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June 18th, 2019

SUPPLEMENTARY MATERIAL

We provide additional analysis through (I) discussion and presentation of the global evolution of ocean carbon fluxes in the Large Ensemble and sensitivity experiments, (II) expanded discussion and presentation of emergence timescales for additional variables related to the ocean carbon pumps that are not covered fully in main text and (III) discussion and presentation of an alternative method for computing time of emergence using a pre-industrial simulation.

PART I : ANTHROPOGENIC CHANGES IN THE OCEAN CARBON PUMPS

The evolution of globally integrated air-sea CO₂ fluxes and biological pumps for the Large Ensemble (LE), two sensitivity experiments, and the pre-industrial control runs are shown in **Fig. S1**. Pre-industrially, the globally-integrated air-sea CO₂ flux is approximately zero PgC/yr, and the soft-tissue and calcium-carbonate pumps (quantified by export of particulate organic carbon (POC) and CaCO₃ at 100 meters depth) are 8.0 and 0.7 PgC/yr respectively. Note that the quasi-zero net air-sea CO₂ pre-industrial flux implies that the export of carbon in the soft-tissue and calcium-carbonate pumps across

the 100 m depth contour is resupplied by upward transport of dissolved inorganic carbon (DIC). The annual mean air-sea CO₂ flux increases by 6 PgC/yr under anthropogenic forcing by the end of the 21st century, whereas the biological pump decreases by ~1 PgC/yr. Until year ~2050, anthropogenic changes in the global air-sea CO₂ flux are due entirely to the impact of rising atmospheric CO₂ concentrations on air-sea gas exchange, not due to warming, with small contributions (<1 PgC/yr) from changes in the biological pumps. For this reason, we interpret changes in the air-sea flux as representing changes in the solubility or physical carbon pump (1). Additional confirmation that changes in biological export are not impacting the emergence characteristics of air-sea CO₂ fluxes can be found in **Fig. S2**, which demonstrates that the impact of changing export of surface pCO₂ is much smaller than the total change over the 21st century (0.5 uatm vs. 550 uatm, global averages). After year 2050, warming decreases uptake globally by ~1PgC/yr or 15%, consistent with CMIP5 estimates of climate-carbon feedback magnitude (2). Anthropogenic changes in the soft tissue pump are driven by warming (concurrence of All-forcings and Rad-only experiments), whereas anthropogenic changes in the CaCO₃ pump are driven by changes in ocean chemistry (concurrence of All-forcings and BGC-only experiments).

PART II : TIME OF EMERGENCE DISCUSSION FOR ADDITIONAL OCEAN PROPERTIES

Changes in the ocean carbon pumps do not occur in isolation, but rather they are coupled to many other changing ocean properties and processes. Some, but not all, of these related ocean fields are discussed fully in the main text. Notably, sea surface temperatures (SST), net primary productivity (NPP), controls on the soft-tissue pump, and oxygen inventories have limited discussion, and so we include a more detailed discussion here.

SST couples to the ocean carbon pumps through driving modulations of the CO₂ solubility (4) and biological productivity (5). Satellite-derived SST is a primary input for estimating air-sea CO₂ fluxes (e.g. 6) and biological export (e.g. 7). Therefore, the timescales of emergence for anthropogenic changes in SST are of interest to projection, detection and attribution of changes in the ocean carbon pumps. We find SST emerges in 20-30 years for localities in the tropics and extra-tropics (**Fig. S3e**), where the signal is strongest and positive (**Fig. S4e**). SST emergence timescales for GFDL-ESM2M LE were first shown in (8) and is consistent with multi-model emergence times and patterns shown in (9) with the exception of the high latitudes. Weak warming, and even cooling in the high latitudes results in delayed emergence, and even non-emergence for localities in the North Atlantic and

Southern Ocean. We note this is model-dependent, as cooling trends in the Southern Ocean in GFDL-ESM2M are stronger than the majority of CMIP5 models (10).

Changes in the soft-tissue pump also occur through a complex sequence of mechanisms. Changing ocean circulation (e.g. stratification, reduced ventilation and mixed layer depths, **Fig. S4u**) reduces nutrient availability in the upper ocean (**Fig. S4k & S4q**), shifting phytoplankton species/size distribution towards smaller phytoplankton (i.e. decreasing phytoplankton biomass, **Fig. S4n**), which are less efficient than large phytoplankton at exporting carbon from the surface ocean (**Fig. S4o,11**).

NPP has limited attributive value for anthropogenic trends in biological export, however, we include discussion of NPP as it is an important ecosystem quantity, and because other ESMs indicate stronger dependence of anthropogenic changes in POC export (soft-tissue pump) on anthropogenic changes of NPP (12). Emergent trends in NPP (**Fig. S3t**) follow the sign of trends in nitrate (NO_3) concentration (**Fig. S3q**). Exceptions occur over (a) the Equatorial Pacific cold tongue, where NO_3 concentrations are not limiting ($> 0.5 \text{ mol kg}^{-1}$) and warming enhances phytoplankton growth rates (b); the Pacific sector of the Southern Ocean, where NPP decreases due to decreasing iron (13, **Fig. 4Sk**); and (c) in the North West Pacific where NPP increases due to alleviation of light limitation associated with decreased mixed layer depths (MLD, **Fig. S4u**). For the North West Pacific, the biological impacts (a reduction in NPP) of a changing physical climate (shallowing MLD) emerge, in a statistical sense, before the physical driver (**Fig. S3t** vs. **Fig. S3u**). We note that increases in NO_3 (and NPP) in the band 45°S - 30°S is due to poleward expansion of the subtropical gyre resulting in an injection of higher- NO_3 at the gyre edge.

Finally, we discuss the emergence of O_2 inventories over 200-600m as it is of critical importance to marine life and impacted by the biological export and subsequent remineralization of organic carbon. The trend in O_2 inventories is determined by the relative contributions of changing solubility (reflected by $\text{O}_{2,\text{SAT}}$), and circulation and biology (primary productivity, biological export and respiration, reflected in Apparent Oxygen Utilization (AOU)). Decreasing $\text{O}_{2,\text{SAT}}$ emerges for most of the globe by mid-century (**Fig. S3i & S4i**), with the exceptions of the subtropical convergence zones/fronts, which have counter-trends, and remain non-emergent through the century. Consistent with CMIP5 projections presented in (14), AOU has positive trends in the high latitudes and negative trends in the mid-to-low latitudes, with these trends emerging mid-to-late century (**Fig. S3r & S4r**). Changes in AOU arise from a slowdown in the ocean overturning circulation which results in increased water age at intermediate depths in the high latitudes (positive AOU) and decreased water

age in intermediate depths in the mid-to-low latitudes (negative AOU), as older, deeper, oxygen-depleted waters have decreased interaction with waters at intermediate depths (15,16). The O_2 signal emerges most rapidly (~ 2030 -2060) in the high latitudes where $O_{2,SAT}$ and AOU have consistent trend directions (i.e. where AOU is increasing and $O_{2,SAT}$ decreasing, **Fig. S3s & S4s**). Regions with opposing $O_{2,SAT}$ and AOU trends emerge slowly (the West Equatorial Pacific and Indian Oceans), if at all (non-emergence over the subtropical gyres of the South Pacific and Atlantic Oceans). The North Atlantic exhibits limited emergence due to the dipole spatial pattern of trends and strong natural decadal variability.

Decreases in POC export (**Fig. S3o & S4o**) contributes to declining AOU, which counteracts the effects of declining O_2 solubility on O_2 inventories, thus extending emergence times for these regions. Contributions from POC export to anthropogenic changes in AOU are not (and cannot be) quantified here, but are generally considered to be important although second order to the impact of changes in ocean physics in modeling studies (15–17). However, it is observed to be of first order importance for some regions on seasonal to inter-annual scales (18).

PART III: ALTERNATIVE METHOD FOR ToE COMPUTATIONS

As a complement to the Large Ensemble calculations, we also consider output from a 1600-year long pre-industrial run with (the same) ESM2M model. This provides alternative means with which to estimate the noise associated with natural variability in determining ToE.

For the purposes of ToE estimates, the LE advantage is twofold: 1st) providing a purely forced **signal** at the local or pixel scale, without requiring a smoothing technique and 2nd) providing robust **noise** estimates for a perturbed (rather than pre-industrial) climate state. To illustrate both these advantages, we re-compute ToE using sub-sets (between 1 and 30 members) of the LE to estimate the forced signal and using 30, randomly selected 111-year segments from a 1600-year pre-industrial (PI) control run to estimate noise. In practice, if a 30-member ensemble were available, then the PI run would not be needed to estimate the noise, however to isolate the importance of noise estimation for ToE computation, we compare the ToE with the same signal (from the 30 member LE) but different noise (30 member LE noise vs. PI noise). **Fig. S7** compares the standard ToE_{LE} (both signal and noise from 30 LE members) to the re-computed ToE_{PI} (signal from 1-30 ensemble members, but noise from the PI control run). Error in air-sea CO_2 flux ToE estimates using PI noise and 3, 10, and

30 ensemble members to generate the signal are shown in **Fig. S7a, b and c**, respectively. Using PI noise and only 3 ensemble members (**Fig. S7a**) can result in errors of 50+ years relative to estimates generated from the full LE. This error reduces as the number of ensembles used to estimate the signal increases (**Fig. S7a, b & c**), however even when all 30 ensembles members are used to estimate the signal (**Fig. S7c**), error remains, particularly at frontal boundaries, where the underlying noise structures have spatially shifted between the pre-industrial and contemporary climate state (e.g. poleward expansion of the subtropical gyres, **Fig. S3m**).

Fig. S7d shows the fractional ocean area with error in the ToE_{PI} estimates greater than 20% of the ToE_{LE} estimates (i.e. $|ToE_{LE} - ToE_{PI}| / ToE_{LE} > 0.2$). Error estimates are generated through subsampling the combination space of “30-choose- n ”, e.g. there are exactly 30 combinations of 30 choose 1, however there are 435 combinations of 30 choose 2 (from which 30 are sampled for these calculations). The “maximum error” is the maximum error that occurs, between ToE_{LE} and ToE_{PI} , at a particular grid cell for a given sub-sample of LE members. For example, when $n = 3$, error is maximized for a particular pixel in the Southern Ocean when ensemble members 2, 14 and 27 are used to estimate the anthropogenic signal. The “expected error” is mean of 30 different combinations of 3 ensemble members. It is the error one would expect if only a 3-member ensemble were used, however the error could also be as large (or larger) than the “maximum error”—and so we calculate and show both errors. By design the “expected” and “maximum” error converge at $X = 30$, as there is only 1 combination of 30 choose 30.

For all fields, the extent of ocean area with ToE_{PI} error declines when an increasing number of ensembles are used to estimate the signal, however persistent errors remains (when $n = 30$) over 15-30% of the global ocean area due to differences between pre-industrial and contemporary noise, highlighting the necessity of LEs to minimize potential error in local ToE estimates.

There are three main messages from **Fig. S7d**: (1st) When using the pre-industrial control run to generate noise statistics, persistent error remains, even when the all 30 ensemble members are used to generate the anthropogenic signal, and (2nd) with the given error threshold of 20%, the mean or expected error between the full ensemble and a subset of the ensemble approaches zero (leveling off of the solid curves) occurs at about 5 ensembles (the persistent 15-30% of the ocean surface area with error > 20% is due to the use of PI noise, rather than a smaller ensemble size) and (3rd), Maximum error (dashed lines) between the full and partial ensemble does not decay exponentially (as does

expected error), but rather linearly, highlighting the necessity of LEs to minimize potential error in local ToE estimates.

We note that the methods employed here are not the only way to calculate emergence times. However, for robust estimates, the “signal” used must be free of natural variability. To achieve this, without a LE, one might take the trend over a longer period of time (e.g. 21st century). This assumes a stationary trend throughout the 21st century, which might be appropriate for some fields, however is not appropriate for others, like air-sea CO₂ fluxes, which are convex at the end of the century as the ocean’s CO₂ buffering capacity is depleted (e.g. **Fig. S1a**). Secondly, alternative non-LE methods need assumptions about the stationarity of noise and noise structures (e.g. frontal boundaries) – however **Fig. S7** demonstrates that assuming stationarity proves erroneous (e.g. the persistent error over 15-30% of ocean area in **Fig. S7d**), at least when the difference in ocean states considered is pre-industrial vs. contemporary. As shown in Frölicher (19) their Fig. 9, differences in noise between PI and RCP8.5 is model and field dependent, so the benefits of the LE method of ToE presented in this paper are also model dependent. We also note there are likely differences in estimates of noise between a historical simulation with natural forcings (i.e. solar, volcanoes) and a pre-industrial control run without. Future work should explore the model and method dependence of noise estimates on ToE estimates.

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Figure S1. Changes in ocean carbon pumps for the LE and sensitivity experiments. Panel **a.** shows the physical pump (air-sea CO_2 fluxes) and panel **b.** shows the biological pumps, where POC is particulate organic carbon. Values from the sensitivity experiments are shown in different colors. **All-forcings** indicates a fully coupled, transient run in which the forcing follows a historical-to-RCP8.5 pathway. In 1950, ensembles 2-29 branch from ensemble member 1, with the ensemble mean shown in darker purple. The **Rad-only** run has only the radiative impacts of anthropogenic forcings, and air-sea gas exchange is not affected by rising atmospheric CO_2 . The **BGC-only** run has only the chemical impacts of anthropogenic warming (i.e. increased CO_2 gas exchange, but no warming). **PreInd** indicates the pre-industrial control run. **Dashed line at 1990** indicates the beginning of the ocean-carbon observation era, and the reference state from which emergence is defined.

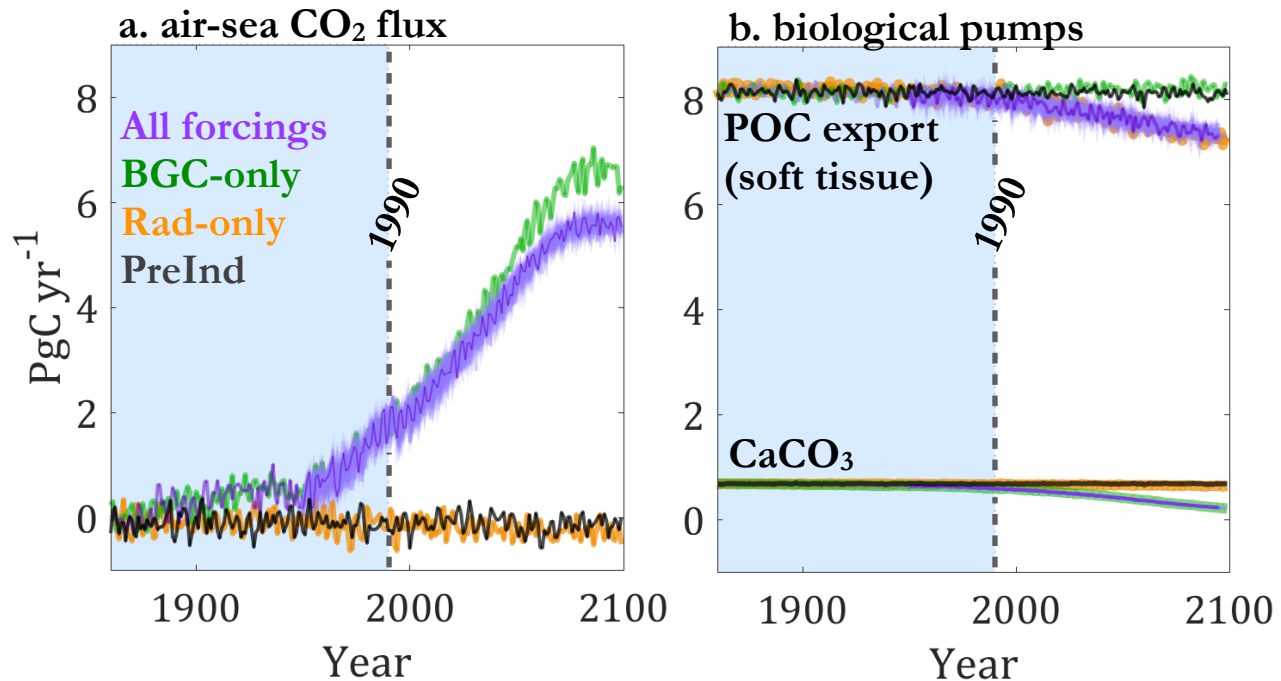


Figure S2. 111-year change in annual mean surface ocean pCO_2 due to biological export of carbon. The export of soft tissue and calcium carbonate impacts surface ocean pCO_2 through changes in the concentration of alkalinity (ALK) and dissolved inorganic carbon (DIC). The impacts are not explicitly diagnosed in ESM2M, however we do offline calculations with CO2SYS (3) to estimate the impact of changing export on surface pCO_2 . This is done by first computing pCO_2 using monthly output from the year 2100, then adjusting the concentration of DIC and ALK to account for changes in the pumps relative to 1990, and recalculating surface pCO_2 with the export-adjusted DIC and ALK concentrations. The difference between the prognostic pCO_2 at 2100 and the diagnostic pCO_2 at 2100 if the pumps were stationary since 1990, is shown below. The annual mean is taken after the computations of pCO_2 are done monthly. For reference, the total pCO_2 changes by ~ 550 uatm over this same time period, due to rising atmospheric pCO_2 and a warming surface ocean.

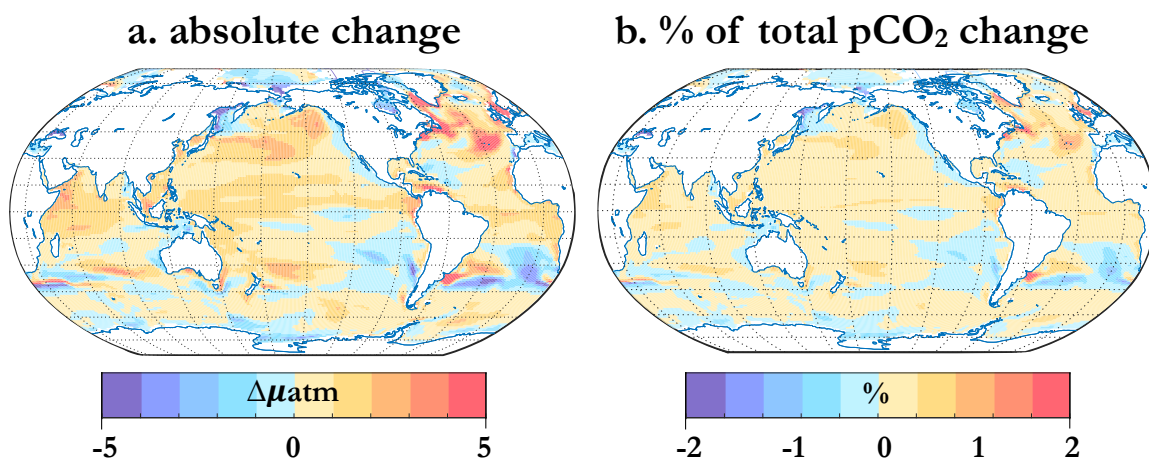


Figure S3. Time of Emergence Maps for selected variables. Omega applies to the saturation of both aragonite and calcite forms of calcium carbonate, for which emergence times are approximately equivalent. The CaCO_3 and soft-tissue pump are calculated as the export flux at 100 meters (m) depth of CaCO_3 and particulate organic carbon, respectively. The heat content is calculated as an integral over 0-700 meters, whereas the O_2 inventories consider the integral 200-600m, and chlorophyll inventories are considered over 0-500m. NPP represents an integral over 0-100m. All other variables represent sea surface properties. Asterisks on the 3 carbon fluxes (pumps) which are repeated from Fig. 4 of the main text. $\Delta p\text{CO}_2$ repeated from Fig. 5. Chlorophyll maps are repeated from Fig. 6.

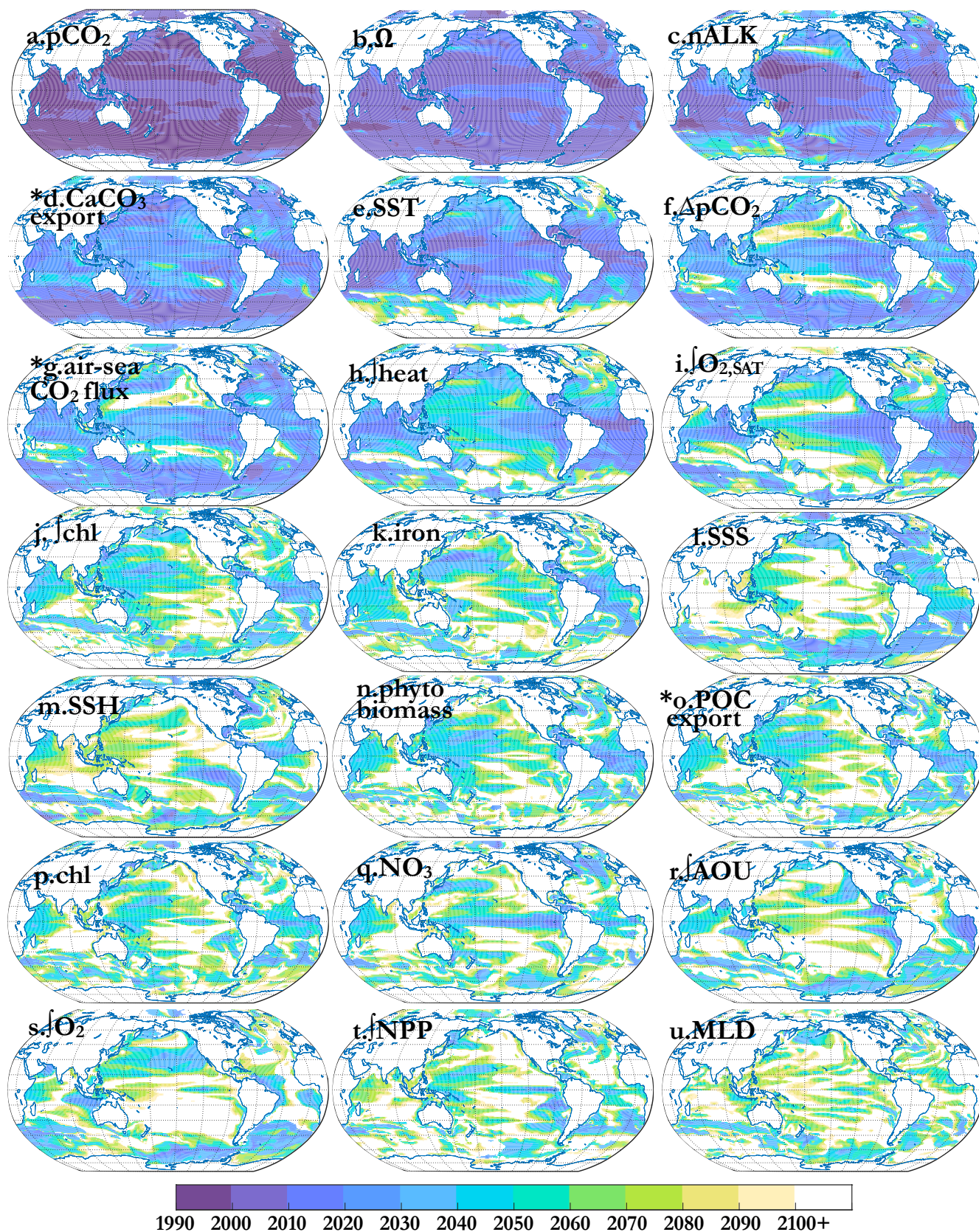


Figure S4. Signal at Time of Emergence or year 2100, whichever comes first. This figure is meant to illustrate the direction and relative magnitude of trends. Blues are negative trends, Reds are positive trend, with increasing color intensity scaling with magnitude of trend. White hatch over non-emergent regions. $[\text{NO}_3] = 0.5 \mu\text{mol kg}^{-1}$ contours (1990 ensemble annual mean values) imposed on some fields for explanatory purposes. $\Delta p\text{CO}_2 = p\text{CO}_{2,\text{ocn}} - p\text{CO}_{2,\text{atm}}$. Caption continues next page.

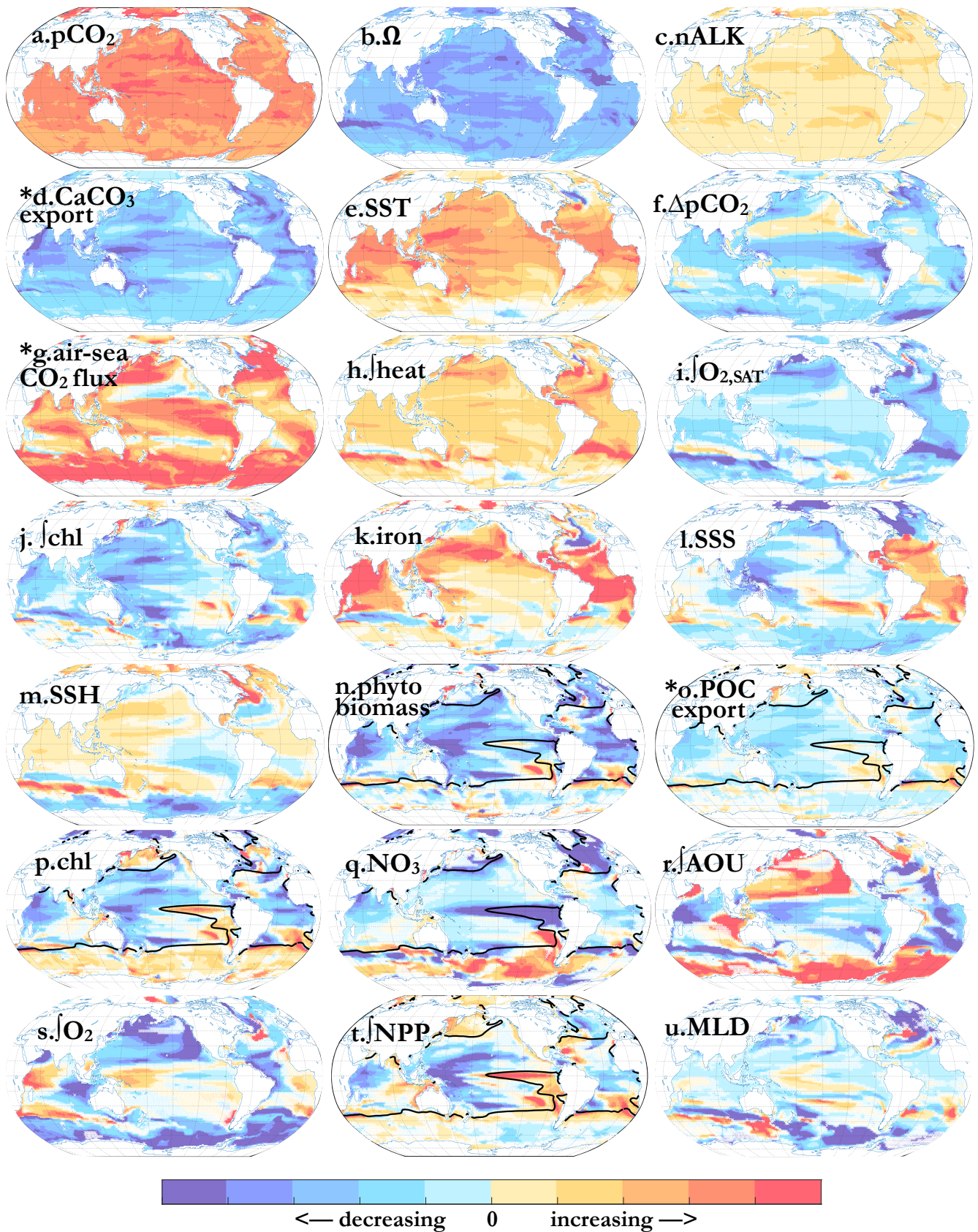


Figure S4. (caption continued) The value of darkest red and corresponding units are as follows: $p\text{CO}_2$ [3 uatm yr^{-1}], Ω [0.01 yr^{-1}], salinity normalized alkalinity (nALK) [$0.3 \text{ meq m}^{-3} \text{ yr}^{-1}$], CaCO_3 export at 100m [$0.02 \text{ gC m}^{-2} \text{ yr}^{-1} \text{ yr}^{-1}$], sea surface temperature (SST) [$0.05 \text{ }^\circ\text{C yr}^{-1}$], $\Delta p\text{CO}_2$ [0.5 uatm yr^{-1}], fgCO_2 [$0.2 \text{ gC m}^{-2} \text{ yr}^{-1} \text{ yr}^{-1}$], heat inventory [$8.37 \times 10^7 \text{ J m}^{-2} \text{ yr}^{-1}$], $\text{O}_{2,\text{sat}}$ inventory 200-600m [$0.22 \text{ mmol m}^{-3} \text{ yr}^{-1}$], chlorophyll inventory [$0.02 \text{ mmol m}^{-2} \text{ yr}^{-1}$], iron concentration [$1.5 \text{ nmol m}^{-3} \text{ yr}^{-1}$], sea surface salinity (SSS) [$0.008 \text{ psu yr}^{-1}$], sea surface height (SSH) [0.3 cm yr^{-1}], large phytoplankton [$1.4 \text{ } \mu\text{mol m}^{-3} \text{ yr}^{-1}$], POC export at 100 meters [$0.2 \text{ gC m}^{-2} \text{ yr}^{-1} \text{ yr}^{-1}$], surface chlorophyll concentration [$0.0023 \text{ mmol m}^{-3} \text{ yr}^{-1}$], NO_3 [$2.0 \text{ } \mu\text{mol m}^{-3} \text{ yr}^{-1}$], apparent oxygen utilization (AOU) inventory 200-600m [$0.22 \text{ mmol m}^{-3} \text{ yr}^{-1}$], O_2 inventory 200-600m [$0.22 \text{ mmol m}^{-3} \text{ yr}^{-1}$], net primary production (NPP) 0-100m [$0.075 \text{ gC m}^{-2} \text{ yr}^{-1}$], mixed layer depth (MLD) [0.3 cm yr^{-1}]

Figure S5. Decadal variability and mean state for CaCO_3 export. (a) Decadal variability in CaCO_3 export, defined by the standard deviation of the 30 ensemble member annual-mean trends over the 20-year period 1990 to 2009, and (b) ensemble mean annual-mean CaCO_3 export at year 1990. Units are in $\text{gC m}^{-2} 20\text{yr}^{-1}$ (per 20 years) for (a) and $\text{gC m}^{-2} \text{ yr}^{-1}$ for (b) and corresponding ranges for the two panels are given below the color bar. Note that the color bar range of the (a) decadal variability is one order of magnitude less than (b) the mean state.

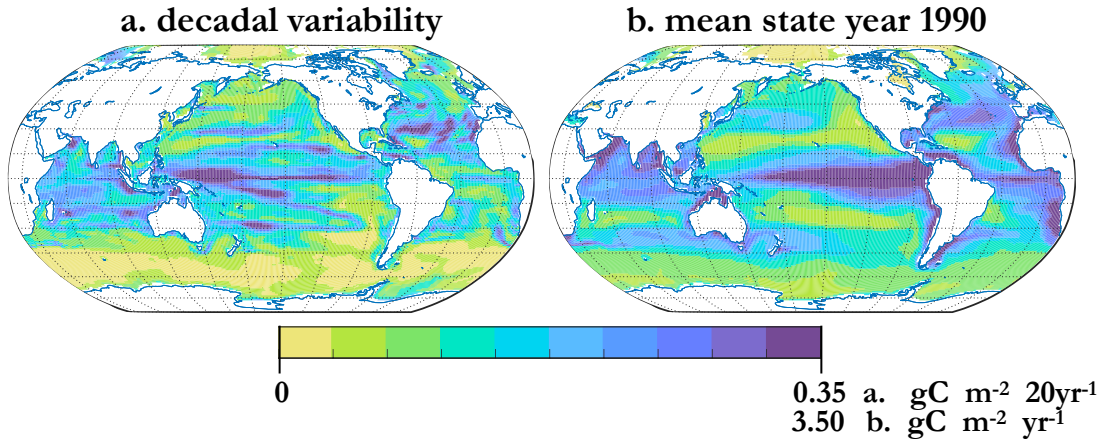


Figure S6. Depth differences in trends for iron limitation. For (a) small and (b) large phytoplankton, the difference between surface trends and 50 m trends (surface minus 50 meters) in iron limitation during austral summer (January-March). The trend is taken from the ensemble mean over the period 1990 to 2050. Units given in $\text{mol kg}^{-1} \text{ yr}^{-1}$. For both large and small phytoplankton, positive values in the Southern Ocean indicate iron limitation trends increases with depth.

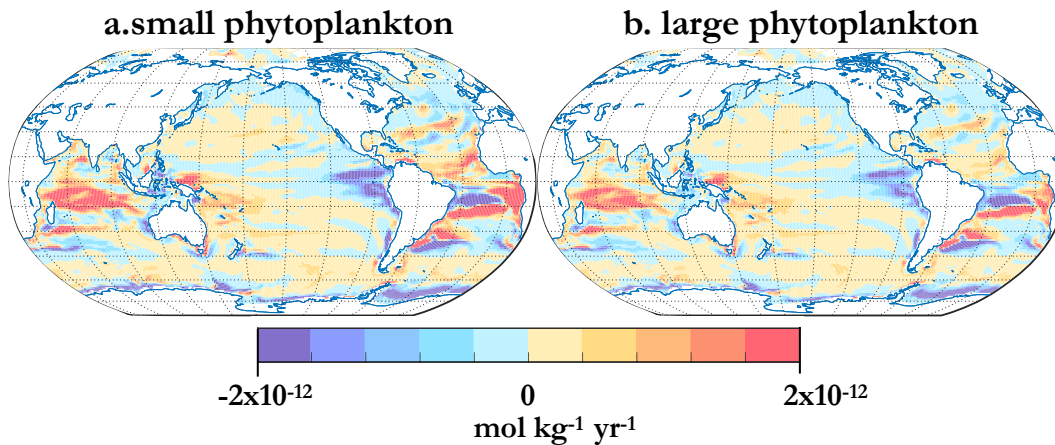


Figure S7. Error for ToE calculation with Pre-Industrial noise. (a), (b), and (c) show the maximum difference between emergence times calculated for air-sea CO₂ fluxes between using the full ensemble for both signal and noise estimates (ToE_{LE}) and using pre-industrial noise (ToE_{PI}) and 3, 10 and all 30 ensemble members to estimate the anthropogenic signal. (d) shows the fractional ocean area with a difference between ToE_{LE} and ToE_{PI} greater than 20%. Increasing the number of ensemble members reduces error, however persistent error remains (~15-30% of the globe), due to the inaccuracy of pre-industrial noise. Dashed lines are the maximum error in ToE estimates, solid lines are mean (expected) error, for the given number of ensemble used to estimate the anthropogenic signal. See supplemental text for full description of methodology.

