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Nitrification amplifies the decreasing trends of atmospheric oxygen and implies a larger land carbon uptake

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[1] Atmospheric O_2 trend measurements are used to partition global oceanic and land biotic carbon sinks on a multiannual basis. The underlying principle is that a terrestrial uptake or release of CO_2 is accompanied by an opposite flux of O_2. The molar ratio of the CO_2 and O_2 terrestrial fluxes should be 1, if no other elements are considered. However, reactive nitrogen produced by human activities (e.g., fertilizers, N deposition) is also being incorporated into plant tissues. The various reaction pathways of the terrestrial nitrogen cycle cause fluxes of atmospheric O_2. Thus the cycles of nitrogen, carbon, and oxygen must be linked together. We report here on previously unconsidered anthropogenic nitrogen-related mechanisms which impact atmospheric O_2 trends and thus the derived global carbon sinks. In particular, we speculate that anthropogenic-driven changes are driving the global nitrogen cycle to a more oxidized state, primarily through nitrification, nitrate fertilizer industrial production, and combustion of fossil fuels and anthropogenic biomass burning. The sum of these nitrogen-related processes acts to additionally decrease atmospheric O_2 and slightly increase atmospheric CO_2. We have calculated that the effective land biotic O_2:CO_2 molar ratio ranges between 0.76 and 1.04 rather than 1.10 (moles of O_2 produced per mole of CO_2 consumed) over the period 1993–2003, depending on which of four contrasting nitrogen oxidation and reduction pathway scenarios is used. Using the scenario in which we have most confidence, this implies a 0.23 PgC yr^{-1} correction to the global land biotic and oceanic carbon sinks of most recently reported estimates over 1993–2003, with the land biotic sink becoming larger and the oceanic sink smaller. We have attributed large uncertainties of 100% to all nitrogen-related O_2 and CO_2 fluxes and this corresponds up to ±0.09 PgC yr^{-1} increase in global carbon sink uncertainties. Thus accounting for anthropogenic nitrogen-related terrestrial fluxes of O_2 results in a 45% larger land biotic sink of 0.74 ± 0.78 PgC yr^{-1} and a slightly smaller oceanic sink of 2.01 ± 0.66 PgC yr^{-1} for the decade 1993–2003.

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1. Introduction

[2] High-precision measurements of atmospheric oxygen trends are used to partition the uptake of anthropogenic CO_2 between land and oceans [see, e.g., *Keeling and Shertz*, 1992]. The quantification of the global carbon budget by the Third Assessment Report of the Intergovernmental Panel on Climate Change [*Prentice et al.*, 2001] rests upon atmospheric oxygen measurements. The underlying principle is that the dissolution of anthropogenic CO_2 into the ocean has no O_2 counterflux, whereas the uptake of CO_2 by terrestrial plants is mirrored by an opposing O_2 flux. The O_2 to CO_2 ratio in land biotic fluxes commonly used in carbon sink calculations is 1.10 ± 0.05 moles of O_2 produced per mole of CO_2 consumed [*Severinghaus*, 1995; *Manning and Keeling*, 2006]. Photosynthesis emits O_2 to the atmosphere and respiration is usually assumed to absorb atmospheric O_2 following this same ratio. Thus the mass of oxygen in the terrestrial biosphere decreases over time if photosynthesis exceeds respiration. However, determining the precise value of this ratio is problematic [*Manning and Keeling*, 2006; *Seibt et al.*, 2004], particularly for respiration, which could have large variations over temporal scales and ecosystem ranges [*Randerson et al.*, 2006]. Recent studies have suggested molar ratios between 1.0 and 1.1 [*Manning*, 2001; *Marca*, 2004; *Seibt et al.*, 2004; *Stephens et al.*, 2007], however these studies have not been very comprehensive.

[3] Although there is no oceanic O_2 counter-flux directly as a result of fossil fuel CO_2 emissions, there is an indirect effect caused by the fact that the global oceans have warmed [*Levitus et al.*, 2000]. The ocean warming signal, combined with changes in ocean dynamics and biology, has likely

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