Revision of the global carbon budget due to changing air-sea oxygen fluxes

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Carbon budgets inferred from measurements of the atmospheric oxygen to nitrogen ratio (O$_2$/N$_2$) are revised considering sea-to-air fluxes of O$_2$ and N$_2$ in response to global warming and volcanic eruptions. Observational estimates of changes in ocean heat content are combined with a model-derived relationship between changes in atmospheric O$_2$/N$_2$ due to oceanic outgassing and heat fluxes to estimate ocean O$_2$ outgassing. The inferred terrestrial carbon sink for the 1990s is reduced by a factor of two compared with the most recent estimate by the Intergovernmental Panel on Climate Change (IPCC). This also improves the agreement between calculated ocean carbon uptake rates and estimates from global carbon cycle models, which indicate a higher ocean carbon uptake during the 1990s than the 1980s. The simulated decrease in oceanic O$_2$ concentrations is in qualitative agreement with observed trends in oceanic O$_2$ concentrations.

INDEX TERMS: 1635 Global Change: Oceans (4203); 1615 Global Change: Biogeochemical processes (4805); 4806 Oceanography: Biological and Chemical: Carbon cycling; 4805 Oceanography: Biological and Chemical: Biogeochemical cycles (1615); 4842 Oceanography: Biological and Chemical: Modeling; KEYWORDS: global carbon budget, changes in ocean heat content, oceanic oxygen outgassing, ocean and land sinks of CO$_2$, carbon cycle modeling


1. Introduction

A quantitative understanding of the carbon cycle is important to develop global warming mitigation strategies. The conventional method, applying an ocean model to estimate the partitioning of anthropogenic carbon between the global terrestrial and ocean carbon sinks [Siegenthaler and Oeschger, 1987] has been complemented by various data-based methods to assess the strength of the terrestrial and oceanic carbon sinks [Keeling et al., 1989; Tans et al., 1990; Keeling and Shertz, 1992; Quay et al., 1992; Heimann and Maier-Reimer, 1996; Peng et al., 1998; Prentice et al., 2001]. Regional estimates of the carbon sinks [e.g., Enting and Mansbridge, 1991; Rayner et al., 1999; Gruber and Keeling, 2001] are constrained by the global estimates. Here we investigate how global warming and volcanic eruptions affect sea-to-air oxygen (O$_2$) and nitrogen (N$_2$) fluxes and, in turn, the carbon budgets for the last 2 decades deduced from the observed trends in atmospheric carbon dioxide (CO$_2$) and O$_2$; the latter estimated from measurements of the ratio of oxygen to nitrogen (O$_2$/N$_2$) in air [Keeling et al., 1996; Battle et al., 2000; Manning, 2001; Prentice et al., 2001].

The terrestrial and oceanic carbon sinks are estimated by solving the atmospheric budgets of O$_2$ and CO$_2$ (Figure 1). The observed increase in atmospheric CO$_2$ [Keeling and Whorf, 2000] equals the known amount of carbon released by fossil fuel burning [Marland et al., 1995] minus the carbon that has been taken up by the ocean and the land biosphere. For each mol CO$_2$ released by fossil fuel burning, about 1.39 mol O$_2$ are consumed [Manning, 2001]. In turn, about 1.1 mol O$_2$ are released for each mol CO$_2$ taken up by the biosphere [Severinghaus, 1995], whereas CO$_2$ uptake by the ocean does not alter atmospheric O$_2$. The basic assumption has been that sea-to-air O$_2$ fluxes are negligible on a multiannual timescale, and thus the effect of oceanic O$_2$ outgassing on the atmospheric O$_2$ budget has typically been omitted [Keeling et al., 1996; Bender and Battle, 1999; Battle et al., 2000] (Figure 1).

The atmospheric and marine inventories of gaseous or dissolved O$_2$ are coupled through air-sea gas exchange. O$_2$ may be released to the atmosphere by a number of processes. First, the solubility of O$_2$ is reduced in a warming ocean. Second, concentrations of dissolved O$_2$ are altered by changes in the balance between O$_2$ transported to depth and O$_2$ consumed during the remineralization of organic material. A slowed circulation or an increased export production of organic material leads to lower O$_2$ concentrations at depth and increased O$_2$ outgassing. And third, increasing storage of organic carbon in dissolved organic matter or in sediments would tend to increase oceanic O$_2$ inventories. Simulations with coupled atmosphere-ocean biogeochemical models yield a reduced ocean circulation and a decrease in the oceanic O$_2$ inventory under global warming [Sar-
Figure 1. Budgets of atmospheric O$_2$, expressed as atmospheric O$_2$/N$_2$ ratio, and CO$_2$ (adapted from Prentice et al. [2001]). The diagram schematically illustrates the principles of the partitioning of fossil fuel carbon using measurements of atmospheric O$_2$ and CO$_2$, by considering simultaneously changes in atmospheric CO$_2$ (horizontal axis) and O$_2$ (vertical axis) [Keeling et al., 1996]. The arrow labeled “fossil fuel burning” denotes the effect of the combustion of fossil fuels assuming that 1.39 mol O$_2$ are consumed for 1 mol CO$_2$ released [Manning, 2001]. The arrow labeled “outgassing” indicates O$_2$ changes from oceanic outgassing primarily due to changes in the marine biogeochemical cycle; its size is estimated from data of ocean heat uptake [Levitus et al., 2000] and a model-derived relationship between ocean heat fluxes and atmospheric O$_2$/N$_2$ changes due to outgassing. Carbon uptake by land and ocean is constrained by the known O$_2$/CO$_2$ stoichiometric ratios of these processes [Severinghaus, 1995; Keeling et al., 1996; Battle et al., 2000; Manning, 2001], defining the slopes of the respective arrows, and the observed changes in atmospheric CO$_2$ [Keeling and Whorf, 2000] and O$_2$. The trend in atmospheric O$_2$ is established from measurements of the ratio of O$_2$ to N$_2$ in air (solid circles: Keeling et al. [1996] and Manning [2001]; solid triangles: Battle et al. [2000]). Changes in atmospheric O$_2$/N$_2$ are usually given in units of per meg. A per meg corresponds to a relative difference in the O$_2$/N$_2$ ratio of two samples of 10$^{-6}$ [Keeling et al., 1996].

2. Model and Methods

[7] We have used a low-order physical-biogeochemical climate model that consists of a zonally averaged ocean model, coupled to an atmospheric energy balance model [Stocker et al., 1992] including an active hydrological cycle [Schmittner and Stocker, 1999], and a representation of the carbon cycle in the ocean [Marchal et al., 1998] and the terrestrial biosphere [Siegenthaler and Oeschger, 1987]. Model configuration and spin-up to a modern steady state are as given by Plattner et al. [2001], except that vertical resolution in the upper 1,000 m has been increased to 20 levels. This has little impact on simulated distributions of dissolved O$_2$ and other tracers and does not alter the conclusions. Nitrogen (N$_2$) was added as an inorganic tracer to simulate solubility driven changes in N$_2$ sea-to-air fluxes. The O$_2$ air-sea gas exchange is now explicitly considered in the model. Air-sea gas exchange of O$_2$ and N$_2$ is dependent on windspeed and temperature [Wanninkhof, 1992]. Details on the incorporation of O$_3$ and N$_3$ in the physical-biogeochemical climate model and on the different model simulations performed are provided in the Appendix.

[8] Radiative forcing from reconstructed (until year 2000) and later projected changes in the abundances of greenhouse gases and aerosols in the atmosphere was calculated based on simplified expressions [see Joos et al., 2001, and references therein]. We consider radiative forcing from changes in atmospheric CO$_2$, CH$_4$, N$_2$O, stratospheric and tropospheric O$_3$, stratospheric H$_2$O due to CH$_4$ changes, SF$_6$, 28 halocarbons (including those controlled by the Montreal Protocol), from direct and indirect effects of sulfate aerosols, and from direct forcing of black and organic carbon. Radiative forcing by explosive volcanic eruptions and changes in solar irradiance was prescribed for the last millennium [Crowley, 2000]. Transient runs start at year 1000 A. D. Output is only analyzed after 1400 A. D. to exclude the transition into a slightly different ocean circulation state after addition of radiative forcing by
modeled ocean heat content is temporarily reversed in the house gas forcing (Figure 2). The increasing trend in warming as compared to simulations without global warming is reduced by about 8% in simulations with global volcanoes; the North Atlantic Deep Water formation rate dropped from 22 to 18 Sv (1 Sv = 10^6 m^3 s^{-1}). The model’s climate sensitivity, that is the increase in global mean surface air temperature for a doubling of atmospheric CO_2, ΔT_{2x}, was set to 3.7°C in the standard simulation and 0°C in the baseline simulation (constant climate and ocean circulation).

3. Results

3.1. Modeled Air-Sea Fluxes of O2 and Heat

The ocean is taking up heat in response to greenhouse gas forcing (Figure 2). The increasing trend in modeled ocean heat content is temporarily reversed in the years after explosive volcanic eruptions (Figure 3a). The modeled change in oceanic heat content between 1955 and 1995 is in close agreement with observations [Levitus et al., 2000]. However, similar to studies applying coupled atmosphere-ocean general circulation models [e.g., Barnett et al., 2001; Levitus et al., 2001; Bopp et al., 2002], decadal scale variability in the ocean heat content is substantially underestimated. The observed decadal scale variations stem mainly from the Pacific Ocean [Levitus et al., 2000] and may be due to internal climate oscillations such as El Niño/Southern Oscillation or the Pacific Decadal Oscillation that are not resolved in our model.

Table 1. Carbon Uptake Rates by the Ocean and by the Terrestrial Biosphere

<table>
<thead>
<tr>
<th>Carbon Fluxes, GtC yr^{-1}</th>
<th>From Atmospheric O2/N2</th>
<th>From Ocean Models</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>IPCC^b</td>
<td>Revised^b</td>
</tr>
<tr>
<td>Period 1980–1989</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ocean-atmosphere flux</td>
<td>-1.9 ± 0.6</td>
<td>-1.7 ± 0.6</td>
</tr>
<tr>
<td>Land-atmosphere flux</td>
<td>-0.2 ± 0.7</td>
<td>-0.4 ± 0.7</td>
</tr>
<tr>
<td>Period 1990–1999</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ocean-atmosphere flux</td>
<td>-1.7 ± 0.5</td>
<td>-2.4 ± 0.7</td>
</tr>
<tr>
<td>Land-atmosphere flux</td>
<td>-1.4 ± 0.7</td>
<td>-0.7 ± 0.8</td>
</tr>
<tr>
<td>Difference Between the 2 Decades (1990s Minus 1980s)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ocean-atmosphere flux</td>
<td>+0.2</td>
<td>-0.7</td>
</tr>
<tr>
<td>Land-atmosphere flux</td>
<td>-1.2</td>
<td>-0.3</td>
</tr>
</tbody>
</table>

^a^ Positive values are fluxes to the atmosphere; negative values represent uptake from the atmosphere. Error ranges denote uncertainty (one standard deviation), not interannual variability, which is substantially larger.

^b^ Based on intradecadal trends in atmospheric CO_2 and O_2 as given by the IPCC [Prentice et al., 2001] and detailed by Manning [2001]. A small correction for solubility driven O_2 outgassing is included.

^c^ Corrected for the effect of oceanic O_2 outgassing caused by changes in the marine biogeochemical cycles. The correction is estimated from data of heat ocean uptake [Levitus et al., 2000] and the model-derived linear relationship between ocean heat uptake rates and changes in atmospheric O_2/N_2 due to outgassing (1.56 per meg/10^{22} J).

^d^ Ocean carbon uptake rates calculated by prescribing annual-mean atmospheric CO_2 from observations at Mauna Loa and South Pole [Keeling and Whorf, 2000] in the standard model setup.

^e^ Ocean carbon uptake rates averaged from simulations of eight models participating in the Ocean Carbon-Cycle Model Intercomparison Project (OCMIP) (adapted from Orr [2000]); model range is given in parentheses. Global warming feedbacks are not considered in the OCMIP simulations. Prescribed atmospheric CO_2 is higher than observed during the 1990s in the OCMIP simulations; the ocean carbon uptake rates shown have been obtained by scaling the original OCMIP results downward by 13%, based on the baseline simulation and the OCMIP simulation with our model.

Figure 2. Ocean heat content anomaly versus time. Observed (dash-dotted line; Levitus et al. [2000]) and modeled (solid line; ΔT_{2x} = 3.7°C) oceanic heat content anomalies during the last 5 decades. Data and model results are 5-year running means relative to the average for the 40-year period. Uncertainties (1 standard deviation) in observations are indicated by the shaded band.
Related to the slow warming, there is a net flux of O$_2$ from the ocean to the atmosphere over the industrial period (Figure 3a). The modeled long-term trend in the sea-to-air flux is interrupted following explosive volcanic eruptions that cause short-term oceanic O$_2$ uptake. The sea-to-air fluxes of O$_2$ and N$_2$ alter atmospheric O$_2$/N$_2$ by about 12.3 per meg during the 1990s in the standard simulation. This corresponds to 30% of the difference between the observed decrease in atmospheric O$_2$ and the amount of O$_2$ consumed by fossil fuel burning (Figure 1). This difference is conventionally attributed to O$_2$ release from the land biosphere associated with carbon uptake. Hence, the terrestrial carbon sink would also be reduced by 30% when considering modeled oceanic O$_2$ outgassing.

3.2. Revision of the Carbon Budget

The close correlation between ocean heat uptake and changes in atmospheric O$_2$/N$_2$ from oceanic outgassing (Figures 3b and 3c) suggests that observations of changes in ocean heat content [Levitus et al., 2000] can be utilized to estimate the O$_2$ outgassing and to revise the recent carbon budget published by the Intergovernmental Panel on Climate Change (IPCC) [Prentice et al., 2001; Manning, 2001] (Table 1). Thereby we attempt to account for internal climate variability not readily reproduced by our model.
for individual decades. Simulated sea-to-air O\(_2\) fluxes and ocean heat uptake rates are tightly correlated on multianual to multidecadal timescales (Figure 3a). A change in ocean heat uptake of 10\(^{22}\) J corresponds to an increase in atmospheric O\(_2\)/N\(_2\) of 1.56 per meg when correlating (\(r^2 = 0.94\)) simulated heat fluxes and associated O\(_2\)/N\(_2\) changes over the period 1900 to 2000 (Figures 3b and 3c). We combine this relation with data-based estimates of ocean heat uptake [Levitus et al., 2000] and atmospheric O\(_2\)/N\(_2\) and CO\(_2\) [Manning, 2001; Prentice et al., 2001]. Observed trends in the oceanic heat content are \(-0.39 \cdot 10^{22}\) J yr\(^{-1}\) for the 1980s and \(+1.24 \cdot 10^{22}\) J yr\(^{-1}\) for the period 1990 to 1995. The available ocean heat content data are extrapolated to the year 2000 assuming the same annual heat uptake rate (\(+1.24 \cdot 10^{22}\) J yr\(^{-1}\)) as during 1990 to 1995. This assumption appears to be supported by data of the heat content anomaly of the ocean’s uppermost 300 m [Levitus et al., 2000] (see section 3.4).

This new approach yields rates of carbon uptake by the terrestrial biosphere that are slightly higher during the 1980s and a factor of 2 lower during the 1990s than the central estimates published by the IPCC [Prentice et al., 2001]. As a result, oceanic CO\(_2\) uptake rates are now found to be higher during the 1990s than the 1980s in contrast to the central estimates of the IPCC, although uncertainties in the carbon budget do not allow to determine a reliable trend in the carbon sinks (Table 1). An increase in ocean CO\(_2\) uptake over the last two decades is in agreement with estimates derived from ocean models [Orr, 2000] (Table 1) and from observations of the \(^{13}\)C/\(^{12}\)C ratio of atmospheric CO\(_2\) [Joos et al., 1999a; Keeling et al., 2001].

3.3. Uncertainties in the Budget Revision

Uncertainties in the estimated sinks of anthropogenic CO\(_2\) due to uncertainties in the atmospheric trends of O\(_2\)/N\(_2\) and CO\(_2\), fossil fuel emissions, and the stoichiometric relation between CO\(_2\) released and O\(_2\) consumed during fossil fuel burning and terrestrial carbon release, are taken from Prentice et al. [2001] (Table 1). Uncertainties in the correction of the carbon sinks arise from uncertainties in the observed ocean heat uptake [Levitus et al., 2000] and in the model-derived relationship between heat uptake rates and atmospheric O\(_2\)/N\(_2\) changes due to oceanic outgassing.

The robustness of the model-derived relationship has been tested by varying key physical and biogeochemical parameters of the model and the applied radiative forcing (Appendix, Tables A1 and A2). For vertical diffusivity, governing the strength of the thermohaline overturning [Wright and Stocker, 1992] and vertical mixing of O\(_2\) and heat, set to 0.2 or 0.6 \(\cdot 10^{-4}\) m\(^2\) s\(^{-1}\) (standard: 0.4 \(\cdot 10^{-4}\) m\(^2\) s\(^{-1}\)) the estimated correction in the carbon sinks is 25% smaller or 31% larger than in the standard simulation. Changes in biological export production, parameterized in the model as a function of dissolved inorganic phosphate, affect dissolved O\(_2\), but not heat uptake. A simulation was performed where the export production was kept constant. Then, the correction is 6% larger than in the standard simulation, where the export production is slightly reduced. Applying a stoichiometric ratio of O\(_2\):P of \(-138\) in the organic matter cycle (Redfield ratio), results in a 15% smaller correction compared to the standard run, where O\(_2\):P was set to \(-170\). A smaller O\(_2\):P Redfield ratio implies less formation of dissolved O\(_2\) during organic matter production and less O\(_2\) consumption during remineralization.

A different temporal evolution and different types of external forcing affect ocean heat uptake and O\(_2\) outgassing differently. Simulations were performed with the climate sensitivity set to 1.5 or 5.0\(^\circ\)C, with anthropogenic forcing only, with volcanic forcing only, with volcanic and solar forcing combined, with indirect aerosol forcing neglected or doubled. For all these cases the estimated correction in the carbon sinks varies by less than \(+16\)% around the standard value of 0.246 Gt C yr\(^{-1}\) for the 1980s and 0.778 Gt C yr\(^{-1}\) for the 1990s (Appendix, Table A1). However, when the correlation was evaluated over the periods 1800 to 2100, 2000 to 2100, or 2100 to 2500 for different emission scenarios [Nakicenovic et al., 2000], and different CO\(_2\) stabilization profiles [Schimel et al., 1997; Wigley et al., 1997], the correction was about halved compared to the standard case (Appendix, Table A2). This suggests that the O\(_2\) to heat flux ratio may decrease in the future.

Overall, the uncertainty in the correction due to uncertainties in the model-derived relationship between heat uptake rates and O\(_2\)/N\(_2\) changes due to outgassing is estimated to be around 30% based on these additional simulations where model parameters and forcing were varied. This corresponds to uncertainties in the corrections of \(+0.1\) Gt C yr\(^{-1}\) for the 1980s and \(\pm 0.2\) Gt C yr\(^{-1}\) for the 1990s.

The uncertainty in the correction of the carbon sinks due to uncertainty in the ocean heat content data for the 1980s is estimated to be \(\pm 0.2\) Gt C yr\(^{-1}\) by combining the errors in the observed heat content anomaly at the beginning and the end of the decade [Levitus et al., 2000] and assuming them to be independent. Heat uptake rates during the 1990s are estimated to be uncertain by a factor of two due to the data extrapolation for the period 1995 to 2000; this alone yields an uncertainty in the correction of \(\pm 0.4\) Gt C yr\(^{-1}\).

Combined uncertainties are calculated by gaussian error propagation. This yields a total correction of the carbon sinks of \(0.25 \pm 0.22\) Gt C yr\(^{-1}\) for the 1980s and \(0.78 \pm 0.44\) Gt C yr\(^{-1}\) for the 1990s. This correction is applied to the carbon budgets of the IPCC for the 1980s and 1990s [Prentice et al., 2001; Manning, 2001] (Table 1). Then, best estimates and uncertainties of the revised CO\(_2\) uptake rates by the terrestrial biosphere are \(0.4 \pm 0.7\) Gt C yr\(^{-1}\) for the 1980s and \(0.7 \pm 0.8\) Gt C yr\(^{-1}\) for the 1990s (Table 1).

3.4. An Alternative Scenario for Ocean Heat Uptake

We have additionally applied an alternative scenario for ocean heat uptake to further investigate the robustness of our approach. Observations of the heat content anomaly for the uppermost 300 m of the ocean are used instead for the uppermost 3000 m. Heat data for the top 300 m are available as annual values until 1998, whereas the available heat data for the top 3000 m are 5-year running mean values that are only available until 1995 [Levitus et al., 2000]. Besides this difference in the applied heat data, the same
method as described in section 3.2 is used to determine the correction of the carbon budget.

[21] Observed trends in the oceanic heat content of the uppermost 300 m are \(-0.16 \cdot 10^{22} \, \text{J} \, \text{yr}^{-1}\) for the 1980s and \(+0.67 \cdot 10^{22} \, \text{J} \, \text{yr}^{-1}\) for the period 1990 to 1998. The uppermost 300 m of the World Oceans exhibit a strong warming after year 1995. Simulated changes in the oceanic heat content of the top 300 m are tightly correlated on multiannual to multidecadal timescales with simulated changes in atmospheric \(\text{O}_2/\text{N}_2\) due to outgassing. A change in heat content of the top 300 m of \(10^{22} \, \text{J}\) corresponds to an increase in atmospheric \(\text{O}_2/\text{N}_2\) of 2.25 per meg \((r^2 = 0.81, \text{period}\, 1900\text{ to } 2000)\) for the standard model setup. The heat content anomaly for the uppermost 300 m of the ocean is extrapolated for the year 1999 assuming the same annual change in heat content \((+0.67 \cdot 10^{22} \, \text{J} \, \text{yr}^{-1})\) as during 1990 to 1998. The deduced corrections in the terrestrial carbon sink are \(+0.15 \, \text{Gt C yr}^{-1}\) for the 1980s and \(-0.60 \, \text{Gt C yr}^{-1}\) for the 1990s. These values are smaller than the corrections obtained from the top 3000 m heat data, but still within the uncertainties of the corrections provided above. Thus, the 300 m heat data support our extrapolation of the trend in total ocean heat uptake for the second half of the 1990s, that was used to revise the carbon budget in section 3.2. Data-based estimates of oceanic heat content anomaly for the entire ocean and the period 1995 to 2000 will probably become available in the near future. This will provide a better basis for the estimated correction in the carbon budget.

### 3.5. Modeled Versus Observed Changes in Oceanic Dissolved \(\text{O}_2\)

[22] The simulated oceanic \(\text{O}_2\) concentration changes are small and roughly consistent with previous results based on oceanic observations. Modeled basin-average changes (1970s to 1990s) are less than 5 \(\mu\text{mol kg}^{-1}\), and largest changes are found in the upper 1000 m in the Pacific, South Atlantic, Indian, and Southern Oceans, and below 1500 m in the North Atlantic (Figures 4a and 4b). Such small long-term changes are difficult to detect from observations. Interpolation of sparse data may introduce biases. Cruises at different times may yield different results due to natural variability, changes in water masses, or intercruise calibration differences. To account for these effects, analyses are usually performed for saturation-normalized \(\text{O}_2\) concentrations on density surfaces, and intercruise deep-water concentration differences are assumed to reflect calibration differences.

[23] Modeled saturation-normalized \(\text{O}_2\) concentrations are averaged on density surfaces (referenced to surface pressure) and compared to previous results based on oceanic observations for the North Pacific. Saturation-normalized \(\text{O}_2\) decreased by up to 5 \(\mu\text{mol kg}^{-1}\) in the upper ocean, and increased slightly below 1000 m (Figure 4c). The simulated changes in the upper water column and below 2000 m are consistent with the \(\text{O}_2\) changes estimated by Keller et al. [2002] for the entire North Pacific, analyzing \(\text{O}_2\) measurements from the 1970s and 1990s. However, \(\text{O}_2\) changes between 1000 and 2000 m are smaller in the model than estimated by Keller et al. [2002].

[24] Modeled trends in \(\text{O}_2\) concentrations are generally less than 1 \(\mu\text{mol kg}^{-1}\) per decade in all basins. Larger changes are simulated only in the top 1000 m in the circumpolar region (reductions up to 5 \(\mu\text{mol kg}^{-1}\) per decade) and in the northermost grid cells in the Atlantic and Pacific Oceans which exhibit marked changes in convection resulting in zonally averaged \(\text{O}_2\) reductions of up to 16 \(\mu\text{mol kg}^{-1}\) per decade. The simulated changes in the zonally averaged model are qualitatively comparable to large local changes observed in the circumpolar region [Matear et al., 2000], in middle and high latitudes of the North Pacific region [Emerson et al., 2001; Kim et al., 2000; Ono et al., 2001; Watanabe et al., 2001], in the South Pacific [Shaffer et al., 2000], in the Indian Ocean [Bindoff and McDougall, 2000], and in the North Atlantic [Garcia et al., 1998; Garcia and Keeling, 2001].
[25] Matear et al. [2000] found reductions in dissolved O$_2$ of up to 6 \textmu mol kg$^{-1}$ per decade in the circumpolar region ($53^\circ$S -- $61^\circ$S, 110$^\circ$E -- 160$^\circ$E) by analyzing data from the recent World Ocean Circulation Experiment (WOCE) and from the 1968 Eltanin cruise. It is an open question whether this decrease is representative for the entire Southern Ocean. Emerson et al. [2001] found local reductions in saturation-normalized O$_2$ of up to 30 \textmu mol kg$^{-1}$ in data from cruises in the 1980s and 1990s along 150$^\circ$W in the North Pacific. A large O$_2$ decrease is also found in available long-term studies. Kim et al. [2000] report large long-term decreases in oceanic O$_2$ concentrations of more than 20 \textmu mol kg$^{-1}$ mainly below 1000 m in the East Sea (Japan Sea) since the mid-1950s and of nearly 10 \textmu mol kg$^{-1}$ since 1979. Ono et al. [2001] and Watanabe et al. [2001] found a reduction in dissolved oxygen of up to 30 \textmu mol kg$^{-1}$ superimposed on a bidecadal oscillation in subsurface waters in the western subarctic Pacific (Oyashio domain) over the period from 1968 to 1998. Shaffer et al. [2000] report decreases of dissolved O$_2$ above 3000 m at 28$^\circ$S in the eastern South Pacific off Chile (72$^\circ$W -- 88$^\circ$W) since 1967, ranging from about 18 \textmu mol kg$^{-1}$ at 300 m to about 4 \textmu mol kg$^{-1}$ at 2000 m. Bindoff and McDougall [2000] observed a pronounced decrease in O$_2$ concentrations of 7 to 8 \textmu mol kg$^{-1}$ between 300 and 1000 m depth occurring at 32$^\circ$S in the Indian Ocean (averaged from 30$^\circ$E -- 117$^\circ$E) over the period 1962 to 1987. Finally, Garcia et al. [1998] found a significant decrease in O$_2$ concentration between 700 and 1800 m at 24$^\circ$N in the North Atlantic over the period 1981 to 1992. The zonally averaged decrease in the O$_2$ concentration is between 3 and 7 \textmu mol kg$^{-1}$, with the maximum occurring around 1100 m.

[26] In summary, detectable decreases in O$_2$ have been observed in intermediate waters in the North Pacific, North Atlantic, South Pacific, and South Indian Oceans, and small increases have occurred in deeper waters in the North Pacific and South Indian Oceans [Keeling and Garcia, 2002]. These observations support the simulated long-term decrease in the oceanic O$_2$ inventory.

3.6. Mechanisms Driving the Modeled Changes in Dissolved O$_2$

[27] Finally, we examine the mechanisms responsible for the changes in the modeled oceanic O$_2$ concentrations. Two additional model simulations were performed to quantify the contribution of different mechanisms to changes in O$_2$ concentrations. In a first simulation, sea surface temperatures and salinities from the standard simulation ($\Delta T_{2\text{m}} = 3.7^\circ$C) were prescribed in the routines to calculate solubilities of O$_2$ and N$_2$ while running the model in the constant-climate baseline setup ($\Delta T_{2\text{m}} = 0^\circ$C) up to year 2000. In a second simulation, the biological export production was kept constant, while otherwise the configuration was identical to the standard simulation. The main cause for the reduction in dissolved O$_2$ and the increase in the net sea-to-air O$_2$ flux in the standard simulation is a reduction in the transport rate of O$_2$ to depth due to changes in ocean circulation and convection. This is also supported by various studies that identify ocean circulation changes as the main cause of observed O$_2$ reductions [Emerson et al., 2001; Keller et al., 2002; Kim et al., 2000; Watanabe et al., 2001; Ono et al., 2001; Shaffer et al., 2000; Bindoff and McDougall, 2000]. The small reduction in simulated biological export production in the standard run tends to slightly increase the marine O$_2$ inventory and to reduce outgassing. Solubility changes, mainly driven by sea surface warming, are responsible for about 22% of the simulated reduction in the marine oxygen inventory, but for only about 10% of the changes in atmospheric O$_2$/N$_2$ as the solubility of both O$_2$ and N$_2$ is reduced. This highlights the importance of ocean circulation changes for atmospheric O$_2$/N$_2$.

4. Discussion and Conclusion

[28] Data-based and model studies suggest that the oceanic O$_2$ inventory has decreased over the past decades. In our standard global warming simulation, the oceanic O$_2$ inventory decreases by 0.42 \cdot 10^{14} \text{ mol yr}^{-1} over the 1990s, well within the range of 0.2 to 0.7 \cdot 10^{14} \text{ mol yr}^{-1} found by others [Sarmiento et al., 1998; Matear et al., 2000; Plattner et al., 2001; Bopp et al., 2002; Keeling and Garcia, 2002; Matear and Hirst, submitted manuscript, 2002]. As discussed in section 3.5, a long-term decrease in dissolved O$_2$ concentration is observed in different oceanic regions.

[29] A close relationship between sea-to-air oxygen fluxes and heat fluxes is found in many modeling studies [Sarmiento et al., 1998; Plattner et al., 2001; Bopp et al., 2002] as well as in a data-based study [Keeling and Garcia, 2002]. The ratios between O$_2$ to heat flux and N$_2$ to heat flux are 5.9 nmol J$^{-1}$ and 2.1 nmol J$^{-1}$ over the period 1900 to 2000 in our standard simulation. This compares well with O$_2$ to heat flux ratios of about 6 nmol J$^{-1}$ found in simulations with increasing atmospheric CO$_2$ [Sarmiento et al., 1998; Bopp et al., 2002] and of 5 nmol J$^{-1}$ derived from oceanic temperature and dissolved O$_2$ observations [Keeling and Garcia, 2002]. Keeling and Garcia [2002] also report a comparable N$_2$ to heat flux ratio (2.2 nmol J$^{-1}$). Sensitivity simulations (Appendix, Table A1) suggest that the O$_2$ to heat flux ratio over the industrial period is relatively insensitive to variations in model parameters and to variations in radiative forcing. However, future scenario simulations (Appendix, Table A2) show that the O$_2$ to heat flux ratio is likely to decrease in the coming decades.

[30] Corrections of the recent carbon budget of the IPCC [Prentice et al., 2001] are suggested in three independently developed studies. Similar to this study, Keeling and Garcia [2002] and Bopp et al. [2002] combine estimates of the O$_2$ to heat flux ratio with estimates of ocean heat uptake to compute sea-to-air O$_2$ fluxes and revise carbon budgets. Similar values of the O$_2$ to heat flux ratio are applied. However, the three studies differ fundamentally in the way ocean heat uptake is considered. Keeling and Garcia [2002] applied a model-derived estimate of the long-term ocean warming trend [Levitus et al., 2001; Barnett et al., 2001] and neglected decadal scale variability in heat uptake. On the other hand, ocean heat content data of Levitus et al. [2000] are used here and in the study of Bopp et al. [2002], assuming that O$_2$ outgassing is related to both the long-term warming trend and the decadal scale variability. The analysis by Bopp et al. [2002] of decadal scale variations in their model and our sensitivity simulation where the model is
driven by the pulse-like volcanic forcing only (Appendix, Table A1) suggest that O₂ outgassing is coupled to decadal scale variations in heat uptake.

[31] The two different approaches yield different corrections for the carbon budget due to O₂ outgassing. The consideration of long-term warming only yields a small upward revision in ocean carbon uptake of +0.2 Gt C yr⁻¹ as suggested by Keeling and Garcia [2002]. This correction would apply for both the 1980s and 1990s. Consideration of the large decadal variability in the heat anomaly data yields a substantial upward revision of ocean carbon uptake for the 1990s, but a small negative correction for the 1980s. Bopp et al. [2002] estimated a correction in the ocean carbon sink of +0.5 ± 0.5 Gt C yr⁻¹ for the period 1990 to 1996 and of −0.1 ± 0.5 Gt C yr⁻¹ for the 1980s comparable to our estimates of +0.7 ± 0.5 Gt C yr⁻¹ and −0.2 ± 0.3 Gt C yr⁻¹.

[32] In conclusion, oceanic O₂ changes need to be considered to determine a self-consistent estimate of the terrestrial and oceanic sinks of anthropogenic CO₂ from atmospheric O₂/N₂ observations. We propose to monitor trends in dissolved O₂ by routine measurements from ships or buoys to further narrow uncertainties in estimates of the carbon sinks and to resolve the discrepancies between the different studies discussed above.

[33] Estimates of the oceanic carbon sink obtained by different approaches [Prentice et al., 2001; Orr, 2000; Joos et al., 1999a; Keeling et al., 2001; Rayner et al., 1999] are consistent when taking into account their uncertainties. However, the central estimates of the IPCC budget show a lower oceanic carbon uptake during the 1990s than the 1980s in contrast to results from process-based carbon cycle models and estimates based on measurements of the 13C/12C ratio of atmospheric CO₂. The consideration of oceanic O₂ outgassing reconciles the central estimates from atmospheric O₂/N₂ data with results from ocean models and analyses of carbon isotopes.

[34] Our results suggest that the global net carbon uptake by the land biosphere during the 1990s was about a factor of two lower than that recently estimated by the IPCC [Prentice et al., 2001]. This is of relevance as the performance of process-based carbon cycle models is evaluated against the recent carbon budget [McGuire et al., 2001; Orr et al., 2001].

### Appendix A

#### A1. Implementation of the Biogeochemical Cycles of O₂ and N₂ in the Model

[35] The atmospheric budget equations for O₂ and N₂ are included in the model by

\[
\frac{\partial F_{atm}}{\partial t} = -F_{air-sea} + 1.39 \cdot F_{fossil} + 1.1 \cdot F_{air-biota} \tag{A1}
\]

and

\[
\frac{\partial F_{sea}}{\partial t} = -F_{air-sea}, \tag{A2}
\]

where \(F_{atm}\) and \(F_{sea}\) are the atmospheric inventories of O₂ and N₂, and \(F_{air-sea}\) and \(F_{air-sea}\) are the net air-to-sea gas exchange of O₂ and N₂. \(F_{fossil}\) denotes the carbon emissions from fossil fuel burning and \(F_{air-biota}\) the net atmosphere to land biota flux of CO₂. The fossil fuel O₂:CO₂ combustion ratio of −1.39 and the land biota O₂:CO₂ exchange ratio of −1.1 are taken from Manning [2001] and Severinghaus [1995], respectively. O₂ and N₂ are allowed to evolve freely in the atmosphere after being initialized by prescribing the preindustrial mole fractions of O₂ and N₂ in the atmosphere. Preindustrial atmospheric mole fractions of O₂ and N₂ are set to 209.460 ppm and 780.840 ppm [see Manning et al., 1999, and references therein]. All inventories, concentrations and fluxes are expressed in molar units in the model.

[36] \(F_{air-biota}\) is the sum of the CO₂ flux from deforestation and land-use change, \(F_{defor}\), and the modeled CO₂ flux into the terrestrial biosphere due to fertilization, \(F_{fert}\):

\[
F_{air-biota} = F_{fert} - F_{defor}, \tag{A3}
\]

[37] \(F_{defor}\) is calculated by closing the atmospheric CO₂ budget (single deconvolution [Siegenthaler and Oeschger, 1987]) from prescribed atmospheric CO₂ [Keeling and Whorf, 2000] and fossil fuel emissions [Marland et al., 1995], and from the modeled fertilization flux, \(F_{fert}\), and CO₂ air-sea gas exchange, \(F_{air-sea}^C\):

\[
F_{defor} = \frac{\partial F_{atm}}{\partial t} - F_{fossil} + F_{air-sea}^C + F_{fert}. \tag{A4}
\]

[38] The net air-sea fluxes of O₂ and N₂ are calculated as the product of the gas transfer coefficient, \(k_g\), and the profile pressure difference between surface air and water:

\[
F_{air-sea}^T = k_g (\rho T_{sat} - \rho T_w) = k_w ([T_{sat}] - [T_w]), \tag{A5}
\]

where the terms \(\rho T_{sat}\) and \(\rho T_w\) denote the saturation and the modeled partial pressures of a tracer \(T\) in seawater. The partial pressures of O₂ and N₂ in seawater are calculated from the concentrations of dissolved O₂ and N₂ in seawater ([T_sat] and [T_w]) and the solubilities of Weiss [1970]. The coefficient \(k_w\) equals the product of the solubility [Weiss, 1970] and the gas transfer velocity related to the liquid phase, \(k_w\). \(k_w\) is calculated applying the formulation of Wanninkhof [1992] for long-term winds and a wind speed climatology [Esbensen and Kushnir, 1981]. Results obtained by applying equation A5 are nearly identical to results obtained assuming dissolution equilibrium with atmospheric O₂ [Plattner et al., 2001] as the equilibration time of the ocean mixed layer with O₂ is short (about 20 days) [Broecker and Peng, 1982].

[39] Tracers are implemented in the ocean as described by Marchal et al. [1998]. The conservation equations for O₂ and N₂ are

\[
\frac{\partial T_{oce}}{\partial t} = -\mathbf{u} \cdot \nabla T_{oce} + \nabla \cdot [\kappa \nabla T_{oce}] + q_{conv}^T + J_{bio}^T + J_{flx}^T, \tag{A6}
\]

where \(T_{oce}\) is the tracer concentration in the ocean. The first term on the right describes advective transport (with the velocity vector \(\mathbf{u}\), the second term on the right describes diffusive transport (with the the diffusion tensor \(\kappa\)), and \(q_{conv}\) denotes the transport by convection. The terms \(J_{bio}^T\) and
Table A1. Ratio of Δ(O2/N2) to Heat Flux: Sensitivity on Model Parameters and Radiative Forcinga

<table>
<thead>
<tr>
<th>Model Setup</th>
<th>Δ(O2/N2)/Heat Flux [per meg/10^{22} J]</th>
<th>r², 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ΔT₂ₓ = 3.7°C, standard simulation</td>
<td>1.56 (—)</td>
<td>0.95</td>
</tr>
</tbody>
</table>

**Climate Sensitivity, ΔT₂ₓ**

<table>
<thead>
<tr>
<th>ΔT₂ₓ</th>
<th>Δ(O2/N2)/Heat Flux [per meg/10^{22} J]</th>
<th>r², 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5°C</td>
<td>1.43 (—9%)</td>
<td>0.90</td>
</tr>
<tr>
<td>5.0°C</td>
<td>1.55 (—11%)</td>
<td>0.95</td>
</tr>
</tbody>
</table>

**Radiative Forcing and Climate Sensitivity, ΔT₂ₓ**

<table>
<thead>
<tr>
<th>ΔT₂ₓ</th>
<th>Δ(O2/N2)/Heat Flux [per meg/10^{22} J]</th>
<th>r², 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5°C, anthropogenic forcing only</td>
<td>1.46 (—6%)</td>
<td>0.98</td>
</tr>
<tr>
<td>5.0°C, anthropogenic forcing only</td>
<td>1.73 (—11%)</td>
<td>0.98</td>
</tr>
<tr>
<td>3.7°C, volcanic forcing only</td>
<td>1.36 (—13%)</td>
<td>0.98</td>
</tr>
<tr>
<td>3.7°C, volcanic and solar forcing only</td>
<td>1.80 (—16%)</td>
<td>0.92</td>
</tr>
<tr>
<td>3.7°C, no indirect aerosol forcing</td>
<td>1.77 (—14%)</td>
<td>0.89</td>
</tr>
<tr>
<td>3.7°C, doubled indirect aerosol forcing</td>
<td>1.51 (—3%)</td>
<td>0.98</td>
</tr>
<tr>
<td>3.7°C, anthropogenic forcing only, starting 1765</td>
<td>1.65 (—6%)</td>
<td>0.95</td>
</tr>
<tr>
<td>3.7°C, scenario WRE starting 1765, CO₂ forcing only</td>
<td>1.46 (—6%)</td>
<td>0.98</td>
</tr>
<tr>
<td>3.7°C, scenario WRE starting 1765, CO₂ forcing only</td>
<td>1.17 (—25%)</td>
<td>0.90</td>
</tr>
</tbody>
</table>

**Marine Biology**

<table>
<thead>
<tr>
<th>ΔT₂ₓ</th>
<th>Δ(O2/N2)/Heat Flux [per meg/10^{22} J]</th>
<th>r², 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.7°C, constant export production</td>
<td>1.65 (—6%)</td>
<td>0.94</td>
</tr>
<tr>
<td>3.7°C, stoichiometric ratio O₂:P set to —138</td>
<td>1.33 (—15%)</td>
<td>0.95</td>
</tr>
</tbody>
</table>

**Ocean Circulation and Mixing**

<table>
<thead>
<tr>
<th>ΔT₂ₓ</th>
<th>Δ(O2/N2)/Heat Flux [per meg/10^{22} J]</th>
<th>r², 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.7°C, vertical diffusivity 0.2 · 10^{-4} m² s⁻¹</td>
<td>1.18 (—25%)</td>
<td>0.92</td>
</tr>
<tr>
<td>3.7°C, vertical diffusivity 0.6 · 10^{-4} m² s⁻¹</td>
<td>2.03 (—31%)</td>
<td>0.87</td>
</tr>
</tbody>
</table>

*aThe model-derived relationship between heat uptake rates and O₂/N₂ changes due to outgassing is calculated from model output (5-year running means) of the period 1900 to 2000. The relationship is given in per meg/10^{22} J for each individual simulation and additionally as %-difference (in parentheses) from the ratio of Δ(O2/N2) to heat flux obtained from the standard simulation. The resulting correction in the carbon budget scales linearly with the Δ(O2/N2) to heat flux ratio. The term r² denotes the coefficient of determination for each individual correlation.

J>Tflux represent source minus sink terms due to biological processes and air-sea fluxes. The term J>Tbio is zero for N₂ and the total inventory of N₂ in the atmosphere-ocean system is conserved.

**A2. Computing the Correction of the Carbon Budget**

[40] Changes in the modeled atmospheric O₂/N₂ ratio are expressed relative to the preindustrial ratio in units of per meg according to Keeling et al. [1996]:

\[ b(O_2/N_2) = \left( \frac{O_2/N_2}{O_2/N_2}_{\text{ref}} - 1 \right) \cdot 10^6. \]  

(A7)

Normalization to N₂ eliminates variations associated with changes in humidity and other atmospheric changes unrelated to O₂ and CO₂ fluxes [Bender and Battle, 1999].

[41] The change in atmospheric O₂/N₂ from air-sea gas exchange (in per meg yr⁻¹), Δ(O₂/N₂), is calculated as the difference between changes in O₂/N₂ from the standard simulation (ΔT₂ₓ = 3.7°C) and the baseline simulation (ΔT₂ₓ = 0°C):

\[ \Delta(O_2/N_2) = b(O_2/N_2)_{\text{rad}} - b(O_2/N_2)_{\text{base}}. \]  

(A8)

[42] Simulated Δ(O₂/N₂) and simulated heat fluxes are correlated to compute the ratio between atmospheric O₂/N₂ changes due to outgassing and ocean heat uptake (see Figures 3b and 3c). Then, observed changes in ocean heat content [Levitt et al., 2000] are multiplied by this model-derived ratio to estimate the change in atmospheric O₂/N₂ due to ocean outgassing, Δ(O₂/N₂)*, for the 1980s and 1990s. By considering Δ(O₂/N₂)* instead of the modeled Δ(O₂/N₂), we account for decadal scale climate variability reflected in the ocean heat data, but not readily simulated for a particular decade.

[45] The correction in the terrestrial and oceanic carbon sinks (in Gt C yr⁻¹) is then obtained by:

\[ \Delta F_{\text{air-sea}}^C = -\Delta F_{\text{air-sea}}^C = \frac{1}{2.49} \cdot \Delta(O_2/N_2)^*. \]  

(A9)

The factor \( \frac{1}{2.49} \) arises from the conversion of per meg O₂/N₂ to gigatons of carbon [Battle et al., 2000].

**A3. Sensitivity and Scenario Simulations**

[44] Uncertainties in the correction of the carbon sinks arise from uncertainties in the model-derived relationship between heat uptake rates and O₂/N₂ changes due to outgassing. The robustness of this relationship has been tested by varying key physical and biogeochemical parameters of the model, the applied radiative forcing, and the period over which model output has been used to derive the relationship.

[45] In Table A1, results from sensitivity simulations on model parameters and on the applied radiative forcing are shown. The model-derived relationship is calculated from 5-year running mean model output of the period 1900 to 2000. Using 1-year running mean or monthly-mean values does not alter results significantly. The calculated slope for the
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Table A2. Ratio of $\Delta(O_2/N_2)$ to Heat Flux: Sensitivity on the Period Used to Derive the Relationship Between Heat Uptake Rates and $O_2/N_2$ Changes due to Outgassing*

<table>
<thead>
<tr>
<th>Model setup</th>
<th>$\Delta(O_2/N_2)/heat \text{ flux}$</th>
<th>$r^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>[per meg/$10^{-22}$ J]</td>
<td></td>
</tr>
</tbody>
</table>

Period 1400–2000

- $\Delta T_{2\times} = 3.7^\circ C$, standard simulation: $1.63(\pm5\%)$ 0.96
- $\Delta T_{2\times} = 3.7^\circ C$, volcanic forcing only: $1.72(\pm10\%)$ 0.93
- $\Delta T_{2\times} = 3.7^\circ C$, volcanic solar forcing only: $1.73(\pm11\%)$ 0.93

Period 1800–2100

- $\Delta T_{2\times} = 3.7^\circ C$, anthropogenic forcing only: $0.98(\pm37\%)$ 0.98
- $\Delta T_{2\times} = 1.5^\circ C$, anthropogenic forcing only: $1.12(\pm28\%)$ 0.97
- $\Delta T_{2\times} = 5.0^\circ C$, anthropogenic forcing only: $0.89(\pm43\%)$ 0.98
- $\Delta T_{2\times} = 3.7^\circ C$, scenario B1 starting 1765, anthropogenic forcing only: $0.98(\pm38\%)$ 0.98
- $\Delta T_{2\times} = 3.7^\circ C$, scenario A1FI starting 1765, anthropogenic forcing only: $0.77(\pm51\%)$ 0.97
- $\Delta T_{2\times} = 3.7^\circ C$, scenario WRE550 starting 1765, CO$_2$ forcing only: $0.93(\pm40\%)$ 0.97
- $\Delta T_{2\times} = 3.7^\circ C$, scenario WRE1000 starting 1765, CO$_2$ forcing only: $0.86(\pm45\%)$ 0.97

Period 2000–2100

- $\Delta T_{2\times} = 3.7^\circ C$, anthropogenic forcing only: $0.78(\pm50\%)$ 0.94
- $\Delta T_{2\times} = 1.5^\circ C$, anthropogenic forcing only: $0.64(\pm59\%)$ 0.88
- $\Delta T_{2\times} = 5.0^\circ C$, anthropogenic forcing only: $0.65(\pm58\%)$ 0.96
- $\Delta T_{2\times} = 3.7^\circ C$, scenario B1 starting 1765, anthropogenic forcing only: $0.78(\pm50\%)$ 0.93
- $\Delta T_{2\times} = 3.7^\circ C$, scenario A1FI starting 1765, anthropogenic forcing only: $0.62(\pm60\%)$ 0.97

Period 2100–2500

- $\Delta T_{2\times} = 3.7^\circ C$, scenario WRE550 starting 1765, CO$_2$ forcing only: $0.94(\pm40\%)$ 0.88
- $\Delta T_{2\times} = 3.7^\circ C$, scenario WRE1000 starting 1765, CO$_2$ forcing only: $0.73(\pm53\%)$ 0.46

*The model-derived relationship between heat uptake rates and $O_2/N_2$ changes due to outgassing is calculated from model output (5-year running means) of the periods 1400 to 2000, 1800 to 2100, 2000 to 2100, and 2100 to 2500. The relationship is given in per meg/$10^{-22}$ J for each individual simulation and additionally as %-difference (in parentheses) from the ratio of $\Delta(O_2/N_2)$ to heat flux obtained from the standard simulation (see Table A1). The resulting correction in the carbon budget scales linearly with the $\Delta(O_2/N_2)$ to heat flux ratio. The term $r^2$ denotes the coefficient of determination for each individual correlation.

standard model setup is reduced from 1.56 per meg/$10^{-22}$ J ($r^2 = 0.94$) to 1.51 per meg/$10^{-22}$ J ($r^2 = 0.96$; 1-year running means) or 1.50 per meg/$10^{-22}$ J ($r^2 = 0.86$; monthly means), respectively. We note that the resulting correction in the carbon budget scales linearly with the $\Delta(O_2/N_2)$ to heat flux ratio.

1. Climate sensitivity: The climate sensitivity, $\Delta T_{2\times}$, in the standard simulation set to 3.7°C, has been varied between 1.5 and 5.0°C.

2. Radiative forcing: In the standard simulation radiative forcing from reconstructed (until year 2000) and later projected changes in the abundances of greenhouse gases and aerosols in the atmosphere was calculated based on simplified expressions [see Joos et al., 2001, and references therein]. The radiative forcing from changes in atmospheric CO$_2$, CH$_4$, N$_2$O, stratospheric and tropospheric O$_3$, stratospheric H$_2$O due to CH$_4$ changes, SF$_6$, 28 halocarbons (including those controlled by the Montreal Protocol), from direct and indirect effects of sulfate aerosols, and from direct forcing of black and organic carbon has been considered. Radiative forcing by explosive volcanic eruptions and solar irradiance changes was prescribed for the last millennium [Crowley, 2000]. Additional simulations have been performed where the applied radiative forcing has been varied between anthropogenic forcing only, volcanic forcing only, volcanic and solar forcing only, and with the indirect aerosol forcing excluded or doubled.

3. Marine biology: Two additional simulations were performed to test the sensitivity on parameters of the biogeochemical model. In the first simulation the export production was kept constant. In the second simulation a stoichiometric ratio of O$_2$:P of −138 in the organic matter cycle (Redfield ratio) has been applied instead of −170 as in the standard.

4. Ocean circulation and mixing: The vertical diffusivity, governing the strength of the thermohaline overturning [Wright and Stocker, 1992] and vertical mixing of O$_2$ and heat, has been varied between 0.2 and $0.6 \cdot 10^{-3}$ m$^2$ s$^{-1}$. In the standard simulation it is set to 0.4 $\cdot 10^{-3}$ m$^2$ s$^{-1}$.

[46] In Table A2, results from sensitivity and scenario simulations are listed. The model-derived relationship between ocean heat uptake rates and $O_2/N_2$ changes due to outgassing is calculated using model output (5-year running means) from the periods 1400 to 2000, 1800 to 2100, 2000 to 2100, and 2100 to 2500, in contrast to Table A1 where model output from the period 1900 to 2000 is used. Future emissions of CO$_2$, non-CO$_2$ greenhouse gases and aerosols are prescribed from the IPCC SRES emission scenarios [Nakićenović et al., 2000] or the WRE CO$_2$ stabilization profiles [Schimel et al., 1997; Wigley et al., 1997].

[47] Acknowledgments. We thank K. Keller and R. Matear for providing data-based estimates for the trend in oceanic oxygen for the North Pacific and the Southern Ocean. We also thank K. Keller, S. Emerson, M. Keeling, K.-R. Kim, L. Bopp, and R. Matear for providing preprints of their papers, and A. Manning for a copy of his Ph. D. thesis. Comments by R. Keeling, K. Keller, O. Marchal, R. Najjar, and I.
References


