Supplementary Information for

How will global carbon cycle respond to negative emissions?

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Taylor expansion for decomposing pCO2 variations

pCO_2 variations are composed by four terms: sea surface temperature (SST), dissolved inorganic carbon (DIC), alkalinity (ALK), and sea surface salinity (SSS). To assess the contributions of potential drivers, we use a Taylor expansion to decompose variations in pCO_2 after neglecting second-order terms, which was well established frame for quantify the contributions (Orr et al., 2022; Takahashi et al., 1993).

$$\Delta pCO_2 = \frac{\partial pCO_2}{\partial SST} \cdot \Delta SST + \frac{\partial pCO_2}{\partial DIC} \cdot \Delta DIC + \frac{\partial pCO_2}{\partial ALK} \cdot \Delta ALK + \frac{\partial pCO_2}{\partial SSS} \cdot \Delta SSS$$

As freshwater flux (FW) also modulates the changes in DIC and ALK, we need to separate the contributions of freshwater flux from carbonate systems by normalizing DIC $(sDIC = DIC \cdot S_0/S)$ and ALK $(sALK = ALK \cdot S_0/S)$ with reference salinity S_0 (Keeling et al., 2004; Wetzel et al., 2005).

$$\Delta pCO_2 = \frac{\partial pCO_2}{\partial SST} \cdot \Delta SST + \frac{\partial pCO_2}{\partial DIC}\left(\frac{S}{S_0}\right) \cdot \Delta sDIC + \frac{\partial pCO_2}{\partial ALK}\left(\frac{S}{S_0}\right) \cdot \Delta sALK + \frac{\partial pCO_2}{\partial FW} \cdot \Delta FW$$

After substituting salinity-normalized DIC (sDIC) and ALK (sALK), the equation can be rewritten with pCO_2 sensitivities γ, and their respective drivers in terms of four potential factors (SST, DIC, ALK, FW).

$$\Delta pCO_2 = (\gamma_{SST} \cdot pCO_2 \cdot \Delta SST) + \left(\gamma_{DIC} \cdot \frac{pCO_2}{DIC} \cdot \Delta sDIC\right) + \left(\gamma_{ALK} \cdot \frac{pCO_2}{ALK} \cdot \Delta sALK\right) + \left(\gamma_{FW} \cdot \frac{pCO_2}{FW} \cdot \Delta FW\right)$$

where sensitivities of individual drivers are defined as: $\gamma_{DIC} = \left(\frac{\partial pCO_2}{\partial DIC}\right) / \left(\frac{pCO_2}{DIC}\right)$, $\gamma_{ALK} = \left(\frac{\partial pCO_2}{\partial ALK}\right) / \left(\frac{pCO_2}{ALK}\right)$, $\gamma_{SST} = \frac{1}{pCO_2}\left(\frac{\partial pCO_2}{\partial ALK}\right) \simeq 0.0423\ ℃^{-1}$, $\gamma_{SSS} = \left(\frac{\partial pCO_2}{\partial ALK}\right)\left(\frac{pCO_2}{SSS}\right) \simeq 1$, $\gamma_{FW} =$

$(\gamma_{SSS} + \gamma_{DIC} + \gamma_{ALK})$ (Sarmiento and Gruber 2006; Eggleston et al., 2010; Ouyang et al., 2020). The sensitivity terms are calculated based on PyCO2SYS (Humphreys et al., 2022).

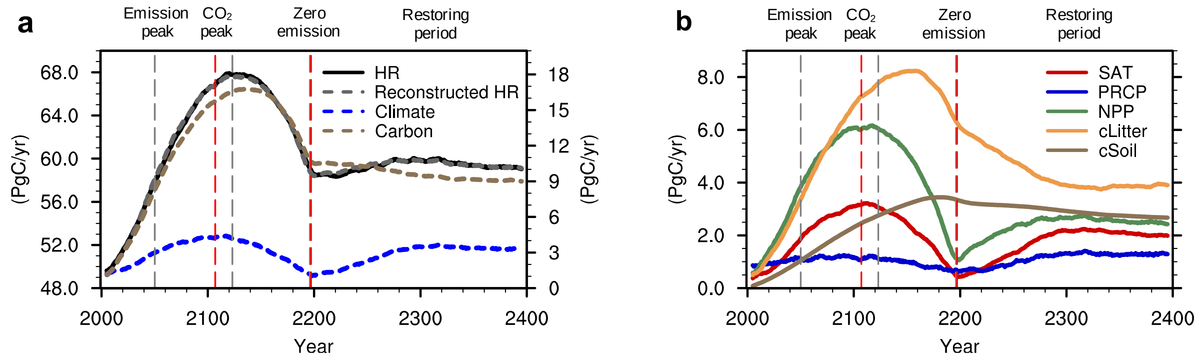

Figure S1. (a) Time-series of yearly HR and reconstructed HR from a multiple linear regression based on climate factors (normalized SAT and PRCP) and factors associated with terrestrial carbon cycle (normalized NPP, litter C, and soil C). Left axis is for the HR and reconstructed HR; Right axis is for the contributions of each factor (climate/carbon) to the change of HR. (b) Contributions of each of factors to the change of HR. All calculations were conducted after taking the 11-year running mean and then averaged (ensemble mean). The peak of anthropogenic emission and the beginning of negative and zero emissions (restoring period) are indicated by the gray dashed vertical line. The points of maximum and minimum CO_2 levels are indicated by the red dashed vertical line.

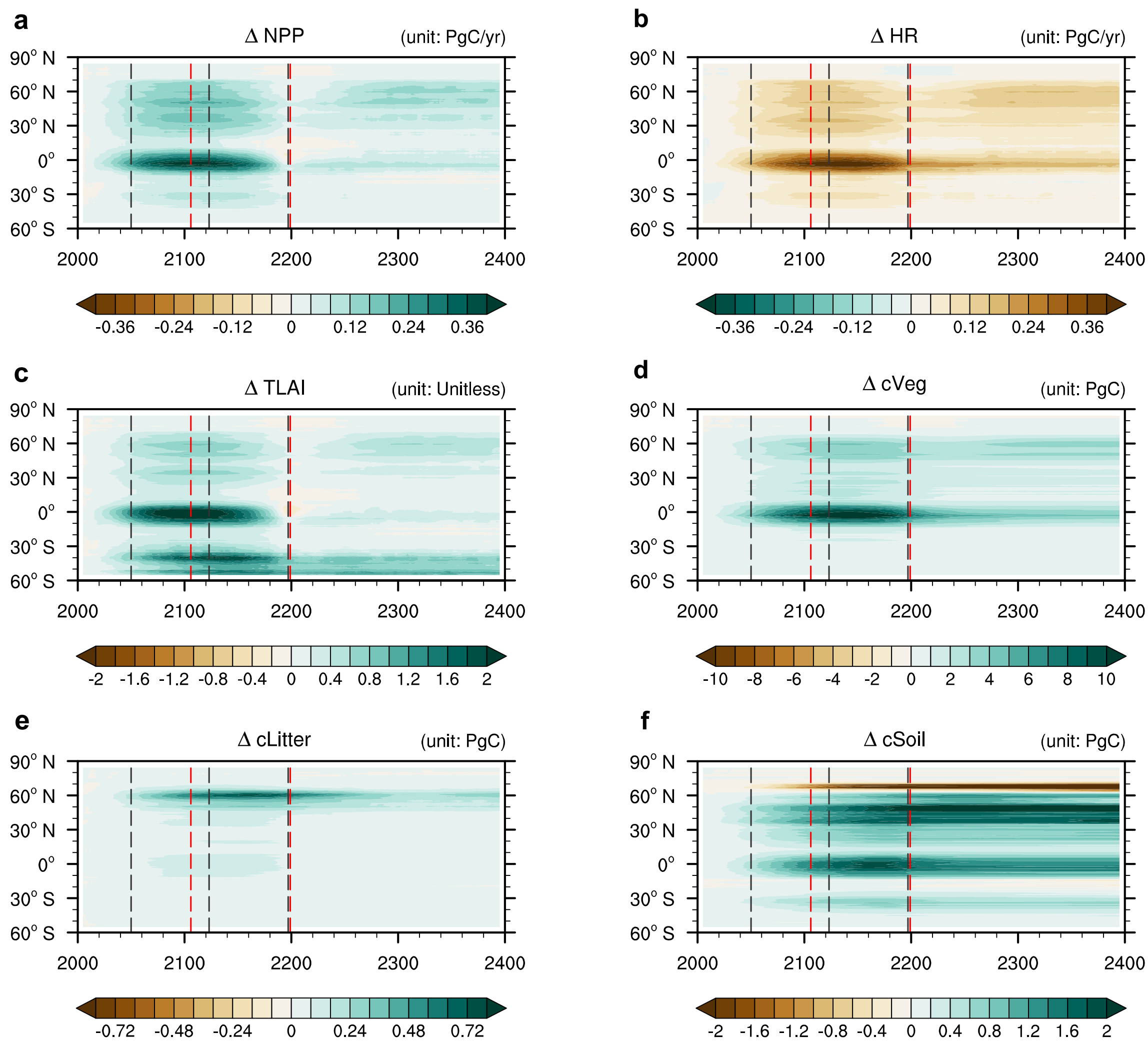

Figure S2. (a) Time-latitude diagrams of yearly NPP and (b) HR anomalies, (c) annual mean leaf area index anomaly, (d) annual mean vegetation, (e) litter, and (f) soil C content anomalies. All values are the ensemble mean and smoothed by the 11-year moving average.

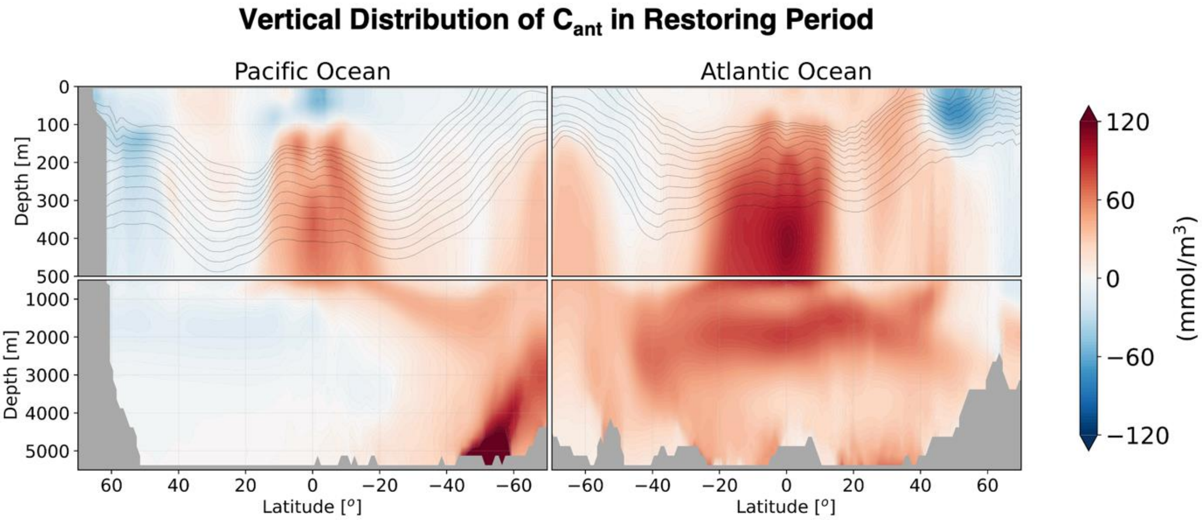

Figure S3. Vertical distribution of dissolved inorganic carbon (DIC) anomaly, C_{ant}, in the restoring periods. Selected black isolines represent the isopycnal lines in potential density (σ) from 25.8 to 28.0 kg/m^3 with 0.2kg/m^3 difference levels.

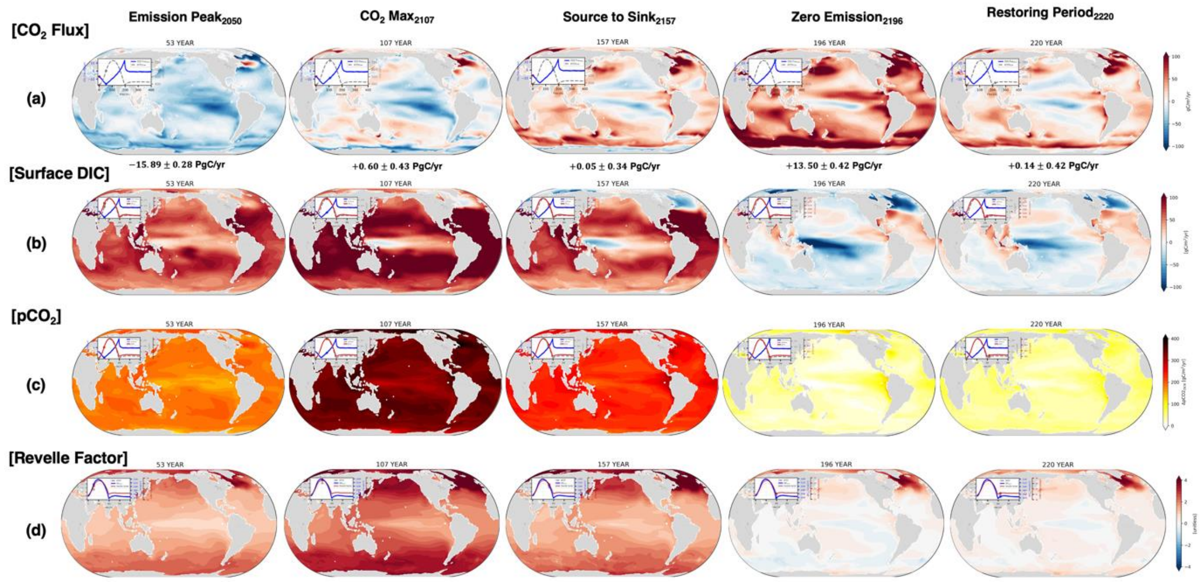

Figure S4. Anomaly contour map of ocean carbonate systems in specific periods such as emission peak (2050), CO_2 concentration maximum (2107), ocean role from source to sink (2157), zero emission (2196), and restoring periods (2220) from present climate (2000). (a) Air-Sea CO_2 Flux, (b) Surface dissolved inorganic carbon (DIC), (c) partial pressure of ocean CO_2, and (d) Revelle Factor.

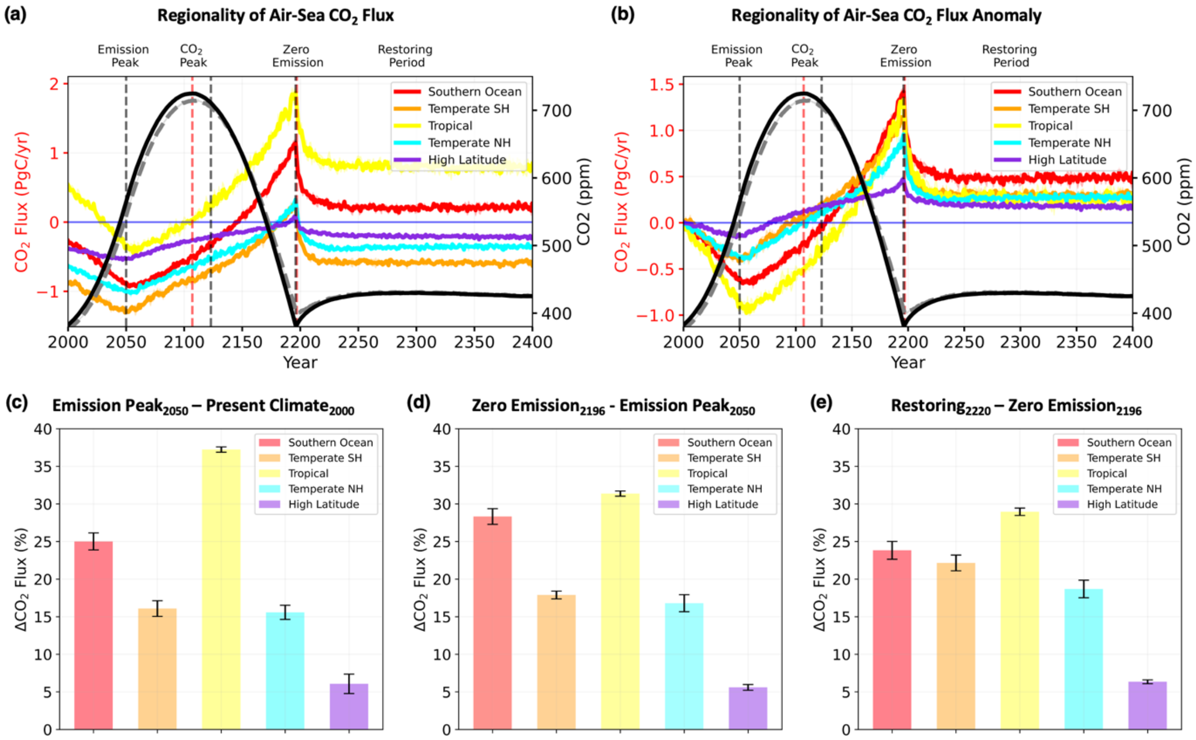

Figure S5. Regional time-series of (a) global averaged air-sea CO_2 flux and (b) anomaly. Ocean basins are divided by latitudes; Southern Ocean (<44 ° S), Temperate southern hemisphere (SH) (44 °S – 18 ° S), Tropical Ocean (18 °S – 18 ° N), Temperate northern hemisphere (NH) (18°N–49°N), and High latitude (>49°N) (Gruber et al., 2009). Each region is represented by different colors; Southern Ocean (red), Temperate SH (orange), Tropical Ocean (yellow), Temperate NH (cyan), and High latitude (purple). Contributions of ocean basins to global air-sea CO2 flux changes in (c) positive emission, (d) negative emission, and (e) zero emission.4