Supporting Information for ”Annually-resolved propagation of CFCs and SF_6 over eight decades”

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Introduction

The supplementary information in Text S1 reports the details of how to construct and solve the modified, non-negative least squares problem to deconvolve the discrete Green's function **G** (together with the information reported in Section 3.2 of the main manuscript).

Supplementary Figures S1 and S2 also provide additional details about the Time-Correction Method, in particular about the decisions made in this study to calculate the surface boundary conditions (Figure S1) and the first guess Green's function (Figure S2).

Figures S3 and S4 report the reconstructed CFC-12 and SF_6 concentrations in 2021 for the hydrographic sections A16-A23, P16 and I8I9. The reconstructed CFC-11 concentrations are shown in the main manuscript (Figure 4).

Figure S5 shows the vertically-integrated CFC-11 concentration reconstructed with the TCM in 1994 and 2020. The 1994 year was chosen for a direct comparison to the CFC-11 inventory by Willey et al. (2004). Note that our product is not gridded on a uniform horizontal and vertical grid, such as the CFC-11 inventory by Willey et al. (2004); as such, in the vertical integration of the reconstructed CFC-11 we examine only the locations where observations describe at least 66% of the water column. This choice explains the lack of data in the southwest Indian Ocean, in the Atlantic sector of the Southern Ocean and partially in the east Pacific Ocean.

Text S1. To solve the least squares problem to find $\mathbf{G}$, at each location we define:

$$\hat{\mathbf{C}} = \left[\begin{array}{c} \mathbf{C}_1 \\ \hline \mathbf{C}_2 \\ \hline \vdots \\ \hline \mathbf{C}_{N_t} \\ \hline \mathbf{G}_0 \end{array}\right], \ \hat{C}_s = \left[\begin{array}{c} C_{s1} \\ \hline C_{s2} \\ \hline \vdots \\ \hline C_{sN_t} \\ \hline \mathbf{I} \end{array}\right], \ \hat{W}^{-1} = \left[\begin{array}{c|c|c|c|c} W_1^{-1} & 0 & 0 & 0 & 0 \\ \hline 0 & W_2^{-1} & 0 & 0 & 0 \\ \hline 0 & 0 & \ddots & 0 & \\ \hline 0 & 0 & 0 & W_{N_t}^{-1} & 0 \\ \hline 0 & 0 & 0 & 0 & S^{-1} \end{array}\right].$$

N_t is the number of tracers used (depending on the available observations), and $\mathbf{C}_i$ represents the array of observations available at one location in the ocean interior for each tracer. The C_{si} matrix returns the surface boundary conditions per each tracer, while W_i^{-1} and S^{-1} indicate the weight matrices for the tracer observations and the Green's function first guess $\mathbf{G}_0$, respectively. We re-arrange the terms reported above to use the non-negative least-squares method of Lawson and Hanson (1995), defining

$$\begin{aligned} \hat{\hat{\mathbf{C}}} &= \hat{W}^{-1/2}\hat{\mathbf{C}} \\ \hat{\hat{\mathbf{C}}}_s &= \hat{W}^{-1/2}\hat{\mathbf{C}}_s \end{aligned} \tag{1}$$

which we can use to rewrite the cost function as

$$J = (\hat{\hat{\mathbf{C}}} - \hat{\hat{C}}_s\mathbf{G})^T(\hat{\hat{\mathbf{C}}} - \hat{\hat{C}}_s\mathbf{G}), \tag{2}$$

which is equivalent to solving the equation $\hat{\hat{C}}_s\mathbf{G} = \hat{\hat{\mathbf{C}}}$, going back to the canonical form of a least-squares problem.

References

Holte, J., Talley, L. D., Gilson, J., & Roemmich, D. (2017). An argo mixed layer climatology and database. *Geophysical Research Letters*, *44*(11), 5618–5626.

Lawson, C. L., & Hanson, R. J. (1995). *Solving least squares problems.* SIAM.

Willey, D. A., Fine, R. A., Sonnerup, R. E., Bullister, J. L., Smethie Jr, W. M., & Warner, M. J. (2004). Global oceanic chlorofluorocarbon inventory. *Geophysical Research Letters*, *31*(1).

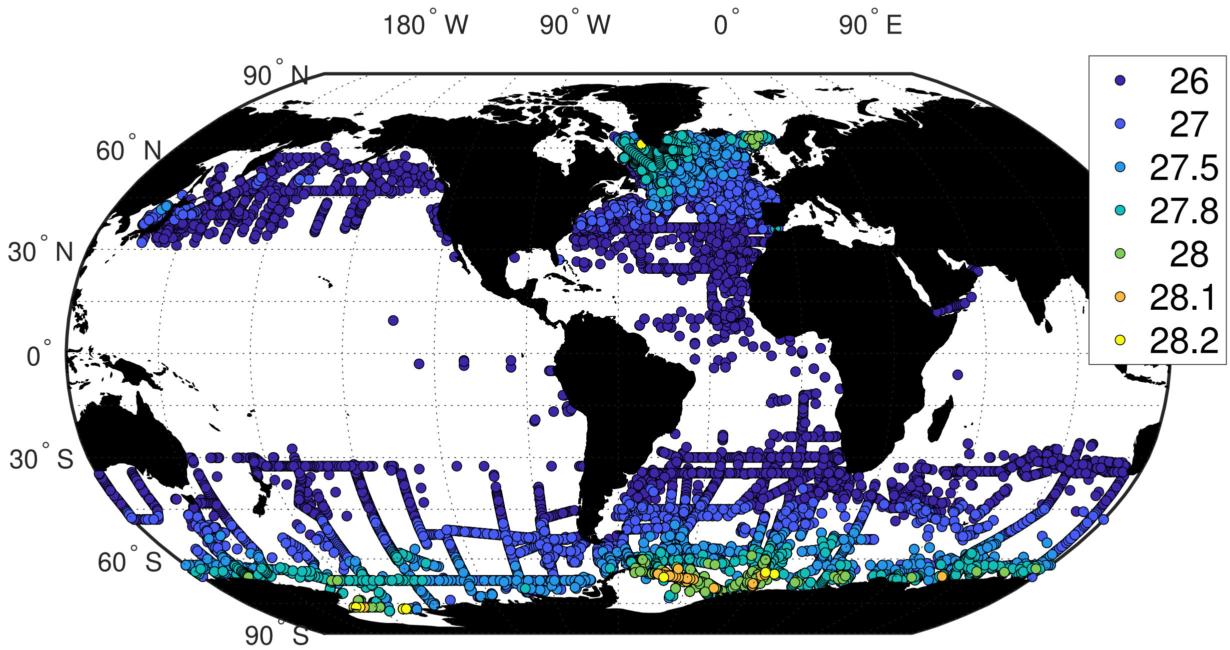

Figure S1. Outcrop points for the neutral density levels indicated in the legend. Outcrop points are defined as locations where the maximum mixed layer depth (from Holte et al. (2017)) is found at a density larger than a specific density level.

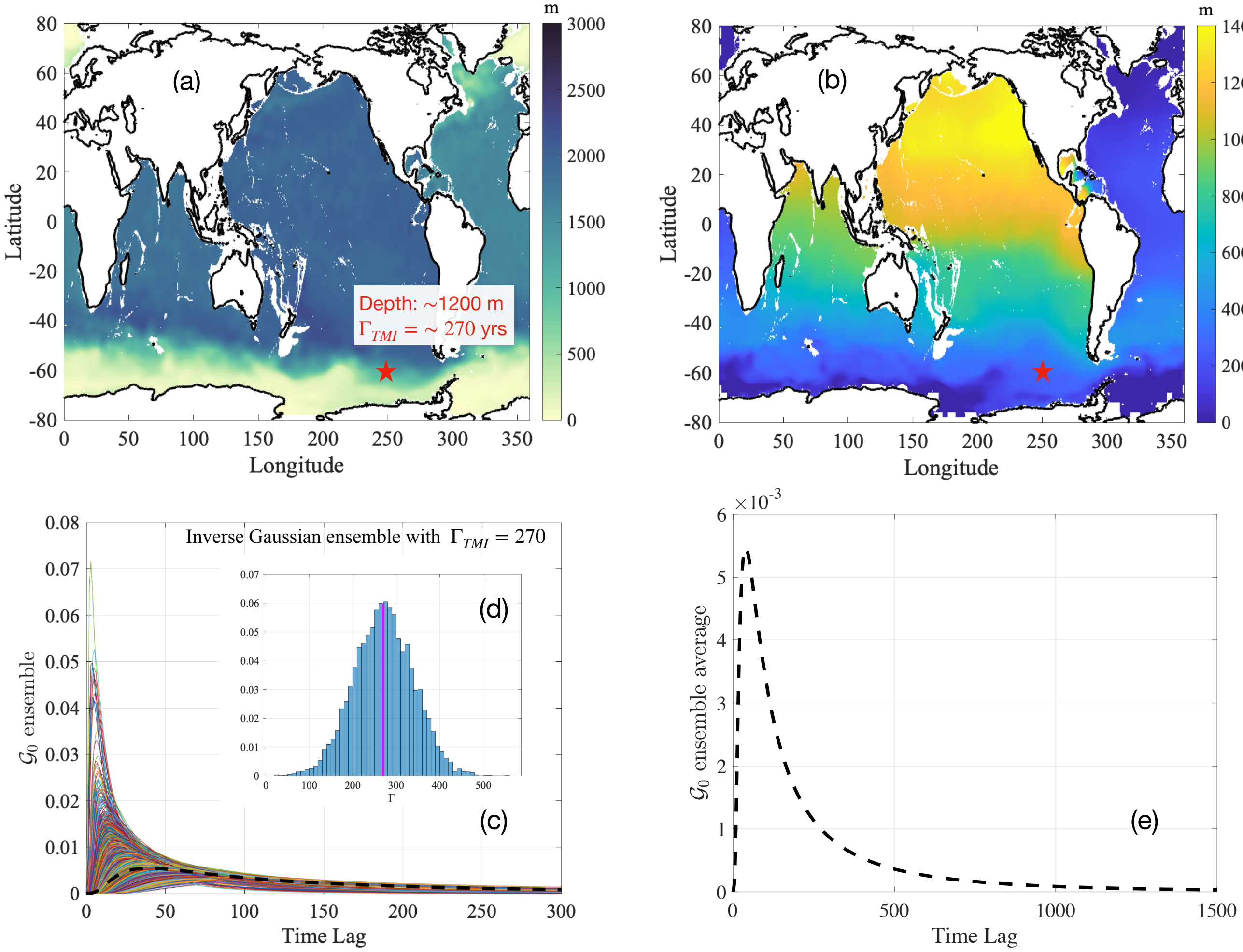

Figure S2. Steps to define the first guess $\mathbf{G}_0$: example at 150°W, 60°S on neutral density surface $\gamma^n = 27.90$ (red star in panels a, b). For each location, we find the closest TMI water age estimate. (a)-(b) Depth and Γ_{TMI} of the density surface. At the starred location, $\gamma^n = 27.90$ is about 1200 m deep and the TMI water age estimate is $\Gamma_{TMI} = 270$ years. (c) Ensemble of Inverse Gaussians with age within the range $\Gamma_{TMI}/2$: Γ_{TMI} x2 (see subset histogram in panel (d)). The ensemble average returns the kernel first guess used here, $\mathbf{G}_0$ (indicated in panel d, and shown in panel e).

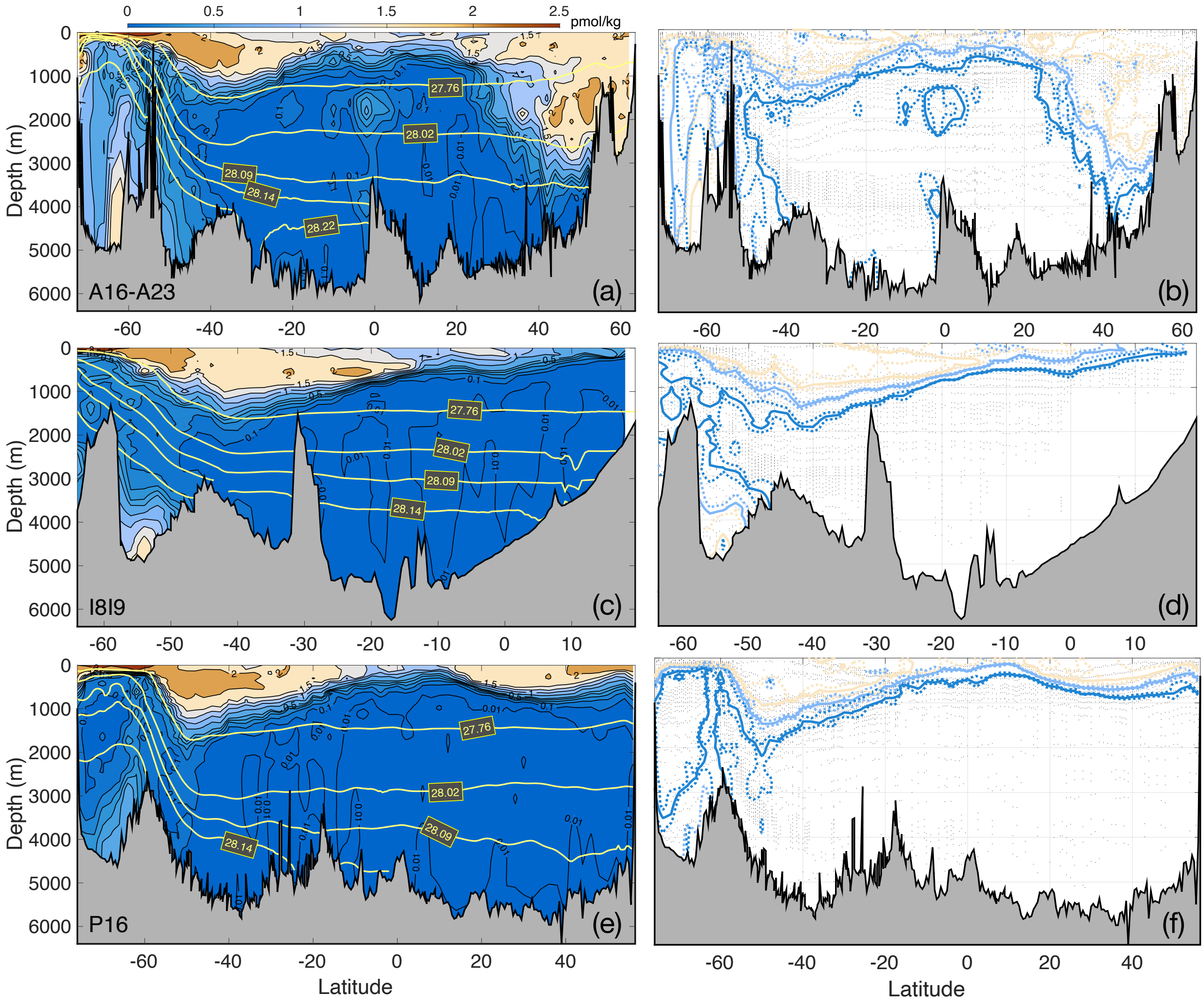

Figure S3. Reconstructed CFC-12 concentrations along A16+A23 (panels a,b), I8I9 (c,d), and P16 (e,f) in 2021. Panels in left column show the reconstructed concentration and neutral density contours (yellow) along each section. Panels in right column show the observation locations throughout all decades of available data (gray dots) and the uncertainty of the time corrected CFC-12 concentrations for three contours (0.2, 0.75 and 1.5 pmol/kg). The solid line shows our best estimate, while the dotted lines show the 95% confidence limits.

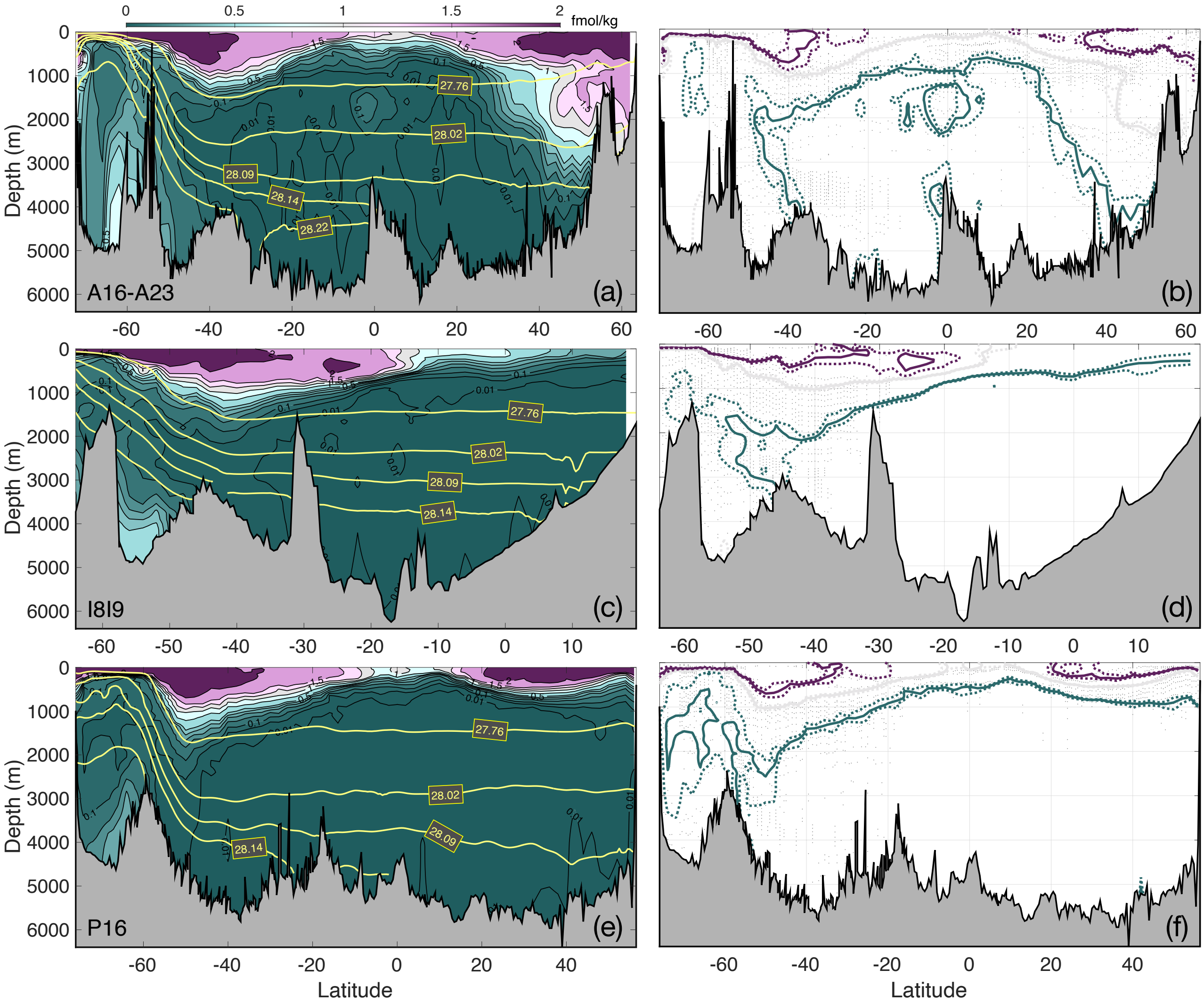

Figure S4. Same as in Figure S3 but for SF_6. The uncertainty of the time corrected SF_6 concentrations is shown for the contours 0.05, 1 and 2 fmol/kg.

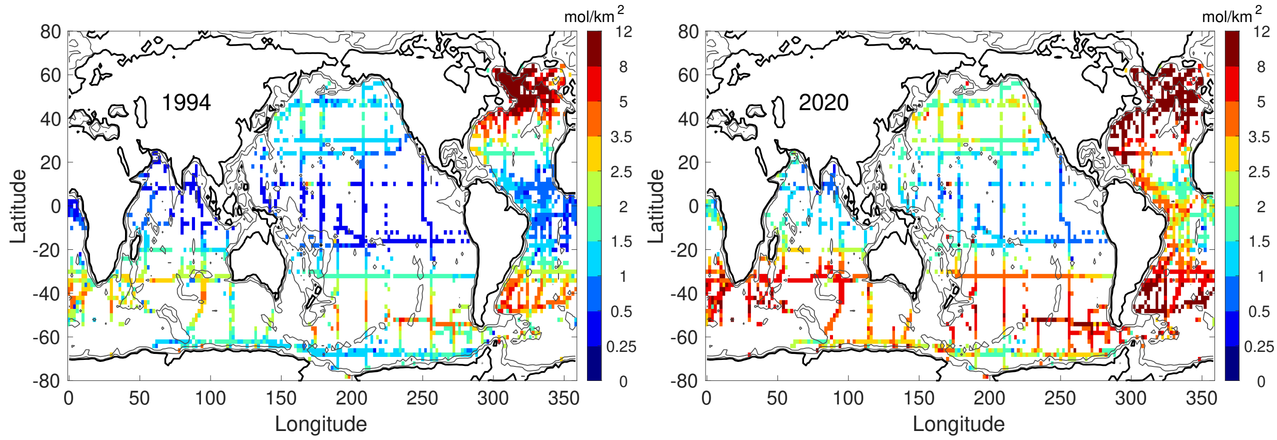

Figure S5. Vertically-integrated CFC-11 concentrations in 1994 (which can be compared with the inventory presented in Willey et al. (2004)) and 2020.