



*Supplement of*

**A 1-D examination of decadal air–sea re-equilibration induced ocean surface anthropogenic CO<sub>2</sub> accumulation: present status, changes from 1960s to 2000s, and future scenarios**

**W.-D. Zhai and H.-D. Zhao**

*Correspondence to:* W.-D. Zhai (wdzhai@126.com)

The supplementary materials include:

Figure S1. The selected wintertime month and the corresponding mixed layer depth distribution.

Figure S2. Distributions of wintertime surface  $p\text{CO}_2$  and salinity normalized TALK.

Figure S3. Wintertime sea surface distributions of salinity normalized DIC and Revelle factor.

Figure S4. Wintertime sea surface TALK versus salinity and the salinity normalized TALK versus temperature.

Figure S5. Global distribution of wintertime air-sea re-equilibration time of  $\text{CO}_2$ .

References

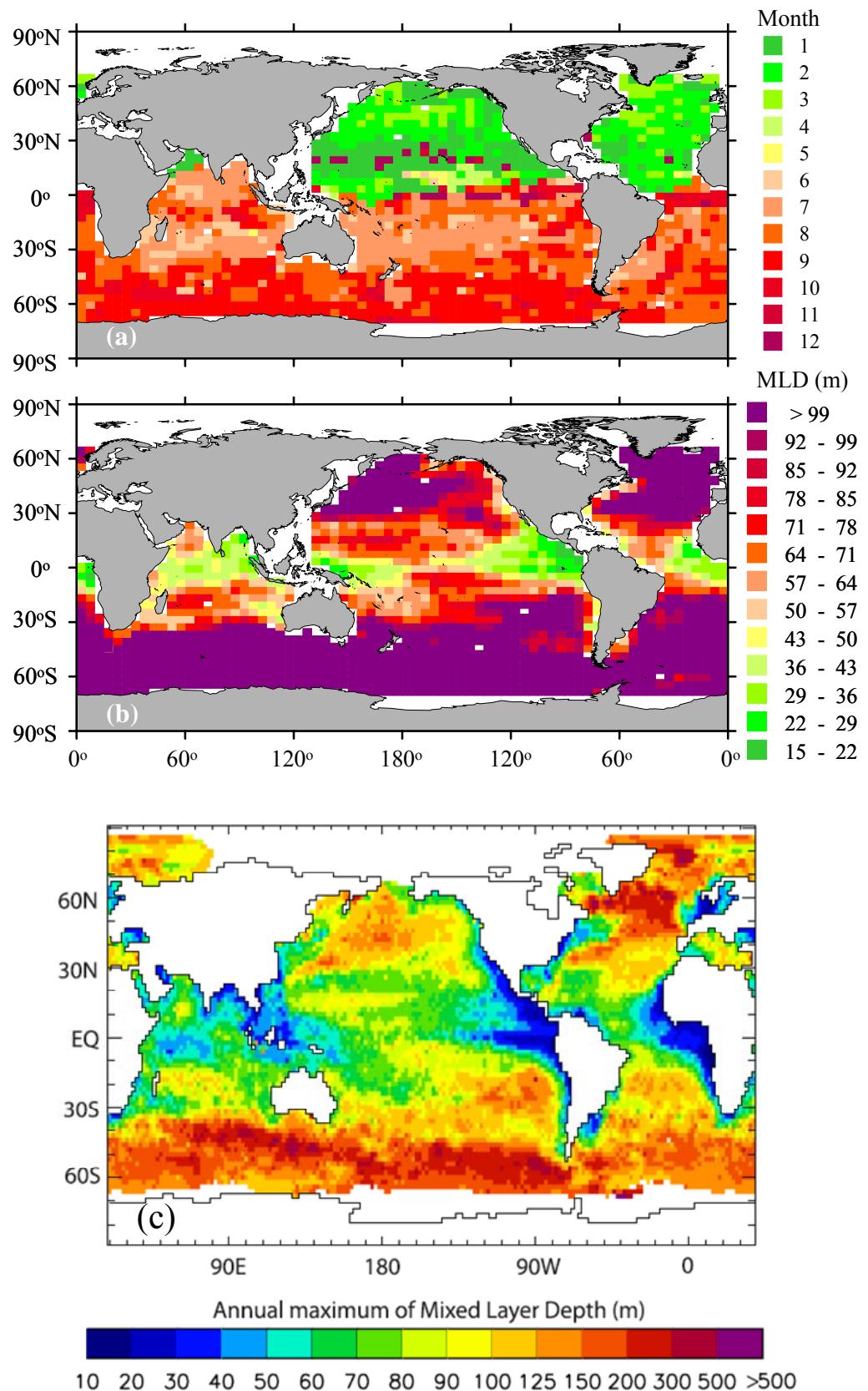


Figure S1. The selected wintertime month (a) and the corresponding mixed layer depth (b) in the  $4^{\circ}$  (latitude)  $\times 5^{\circ}$  (longitude) grid boxes under study, based on the  $2^{\circ} \times 2^{\circ}$  dataset released by de Boyer Montégut et al. (2004). Panel (c) is the maximum annual mixed layer depth map presented by de Boyer Montégut et al. (2004).

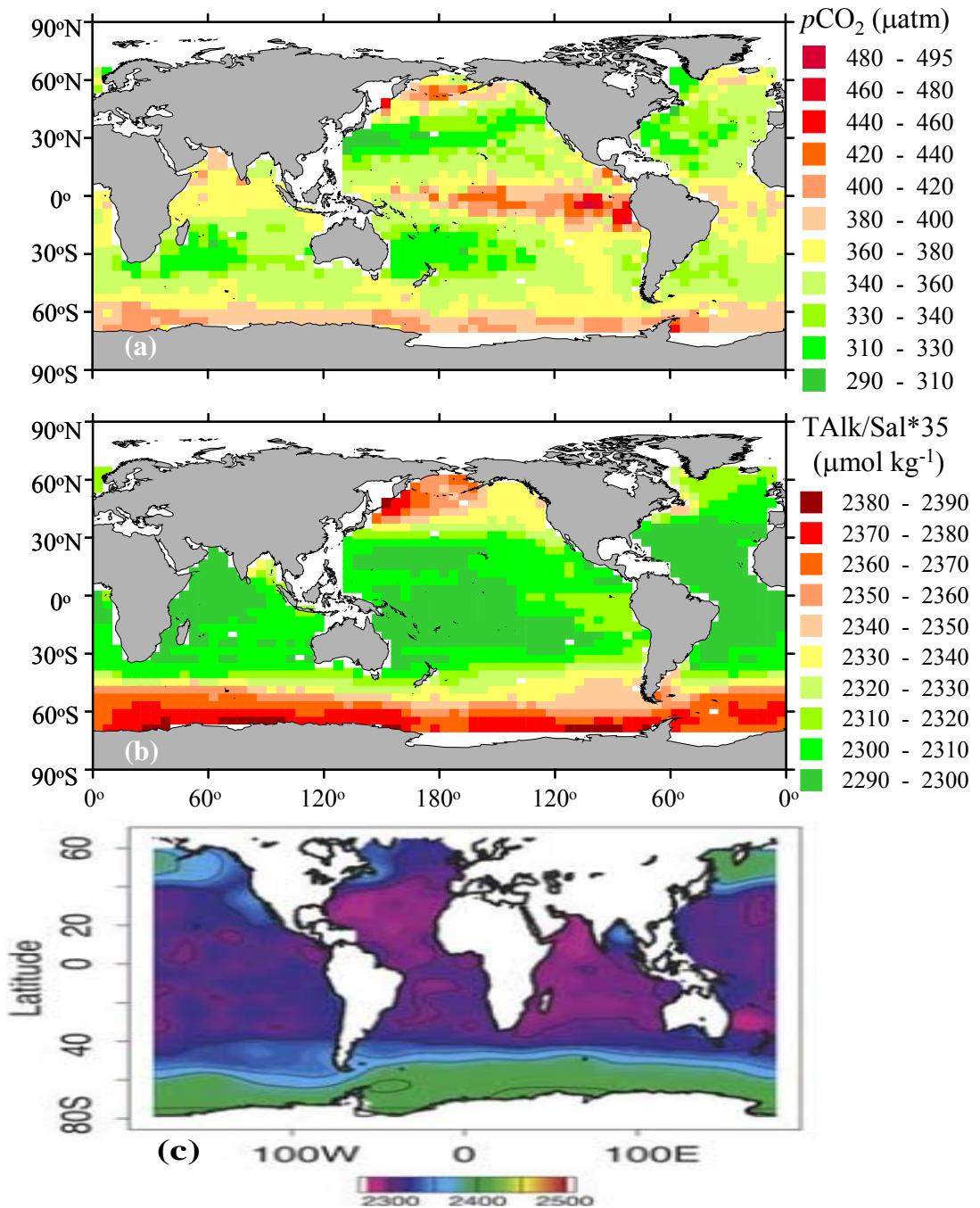


Figure S2. Wintertime sea surface  $p\text{CO}_2$  (a) and salinity normalized TALK (b) in the  $4^\circ$  (latitude)  $\times$   $5^\circ$  (longitude) grid boxes under study, corresponding to the selected months, as shown in Figure S1a. Sea surface  $p\text{CO}_2$  data are from Takahashi et al. (2009) for a reference year 2000 (Version October 2009). Following Lee et al. (2006), TALK is reconstructed using the sea surface temperature and salinity in the Takahashi et al. (2009) dataset. To eliminate the dilution and concentration effects of precipitation and evaporation on the seawater carbonate system, we normalized seawater TALK to a uniform salinity of 35. Note that our salinity normalized TALK distribution is very similar to the Key et al. (2004) climatological result of sea surface potential alkalinity ((TALK + Nitrate)  $\times$  35/Salinity) (c), since sea surface nitrate concentrations are usually low.

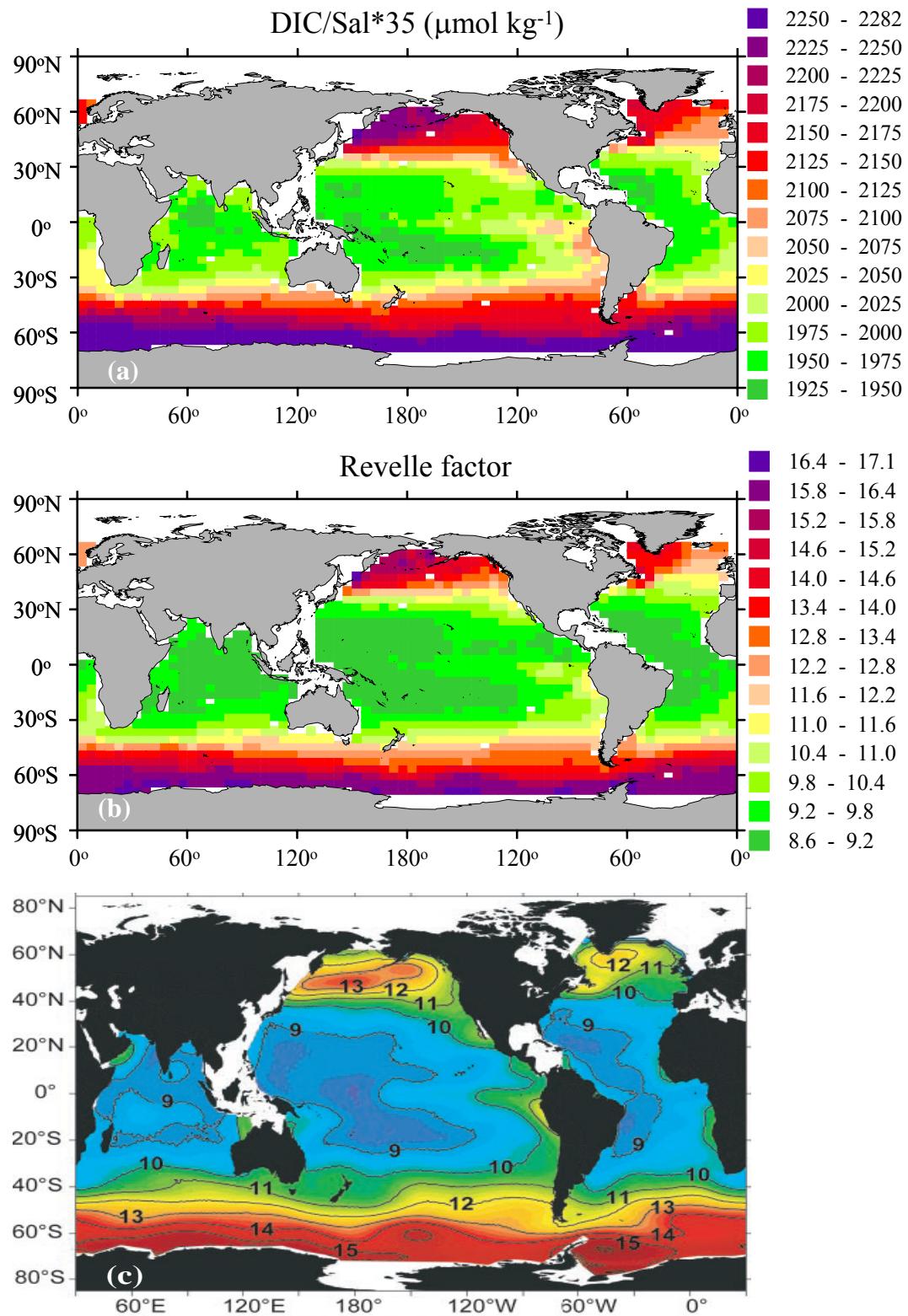


Figure S3. Wintertime sea surface distributions of salinity normalized DIC (a) and Revelle factor (b), calculated for a reference year 2000 from sea surface  $p\text{CO}_2$  and TALK data shown in Figure S2. To eliminate the dilution and concentration effects of precipitation and evaporation on the seawater carbonate system, we normalized seawater DIC to a uniform salinity of 35. The Revelle factor distribution (b) is quite similar to the 1994 result (c) presented by Sabine et al. (2004).

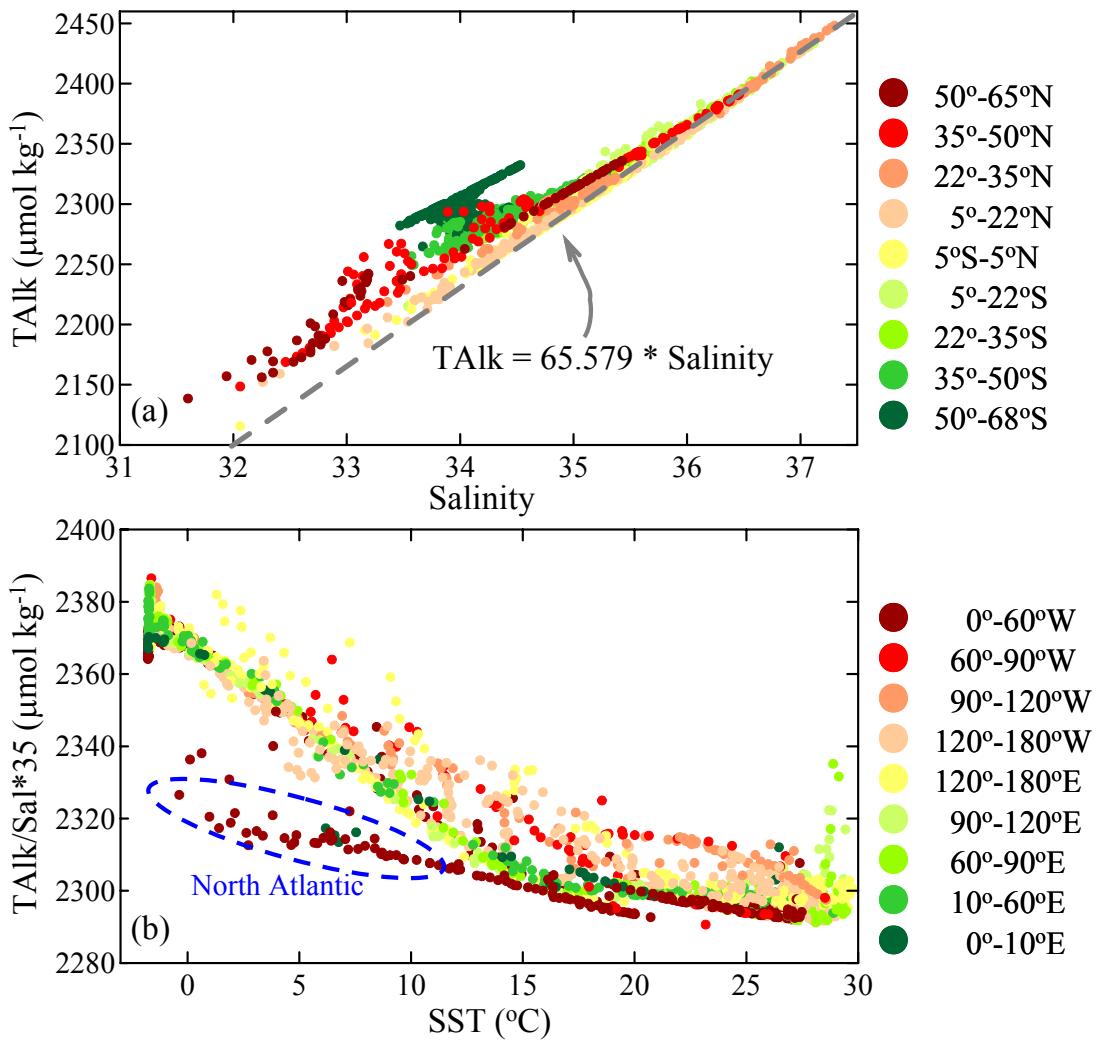


Figure S4. Wintertime sea surface TALK versus salinity (a) and the salinity normalized TALK (via  $TALK/\text{Sal}*35$ ) versus temperature (b) over the grid boxes under study. Colors of data points denote latitude (a) and longitude (b). Panel (a) shows that, surface TALK in low latitude areas is mainly controlled by evaporation induced condensing and/or precipitation induced dilution, while surface TALK has additional sources in mid- or high latitude areas. Panel (b) suggests that the excess TALK are mostly added from deep water upwelling, as indicated with very low temperature of  $<5^{\circ}\text{C}$  associated with high values of the salinity normalized TALK. Among those major cold-waters, the North Atlantic cold-waters have a relatively weak effect on the TALK addition, as shown in panel (b).

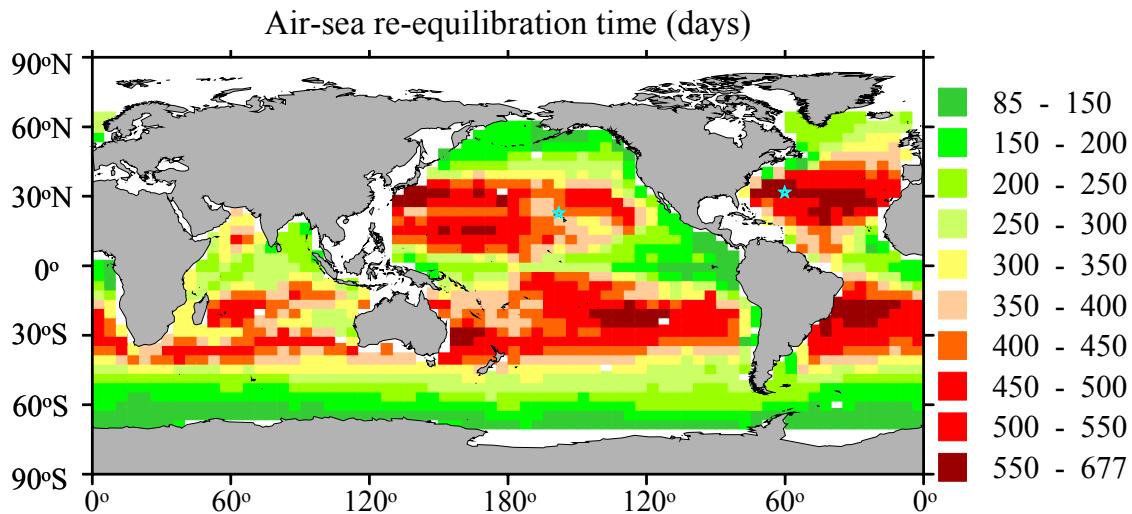


Figure S5. Global distribution of wintertime air-sea re-equilibration time ( $\tau$ ) of  $\text{CO}_2$ , estimated following the equation (1.6.102) in Zeebe and Wolf-Gladrow (2001), using the global mean gas transfer velocity of  $3.5 \text{ m d}^{-1}$  (Sweeney et al., 2007). Stars with the colour of cyan are sites of the Hawaii Ocean Time series program (HOT) and Bermuda Atlantic Time-series Study (BATS). Note that the estimated  $\tau(\text{CO}_2)$  at HOT is much lower than that at BATS.

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