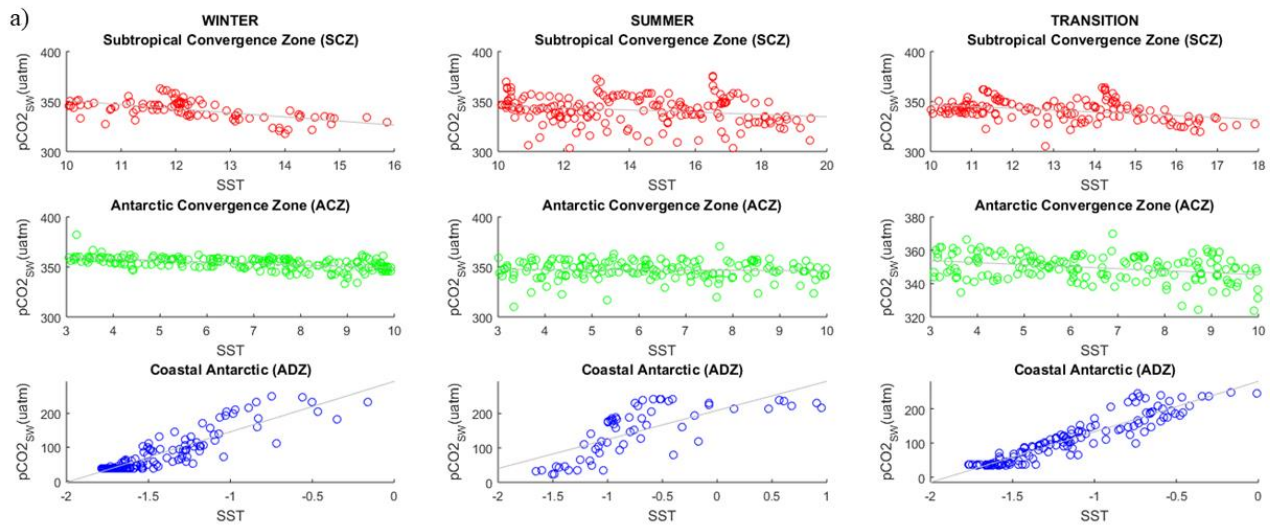


**Fig. S7: Bias terms as computed in the Taylor expansion of the model bias for the air-sea flux of CO<sub>2</sub>.**





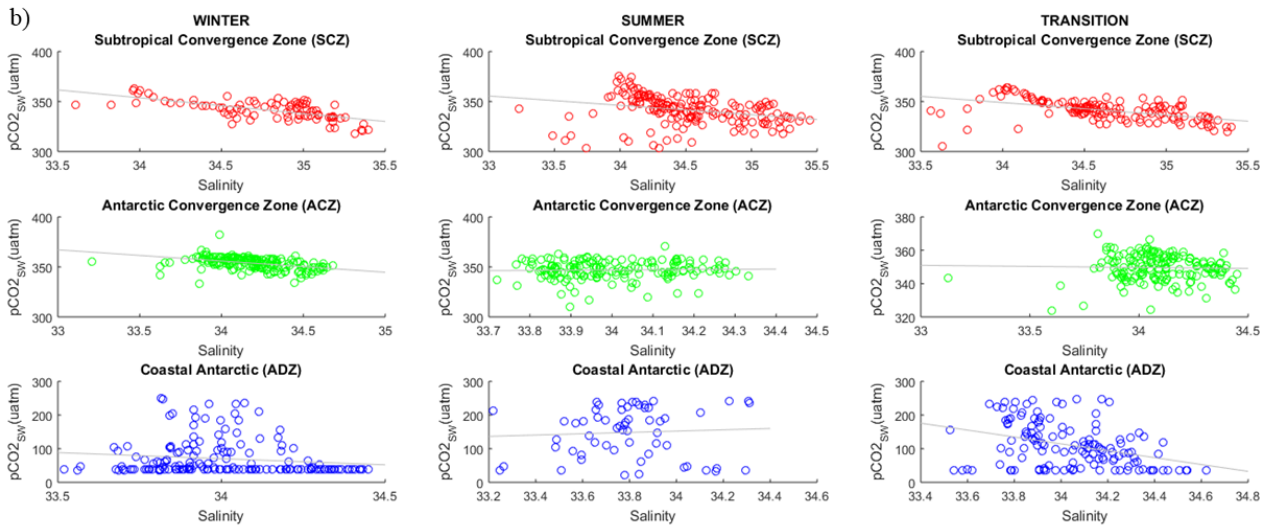
**Fig. S8: Scatter diagrams and linear fits of the air-sea flux of CO<sub>2</sub> with a) pCO<sub>2sw</sub>, b) SST, c) salinity, and d) wind speed in each of the Southern Ocean regions and for each regime.**



|     | 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       |



|     | 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 |
|-----|---------|---------|--------------|
| SCZ | 0.45174 | -9.3632 | -4.2298      |
| ACZ | 0.30756 | 2.3781  | 0.73142      |
| ADZ | 0.53836 | 19.523  | 10.51        |

|     | RMSE    | WEIGHT  | Contribution |
|-----|---------|---------|--------------|
| SCZ | 0.48722 | -12.445 | -6.0635      |
| ACZ | 0.33487 | -1.2514 | -0.41907     |
| ADZ | 0.42904 | -102.26 | -43.873      |

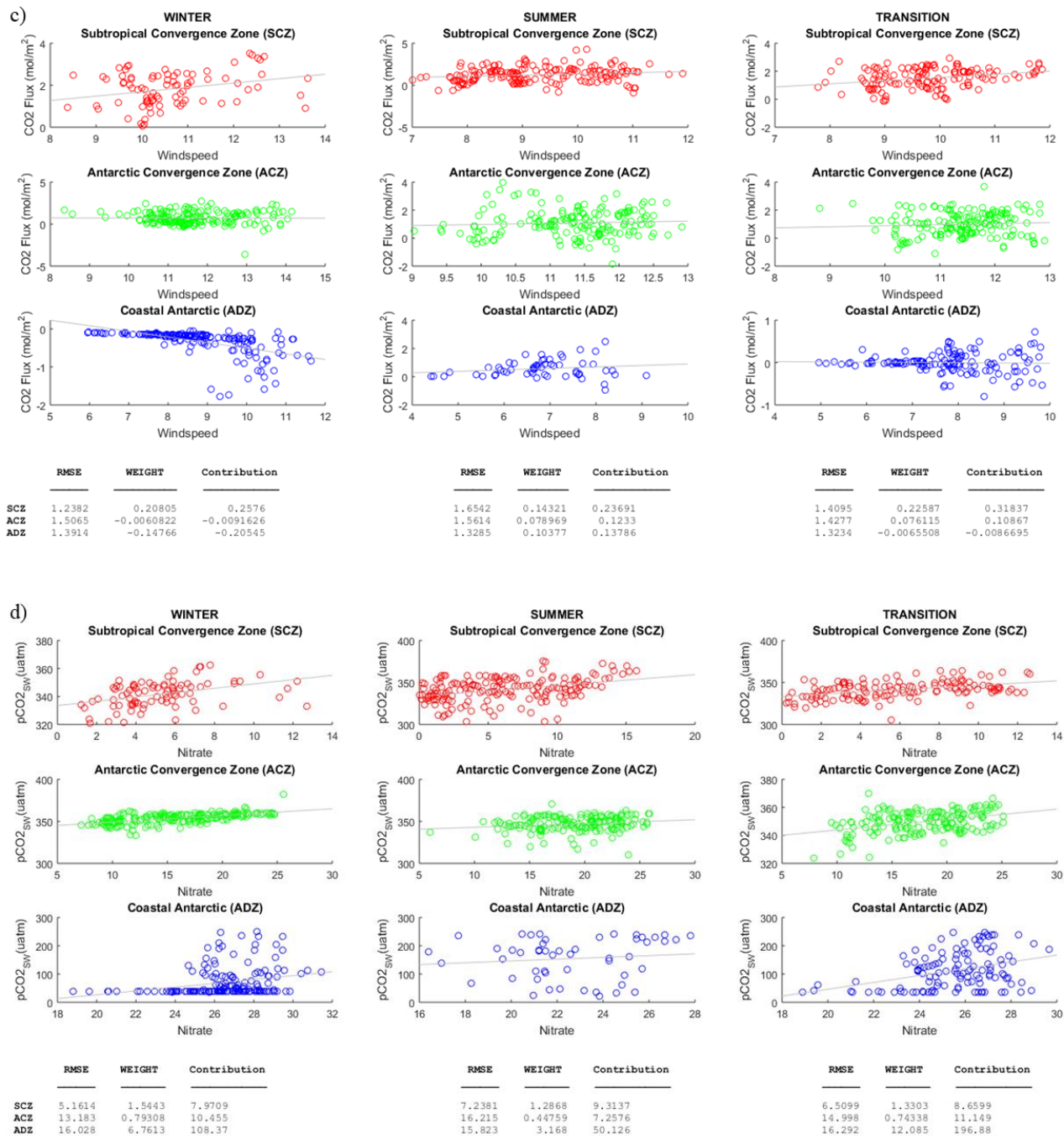


Fig. S9: Scatter diagrams and linear fits of pCO<sub>2sw</sub> with a) SST, b) salinity, c) wind speed and d) nitrate in each of the Southern Ocean regions and for each regime.