

Text S1

1. Biogeochemical Model

The ocean biogeochemical model includes in simplified form the main physico-chemical and biological processes governing the cycling of the tracers dissolved inorganic carbon, alkalinity, phosphate, iron and oxygen and the export and remineralization fluxes of dissolved and particulate organic carbon and calcium carbonate (CaCO_3). In the ocean model, the biological source-sink term has been changed from a nutrient restoring formulation as used in OCMIP-2 [Najjar *et al.*, 2007] to a prognostic formulation. Biological productivity is computed prognostically from temperature, surface solar irradiance, mixed layer depth, and macro- and micronutrients. Following the OCMIP-2 protocols, total biological productivity is partitioned 1/3 into sinking particulate organic matter (POC), and 2/3 into the formation of dissolved or suspended organic matter, much of which is remineralized within the model euphotic zone. Total productivity thus contains both new (upwelling or upward mixing of nutrients from the thermocline) and regenerated (recycling of organic matter within the surface ocean) production. While not strictly equivalent to primary production as measured by ^{14}C methods, model primary production is a reasonable proxy for the time and space variability of observed primary production, but has a lower magnitude. Since the model doesn't explicitly include sediments, any organic matter reaching the bottom of the ocean is assumed to be remineralized in the lowest

layer of the model with a first-order rate. The marine iron cycle is parametrized following *Doney et al.* [2006] including atmospheric dust deposition/iron dissolution, biological uptake, vertical particle transport and scavenging.

The modeled atmospheric oxygen mass balance equation is given by

$$\frac{\partial[\text{O}_2]_{\text{atm}}}{\partial t} + \xi = -\Phi_{\text{foss}} + \Phi_{\text{oa}} + \Phi_{\text{ta}}. \quad (\text{S1})$$

Atmospheric O_2 , $[\text{O}_2]_{\text{atm}}$, is advected and mixed as a dry-air mixing ratio using a semi-Lagrangian advection scheme, where ξ is the atmospheric transport operator for oxygen. Φ represent the source-sink terms due to O_2 consumption by fossil fuel burning (Φ_{foss}), the net exchange between the ocean and the atmosphere (Φ_{oa}), and the net exchange between the terrestrial biosphere and the atmosphere (Φ_{ta}).

Oxygen concentrations in the ocean interior, $[\text{O}_2]$, are governed by

$$\frac{\partial[\text{O}_2]}{\partial t} + \text{L} = \Phi_{\text{b}} + \Phi_{\text{ao}}, \quad (\text{S2})$$

where L is the 3-D transport operator for oxygen, which represents effects due to advection, diffusion, and convection. Φ_{b} is the biological source-sink term for oxygen and Φ_{ao} is the source-sink term as a result of air-sea gas exchange. The production and remineralization of organic phosphorus produces and consumes O_2 in a constant stoichiometric O_2 :P ratio of -170 mol O_2 per mol P [*Anderson and Sarmiento, 1994*]. Denitrification in oxygen minimum zones is dealt with implicitly in the model by halting oxygen consumption below an oxygen threshold ($[\text{O}_2]_{\text{crit}} = 4 \mu\text{mol kg}^{-1}$). The ocean will then be a net source of oxygen to the atmosphere because oxygen production above the compensation depth will be greater than oxygen consumption below the compensation depth.

Air-sea fluxes of O_2 are computed according to OCMIP-2, except that surface wind speed,

atmospheric pressure, and ice fraction are from the atmosphere and sea-ice components.

The source-sink term for the surface layer is:

$$\Phi_{ao} = \frac{k_w}{dz_1} * ([O_2]^{surf} - \frac{P^0}{P} * [O_2]_{sat}^0), \quad (S3)$$

where $[O_2]^{surf}$ is the oxygen concentration in the top layer of the ocean ($dz_1 = 12$ m) and $[O_2]_{sat}^0$ is the oxygen saturation concentration at surface pressure ($P^0 = 1$ atm) in mol m^{-3} for a given temperature and salinity [*Garcia and Gordon, 1992*]. P is the total atmospheric pressure and the windspeed-dependent gas exchange coefficient k_w across the air-sea interface is adopted from *Wanninkhof [1992]*:

$$k_w = (1 - f_{ice}) * 0.31 * U_{10}^2 * (S_c/660)^{-1/2}, \quad (S4)$$

where k_w has units of $cm\ hr^{-1}$ and U_{10} is the wind speed in ms^{-1} interpolated to a height of 10 m above the sea level. f_{ice} is the fractional sea-ice coverage and the Schmidt number S_c for oxygen is computed using the formulation proposed by *Keeling et al. [1998]*.

2. Climatological Mean State of AOU: Data versus Model Results

The modeled large-scale pattern of AOU compares reasonably well with observations from the World Ocean Atlas 2005 [*Garcia et al., 2006*] (Figure SS1). However, the AOU maximum in the equatorial Atlantic is slightly overestimated because biological production (as defined in section 2 in the supplementary material) is too high compared with observations as a consequence of too intense upwelling [*Schneider et al., 2008*]. An obvious difference between model and observations is the much shallower penetration of North Atlantic Deep Water (NADW) in the model indicated by high concentrations of AOU in the deep North Atlantic. The model underestimates AOU concentrations (overestimates O_2) by up to 100 $\mu mol\ kg^{-1}$ in the North Pacific. This is caused by too little nutrient inputs

into the upper, sunlit surface ocean, overly strong iron limitation and consequently too low export of organic matter in the North Pacific, and problems with upwelling parametrization [Schneider *et al.*, 2008; Doney *et al.*, 2006]. Excessive iron limitation occurs mainly in the subtropical Pacific and is linked to overly strong iron scavenging.

3. Oxygen Drift and Detrending Procedure

For this study, all runs were performed on an IBM SP4 and on an IBM P5 at the Swiss National Supercomputing Centre. Results for a benchmark simulation over the historical period agree with the results presented by Fung *et al.* [2005]. The control simulation exhibits a drift in dissolved oxygen concentrations and other tracers related to the long time-scale of deep ocean overturning. The total oxygen inventory is increasing by about 0.9 Pmol over the simulated period (Figure 2a, main text). The increase in oxygen occurs at intermediate depth (1500 m) in the Pacific and oxygen is decreasing at deeper depth, mainly in the Atlantic Ocean. The total sea-air O₂ flux is increasing by about 0.01 Pmol over the simulated period with a permanent outgassing of 0.03 Pmol yr⁻¹. Thus, we apply a grid-point based correction to remove model drift. This is done by splining monthly-mean values of the control simulation at each grid point and for each calendar month individually with a spline that eliminates high-frequency variations (nominal cut-off period of 200 years for dissolved oxygen, air-sea oxygen flux, phosphate (PO₄) and dissolved inorganic carbon (DIC), and of 1000 years for temperature and salinity). These splines, representing low-frequency trends, are then subtracted from the results of the transient simulations. The sea-air O₂ flux offset of 0.03 Pmol yr⁻¹ is subtracted for analysis in section 3.1. One reason for the slight trend in dissolved O₂ is drift in the coupled

physical simulation resulting in surface ocean freshening and a corresponding increase in deep-water salinity due to problems in the representation of sea-ice. Additionally, the ocean oxygen is not in balance (i.e not conserved) in the ocean biogeochemistry model (as defined in OCMIP-2 BGC models) because of denitrification.

4. Definitions of Different Internal Oscillation Modes

We calculated the different circulation indices from a 480 year control run. The NAO index is calculated as the amplitude of the pattern defined by the leading empirical orthogonal function (EOF) of winter (DJFM) monthly mean surface layer pressure anomalies between 20°N and 70°N and between 90°W and 40°E [Hurrell, 1995]. The simulated explained variances of the NAO is 51 %. The PDO index is computed from the first orthogonal function of SST over the North Pacific north of 20°N [Deser *et al.*, 2004]. The explained variance of the PDO index is 34 %. The performance of the model in representing the different climate indices is described in detail by Meehl and Arblaster [1998].

5. Data Sources and Interpolation Method

Data used in section 3.3 of the main text for illustrative model-data comparisons have been sampled along 152°W in the eastern subtropical/subarctic North Pacific in the years 1980, 1984, 1991 and 1997 [Deutsch *et al.*, 2006; Emerson *et al.*, 2004, 2001]. In the Atlantic, data along 20°W starting south of Iceland are used. The Atlantic sections were sampled in the years 1988 [Tsuchiya *et al.*, 1992], 1993 [Doney *et al.*, 1998], 1998 [Smythe-Wright, 1999] and 2003 (<http://ushydro.ucsd.edu>), and are discussed in Johnson and Gruber [2007].

Observed AOU and O₂ changes in the North Atlantic and Pacific have been interpolated onto a grid with a horizontal resolution of 1° × 1° and potential density as vertical coordinate. Potential density is referenced to 1000 dbar (σ_1) and 0 dbar (σ), respectively. A Gaussian interpolation function with a 2° mapping scale for Gaussian weights is used. Modeled tracer concentrations are converted from volume normalized units to conventional mass normalized units using a constant conversion factor of 1024.5 kg m⁻³.

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Figure S1. Observed (top) and modeled (bottom) zonal mean of AOU for the Atlantic Ocean (left) and the Indo-Pacific Ocean (right). Observed distributions are from the World Ocean Atlas 2005 [*Garcia et al.*, 2006]. The model ensemble mean is averaged from 1990 to 1999. Contours are every $1 \mu\text{mol kg}^{-1}$ and represent one standard deviation among the six ensemble members for the zonal and decadal mean.