Extended Methods

S. E. Mikaloff Fletcher,[1] N. Gruber,[1] A. R. Jacobson,[2] S. C. Doney,[3] S. Dutkiewicz,[4] M. Gerber,[5] M. Follows,[4] F. Joos,[5] K. Lindsay,[6] D. Menemenlis,[7] A. Mouchet[8] S. A. Müller[5] and J. L. Sarmiento[2]

1. Basis function simulations

For a model grid box at position i, j and time, t, the surface flux in units of mass or moles per unit time, $F(i,j,t)_{dye}$, is equal to

$$F(i,j,t)_{dye} = F_{tot} \cdot P_{flux}(i,j,t_{seas}) \cdot \phi(t). \quad (1)$$

The arbitrary unit flux, F_{tot}, is distributed within the model region by a dimensionless, seasonal spatial pattern, $P(i,j,t_{seas})$, and scaled by the anthropogenic perturbation to atmospheric CO_2, $\phi(t)$.

The spatial pattern is determined from the seasonal climatology of Takahashi et al. [2002] extrapolated to areas not covered by the observations without using a sea-ice mask. Previous ocean inversion studies have found that the inverse estimates are relatively insensitive to the choice of the spatial pattern (Gloor et al., 2001).

2. Region aggregation and discussion of the covariance matrix

Basis functions were originally generated for 30 surface regions (Mikaloff Fletcher et al., 2003), and later aggregated to 24 regions. The regions that were aggregated were combined prior to the inversion by weighting their corresponding basis functions by their relative fluxes in the Takahashi et al. [2002] CO_2 climatology. The inversion has been tested for biases associated with the aggregation to 24 regions by comparing the 24 region inversion with a 30 region inversion where the inverse estimates are then summed to the 24 regions using three of the ten contributing models, NCAR, PRINCE-LL, and PRINCE-HH. The differences between the pre- and post- aggregated inverse estimates are less than 0.10 Pg C yr$^-$1 regionally and are also less than the reported error except in a few cases where both the reported error and aggregation differences are less than 0.03 Pg C yr$^-$1.

These aggregations were selected to minimize the covariance between the modeled response to surface fluxes into each pair of regions, while maintaining low residuals between the observationally-based and inverse anthropogenic carbon concentration estimates. A matrix of regional covariances was calculated from the matrices produced by the singular value decomposition, where the diagonal elements represent the square of the random error for each flux estimate and the off-diagonal elements represent the co-variances between model regions.

The covariance matrix for the 24 region aggregation generally has small off-diagonal elements for all of the models relative to the variances (shown in Figure 4 of the online supplement), indicating that the errors due to The covariance matrix for the 24 region aggregation generally has small off-diagonal elements for all of the models relative to the variance, indicating that the errors due to correlations between regions are smaller than the random errors. Some models have high covariances between the Arctic Ocean (Region 1) and the North Atlantic High Latitudes (Region 2). We chose not to aggregate these regions because the partitioning of anthropogenic carbon uptake between these two regions is of particular scientific interest. The diagonal elements of the covariance matrix indicate that the random errors are on the order of 0.01 Pg C yr$^-$1. This low random error estimate is due to the very large number of observations constraining the flux estimates. However, this does not take into account systematic errors, which are addressed in Section 4 of the manuscript.

3. CFC skill score

In order to consider which models are likely to have the most accurate transport on the time scale of anthropogenic carbon perturbation, we compare the GLODAP gridded CFC-11 data set with forward simulations of CFC-11 from OCMIP (Dutay et al., 2002). Table 1 of the manuscript shows the correlation between the gridded CFC-11 data and the modeled CFC-11, R, the standard deviation of the modeled CFC-11 normalized by the standard deviation of the gridded CFC-11 data, $\hat{\sigma}$, and a CFC-11 model skill score based on these two quantities, S (Taylor, 2001).

$$S = \frac{4(1+R)^4}{(\hat{\sigma} + 1/\hat{\sigma})^2 (1+R_o)^4} \quad (2)$$

Where R_o represents the maximum attainable correlation attainable. This formula provides a skill score between one and zero, with 1 representing a perfect model representation of the observations. R_o was calculated based on the

[1]Department of Atmospheric and Oceanic Sciences and the Institute for Geophysics and Planetary Physics, University of California, Los Angeles, California, USA.
[2]Atmospheric and Oceanic Sciences Department, Princeton, New Jersey, USA.
[3]Marine Chemistry and Geochemistry, Woods Hole Oceanographic Institution, Woods Hole, MA, USA.
[4]Department of Earth, Atmosphere, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA.
[5]Climate and Environmental Physics, Physics Institute, University of Bern, Bern, Switzerland.
[6]Climate and Global Dynamics Division, National Center for Atmospheric Research, Boulder, Colorado, USA.
[7]Estimating the Circulation and Climate of the Ocean, Jet Propulsion Laboratory, Pasadena, California, USA.
[8]Astrophysics and Geophysics Institute, University of Liege, Belgium.

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correlation between the GLODAP gridded data set and the gridded data set plus the estimated error due to griding the data. The choice of R_o is critical in determining the absolute skill of a model; however, it is of limited importance when evaluating the relative skill of a variety of different models as in this study.

In the manuscript, the skill-weighted cross-model mean refers to a mean of the quantity, X, of the *nmod* models weighted according to their model skill scores, S_i.

$$\bar{X} = \frac{\sum_{i=1,nmod} S_i * X_i}{\sum_{i=1,nmod} S_i} \quad (3)$$

Similarly, the weighted standard deviation refers to the standard deviation of the models weighted by the skill score.

$$stdev_{wt} = \sqrt{\frac{\sum_{i=1,nmod} S_i * (X_i - \bar{X})^2}{(\sum_{i=1,nmod} S_i)(nmod - 1)}} \quad (4)$$

4. Construction of scenarios that test possible biases in the data-based anthropogenic carbon estimates

Matsumoto and Gruber [2005] examined the accuracy of the anthropogenic carbon estimates by applying the ΔC^* method to a forward model simulation with known anthropogenic carbon concentrations. The authors found substantial biases in the ΔC^* method due to the time evolution of the air-sea disequilibrium, biases in the pCFC ventilation age and errors in identifying water masses which contribute to a given water parcel. As a result, they found that the ΔC^* method tends overestimate the anthropogenic carbon inventory in shallower waters by about 10% and underestimate it in deeper waters. Globally, the ΔC^* inferred anthropogenic carbon inventory was 7% larger than the true inventory.

It is not within the scope of this paper to re-assess the anthropogenic carbon dataset based on the findings of Matsumoto and Gruber [2005]; however, it is critical to address the effect these biases might have on the inverse estimates. To this end, we constructed a 'Matsumoto corrected' scenario, in which a hypothetical correction factor was applied to the data-based anthropogenic carbon estimates and these corrected anthropogenic carbon estimates were used in the inversion. In addition, two scenarios were constructed to assess the impact of a globally uniform shift in the oxygen to carbon remineralization ratio used to remove the effects of biology.

The hypothetical correction factor is a function of potential density. Below a given density threshold, σ_θ^0, the anthropogenic carbon concentration is decreased and above it the anthropogenic carbon concentration is increased.

$$C(\sigma_\theta) = A(ae^{(\sigma_\theta - \sigma_\theta^0)/k_1} - 1) - B(be^{-(\sigma_\theta - \sigma_\theta^0)/k_2}), \quad (5)$$

The correction factor, $C(\sigma_\theta)$, is the fractional correction to the data-based anthropogenic carbon estimates, σ_θ^0 is 27.4, and the constants A, a, B, b, k_1, and k_2 have been tuned based on the GLODAP gridded anthropogenic carbon data set such that the global anthropogenic carbon inventory is decreased by 7% and the anthropogenic carbon inventory at densities less than σ_θ^0 is decreased by 10% (Figure 10 of the online supplement).

In addition, two scenarios were constructed to assess the impact of biases in the stoichiometric ratio between carbon and oxygen, $r_{C:O_2}$. Gruber et al., [1996] estimated the data-based anthropogenic carbon for the Atlantic Ocean using high and low values of $r_{C:O_2}$, which were 0.780 and 0.596 based on the $r_{C:O_2}$ and uncertainty estimates Anderson and Sarmiento [1994]. In order to extrapolate these estimates to the entire ocean, we fit the high and low $r_{C:O_2}$ estimates of Gruber et al. [1996] in Atlantic to AOU and the anthropogenic carbon concentration using multiple linear regression (coefficients shown in Table 3 of the online supplement).

$$\sigma = \alpha AOU + \beta C_{ant} + \gamma. \quad (6)$$

where α, β, and γ are optimized in the regression and σ is the difference between the standard anthropogenic carbon estimates and the anthropogenic carbon based on high or low $r_{C:O_2}$ estimates.