Global and hemispheric CO_2 sinks deduced from changes in atmospheric O_2 concentration

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The global budget for sources and sinks of anthropogenic CO_2 has been found to be out of balance unless the oceanic sink is supplemented by an additional 'missing sink', plausibly associated with land biota[1,25]. A similar budgeting problem has been found for the Northern Hemisphere alone[2,3], suggesting that northern land biota may be the sought-after sink, although this interpretation is not unique[2–5]; to distinguish oceanic and land carbon uptake, the budgets rely variously, and controversially, on ocean models[2,6,7], $^{13}CO_2/^{12}CO_2$ data[2,4,5], sparse oceanic observations of p_{CO_2} (ref. 3) or $^{13}C/^{12}C$ ratios of dissolved inorganic carbon,[4,5,8] or single-latitude trends in atmospheric O_2 as detected from changes in O_2/N_2 ratio.[9,10] Here we present an extensive O_2/N_2 data set which shows simultaneous trends in O_2/N_2 in both northern and southern hemispheres and allows the O_2/N_2 gradient between the two hemispheres to be quantified. The data are consistent with a budget in which, for the 1991–94 period, the global oceans and the northern land biota each removed the equivalent of approximately 30% of fossil-fuel CO_2 emissions, while the tropical land biota as a whole were not a strong source or sink.

We focus on time series from Alert (82.5° N), La Jolla (32.9° N) and Cape Grim (40.7° S), as shown in Fig. 1. The data reveal seasonal cycles, interannual trends, and gradients with latitude. We express changes in O_2/N_2 relative to a reference according to

$$\delta(O_2/N_2) = (O_2/N_2)/(O_2/N_2)_{ref} - 1$$

where we multiply $\delta(O_2/N_2)$ by 10^6 and express the results in units of 'per meg' (used here to express parts-per-million changes in a ratio). For each site we compute the annual means centred on 1 January and 1 July. We average together the annual means for La Jolla and Alert which we use as a proxy $\delta(O_2/N_2)_{NH}$ for the Northern Hemisphere as a whole, and we use the Cape Grim data as a proxy $\delta(O_2/N_2)_{SH}$ for the Southern Hemisphere as a whole. We then compute $\delta(O_2/N_2)_{GLOB} = (\delta(O_2/N_2)_{NH} + \delta(O_2/N_2)_{SH})/2$, which we use as a measure of the global mean, and $\delta(O_2/N_2)_{GRAD} = \delta(O_2/N_2)_{NH} - \delta(O_2/N_2)_{SH}$, which we use as a measure of the north–south gradient. We compute the same statistics for concurrent CO_2 data from the same flask samples.

Figure 2 shows that the temporal trends in $(CO_2)_{GLOB}$ and $\delta(O_2/N_2)_{GLOB}$ are well correlated, with a period of anomalously slow growth in CO_2 concentration centred around 1992[11,12] coinciding with a period of slower decrease in O_2/N_2. If fossil-fuel burning were the only process inducing changes, then atmospheric CO_2 and O_2/N_2 would have changed by +8.9 p.p.m. and −60.4 per meg, respectively, from 1991 to 1994. In comparison, the observed changes were 3.2 ± 0.26 p.p.m. and -42 ± 6 per meg. Assuming the N_2 concentration is constant, and that exchanges with land biota are the only additional long-term influence on O_2 levels, we can use the trend in O_2/N_2 to allocate CO_2 uptake between land biota and the oceans[9]. The system can be solved graphically, as shown in Fig. 2. We find that the land biosphere and the oceans removed 2.9 ± 1.3 and 2.4 ± 1.2 p.p.m., respectively, over the three-year period, corresponding to average annual sinks of 2.0 ± 0.9 and 1.7 ± 0.9 Pg C yr^{-1}, respectively (1 Pg = 10^{12} kg). Similar analysis using La Jolla data alone to infer global trends yields a land biotic sink of 1.8 ± 0.7 Pg C yr^{-1} and an oceanic sink of 1.9 ± 0.5 Pg C yr^{-1} over the longer period from 1989 to 1994.

The north-south gradients in atmospheric CO_2 concentrations and O_2/N_2 ratio depend on the spatial and temporal pattern of CO_2 and O_2 sources and sinks as modified by atmospheric

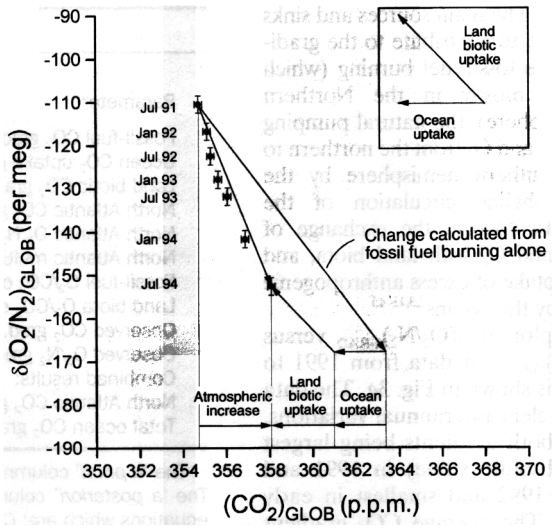

FIG. 2 Globally and annually averaged $\delta(O_2/N_2)$ versus CO_2. Annual means are computed by first fitting the data to the sum of a stiff spline plus a four-harmonic annual cycle. The harmonic fit is then used to adjust data to the 15th of each month, and monthly means are computed. Twelve consecutive monthly means are averaged to compute annual means. Months with missing data are filled in using the complete fitted curve. Also shown are the trends in CO_2 and O_2/N_2 computed from fossil-fuel burning and cement manufacture[32,27], and the unique combination of oceanic and land biotic CO_2 uptake required to account for the observations. Oceanic uptake of CO_2 is assumed to have no effect on O_2 concentration[9], while land biotic uptake is assumed to occur with O_2/CO_2 ratios of −1.1:1 (ref. 33). The error bars indicate the estimated standard errors of the globally and annually averaged values. In computing global oceanic and land biotic uptake, we additionally allow for uncertainty of ±6 per meg and ±0.1 p.p.m. in the overall trends in O_2/N_2 and CO_2, respectively, due to uncertainty in long-term calibration.

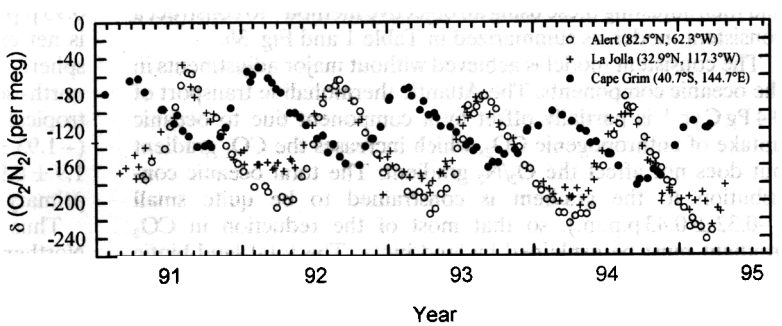

FIG. 1 $\delta(O_2/N_2)$ of air samples collected at Alert, La Jolla and Cape Grim and measured using the interferometric method[34]. Data from additional sites in the Scripps O_2/N_2 network (not shown) indicate that the interannual trends and north–south gradients inferred from these three sites are characteristic of large-scale atmospheric patterns.

mixing. The main sources and sinks believed to contribute to the gradients are fossil-fuel burning (which occurs mostly in the Northern Hemisphere), the natural pumping of CO_2 and O_2 from the northern to the southern hemisphere by the thermohaline circulation of the Atlantic Ocean, the exchange of CO_2 and O_2 with land biota, and the uptake of excess anthropogenic CO_2 by the oceans[2,3,13–15].

A plot of $\delta(O_2/N_2)_{GRAD}$ versus $(CO_2)_{GRAD}$ for data from 1991 to 1994 is shown in Fig. 3*a*. The data show clear interannual variations, with both gradients being largest (in absolute value) in 1991 and early 1992 and smallest in early 1993. The average CO_2 gradient is close to the 3 p.p.m. gradient observed during the middle 1980s. As seen in Fig. 3*a*, the time-averaged gradients, $\delta(O_2/N_2)_{GRAD}$ and $(CO_2)_{GRAD}$, are both smaller in absolute value than predicted from fossil-fuel burning alone using an atmospheric transport model[16]. In seeking an explanation for the reduction of both gradients, we note that CO_2 uptake by land biota in the Northern Hemisphere decreases both gradients, whereas transport by the North Atlantic thermohaline circulation decreases the CO_2 gradient while increasing the O_2/N_2 gradient. The Atlantic Ocean pumps O_2 (relative to N_2) in the same direction (north to south) as CO_2 because the flux of both gases is primarily caused by temperature-dependent solubility differences between northward- and southward-flowing waters[15,17]. The relative oceanic transport of O_2 and CO_2 can be estimated from ocean tracer distributions[15].

TABLE 1 Components of O_2/N_2 and CO_2 north–south gradients

Parameter	(Units)	Symbol	*a priori* Value	*a priori* σ	*a posteriori* Value	*a posteriori* σ
Fossil-fuel CO_2 grad.*	(p.p.m.)	C_f	4.91	0.69	4.90	0.68
Ocean CO_2 uptake grad.†	(p.p.m.)	C_o	0.42	0.42	0.42	0.41
Land biotic CO_2 grad.‡	(p.p.m.)	C_t	−1.15	5.0	−1.31	0.87
North Atlantic CO_2 grad.§	(p.p.m.)	C_{na}	−0.78	0.35	−0.78	0.34
North Atlantic O_2/N_2 grad.§	(per meg)	O_{na}	−4.0	1.9	−4.0	1.9
North Atlantic multiplier§		β_{na}	1.0	1.0	0.96	0.49
Fossil-fuel O_2/CO_2 combust. ratio‖		α_f	1.38	0.04	1.38	0.04
Land biota O_2/CO_2 exch. ratio¶		α_t	1.10	0.05	1.10	0.05
Observed CO_2 grad.#	(p.p.m.)	C_{obs}	3.27	0.26	3.27	0.26
Observed O_2/N_2 grad.#	(per meg)	O_{obs}	−29.2	2.4	−29.2	2.3
Combined results:						
North Atlantic CO_2 gradient	(p.p.m.)	$\beta_{na}C_{na}$			−0.75	0.41
Total ocean CO_2 gradient	(p.p.m.)	$\beta_{na}C_{na} + C_o$			−0.32	0.43

The '*a priori*' column contains estimates based on prior information, as described in the footnotes below. The '*a posteriori*' column contains the results of the objective optimization consistent with the governing equations which are: $C_{obs} = C_f + C_o + C_t + \beta_{na}C_{na}$ and $O_{obs} = -(\alpha_f C_f + \alpha_t C_t)/X + \beta_{na}O_{na}$ where $X = 0.2095$ is the O_2 mole fraction of dry air.

* The gradient induced by fossil-fuel burning in 1992. The gradients were computed using the TM2 atmospheric transport model[16] with fossil-fuel production (6.1 Pg C yr^{-1}) and source distribution data from refs. 26,27. We allow for 10% uncertainty (1σ) in fossil-fuel production and distribution, and 10% uncertainty in transport, which we sum in quadrature to yield a total uncertainty of 14%. The fossil-fuel gradients were corrected for fossil-fuel emissions of CO and CH_4 and corrected for photochemical oxidation of CO and CH_4 (whether from fossil fuels or other sources) using a photochemistry model[28].

† The gradient induced by the uptake of anthropogenic CO_2 by the oceans, estimated using a model estimate[29] of the CO_2 uptake pattern scaled to 1.7 Pg C yr^{-1} and using the TM2 model. We allow an uncertainty of 100%.

‡ The CO_2 gradient induced by exchanges with land biota. This component includes the contributions from net exchanges plus the rectification of seasonal exchanges. We use essentially no *a priori* constraint here.

§ The gradients induced by pre-industrial transport of O_2 and CO_2 from the North Atlantic into the Southern Hemisphere by the Atlantic's thermohaline circulation. The *a priori* estimates use transports of $(0.36 \pm 0.17) \times 10^{14}$ mol O_2 and $(0.33 \pm 0.15) \times 10^{14}$ mol CO_2, as deduced from water-mass differences in O_2, total carbon, nitrate-corrected alkalinity, phosphate and salinity[15]. The O_2 flux here is corrected for the compensating flux of N_2, where the N_2 flux was assumed proportional to the heat flux (see also ref. 14). The *a priori* CO_2 transport adopted here is consistent with other recent estimates although uncertainties are large[17,30,31]. We include an arbitrary scaling factor β_{na} to allow for additional uncertainty in the overall strength of the thermohaline circulation[15]. The CO_2 and O_2 fluxes were distributed according to the source/sink pattern for the 'North Atlantic sink' defined in ref. 20, and used as input to the TM2 model.

‖ Based on fossil-fuel O_2 consumption factors for different fuel types[32]. We allocate O_2 consumption by country, allowing for differences in the mixture of fuels used by different countries. The global O_2/CO_2 ratio for fossil-fuel burning (plus cement manufacturing) for 1992 is 1.39 assuming complete combustion. The factor of 1.38 used here also allows for differences in the spatial pattern of O_2 consumption and CO_2 production as this affects ratios of the signals predicted at the three sampling sites using the TM2 model, it allows for incomplete combustion of fossil-fuel, and it allows for atmospheric photochemical oxidation of CH_4 and CO, as per the first footnote to this table.

¶ From ref. 33.

\# Computed by taking the mean and standard error of the gradients for the four calendar years 1991–94.

We find from model simulations similar to those in Table 1 that the gradients in O_2/N_2 ratio and CO_2 concentration can neither be simultaneously explained by combining fossil-fuel burning and land biotic exchanges alone, nor can they be explained by additionally including an Atlantic thermohaline component, if the latter is based on north–south transport as large as 0.6 Pg C yr^{-1} (ref. 13) or larger[2]. A consistent explanation of both gradients is possible, however. We illustrate this by starting with *a priori* estimates of the oceanic and fossil-fuel contributions to the north–south gradients plus an arbitrary land biotic contribution, and then adjusting these using an objective method[18] to construct a consistent model, as summarized in Table 1 and Fig. 3*b*.

The consistent model is achieved without major adjustments in the oceanic components. The Atlantic thermohaline transport of 0.4 Pg C yr^{-1} is partially offset by a component due to oceanic uptake of anthropogenic CO_2, which increases the CO_2 gradient but does not affect the O_2/N_2 gradient. The total oceanic contribution to the gradient is constrained to be quite small (-0.32 ± 0.43 p.p.m.), so that most of the reduction in CO_2 gradient must be explained by land biota. The total land-biotic component (-1.31 ± 0.87 p.p.m.) has a relatively large uncertainty, however, because it has an O_2/CO_2 signature similar to the much larger fossil-fuel component, and is thus affected by small percentage errors in the estimated production and atmospheric transport of fossil-fuel CO_2.

Land biota can influence the CO_2 and O_2/N_2 gradients both by net annual exchanges and also by purely seasonal exchanges that are rectified by seasonal variations in atmospheric mixing[2,19]. Taking an estimate of $+0.62 \pm 0.33$ p.p.m. for the gradient due to rectification based on the range of two model predictions[2,19] for the sites under consideration, we calculate, by difference, a residual component due to net exchanges (-1.93 ± 0.93 p.p.m.). The only plausible explanation for such a gradient is net exchange of CO_2 with land biota in the Northern Hemisphere, because tropical exchanges have minimal effect on the north–south gradient, and because there is relatively little extratropical land biota in the Southern Hemisphere. The gradient (-1.93 ± 0.93 p.p.m.) requires a northern sink of about 1.9 ± 0.9 Pg C yr^{-1}, assuming that the sink is proportional to net primary production on land between 39° and 63° N (ref. 20).

Thus we find that the net CO_2 uptake by land biota in the Northern Hemisphere which is needed by our analysis to explain the north–south gradients is similar in magnitude to the net uptake required at the global scale by the interannual trends in

O_2/N_2 ratio and CO_2 concentration. One implication is that tropical ecosystems were not a strong net source or sink for CO_2 over the 1991–94 period. If so, then any releases of CO_2 from tropical deforestation must have been offset by CO_2 uptake occurring elsewhere in the tropics, as indicated by measurements during 1992/93 in Amazonia[21].

Our results are sensitive to the transport model due, in part, to the sparseness of the sampling network. Errors arising from such uncertainties in the fossil-fuel and rectifier terms have been explicitly considered. Our analysis does not, however, consider all possible sources of error. The estimated northern land biotic sink assumes that the sink is widespread over middle latitudes; a sink of unknown distribution would have larger uncertainty. Also, although our data indicate the need for an oceanic contribution to the combined O_2/N_2 and CO_2 gradients, our attribution of this component to the Atlantic thermohaline circulation remains speculative. More work is needed to characterize better the patterns of O_2 and CO_2 exchange in the Atlantic and other oceans and to account for river fluxes.

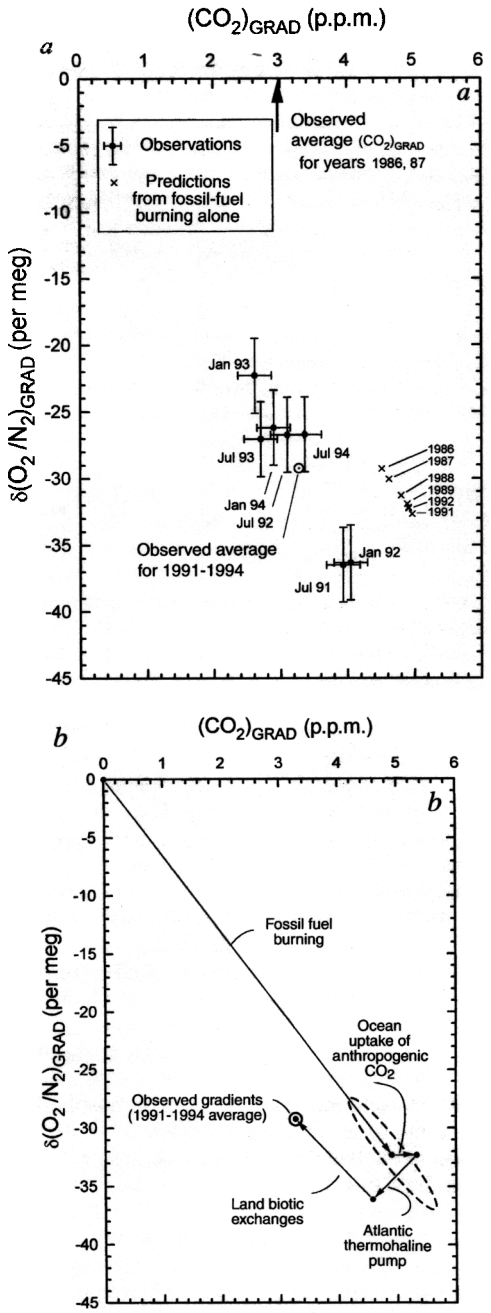

FIG. 3 *a*, Observed gradient components for 1991–94 plus modelled gradients from fossil-fuel burning (corrected for CO and CH_4 oxidation, see Table 1) plus cement manufacturing for years 1986 to 1992. Also shown are the average gradient $(CO_2)_{GRAD}$ for the years 1986 and 1987, computed using data from Cape Grim, Alert and Kumukahi[3] after correcting for an average offset of 0.815 p.p.m. observed between Kumukahi and La Jolla[6]. *b*, The *a posteriori* components of the average $\delta(O_2/N_2)$ and CO_2 gradients as resolved by the objective optimization (Table 1). The ellipsoid shows the range allowed by uncertainty in fossil-fuel burning and transport only.

Our analysis neglects possible gradients induced by the rectification of purely seasonal oceanic exchanges. Preliminary results from the Hamburg model of the ocean carbon cycle[22] suggest the effect on $\delta(O_2/N_2)_{GRAD}$ is smaller than 1 per meg. An ocean rectifier effect of at least 4 per meg would be needed to significantly affect our results.

Another possible source of error is interannual variations in air–sea exchange of O_2 driven by imbalances in marine photosynthesis and ventilation rates. Indeed, we see evidence for such exchanges in our data. The ~ 14 per meg reduction in the O_2/N_2 gradient between January 1992 and January 1993 (Fig. 3*a*) was ~ 7 per meg too large relative to the 1.4 p.p.m. shift in CO_2 gradient if both shifts were caused solely by exchanges with land biota. We speculate that the ~ 7 per meg excess shift in O_2/N_2 gradient was produced by anomalous exchanges of O_2 with the Southern Ocean. Interestingly, the steepest O_2/N_2 gradient and the largest seasonal O_2/N_2 increase at Cape Grim (see Fig. 1 and ref. 10) both occurred in 1991/92. Both features could be explained by an anomalous pulse of $\sim 1 \times 10^{14}$ mol O_2 released from the Southern Ocean around January 1992. Allowing for an uncompensated oceanic O_2 pulse of this magnitude, our estimated global land biotic uptake decreases from 2.0 to 1.7 Pg C yr^{-1} and our estimated northern land biotic uptake increases from 1.9 to 2.05 Pg C yr^{-1}. This suggests that errors in our previous estimates due to such an event are relatively small. Nevertheless, the O_2/N_2 and CO_2 data alone are not sufficient in general to quantify interannual imbalances in O_2 or CO_2 driven by ocean biota[10,23]. Our analysis assumes these exchanges are small; other data sets are needed to quantify such exchanges.

The growth rate of atmospheric CO_2 was unusually slow during the 1991–94 period compared to the previous decade, and particularly compared to the period of high growth rate in 1987 and 1988[11,12]. The slowdown has been ascribed (based on $^{13}C/^{12}C$ data[12,24]) to increasing CO_2 uptake by land biota, an interpretation which is compatible with our analysis for the 1991–94 period only if the northern land biota were a weaker sink or tropical land biota were a net CO_2 source in the 1980s. Our global oceanic sink is consistent with model estimates of long-term oceanic uptake that require net land biotic uptake averaging 0–1 Pg C yr^{-1} to balance the carbon budget over the past 30 years (ref. 25).

Our results are in rough agreement with two recent studies, one by Bender *et al.*[10], who found a global land biotic sink of 3.3 ± 1.6 Pg C yr^{-1} for the 1992–93 period based on independent O_2/N_2 records from 40° S and 41° S, and the other by Ciais *et al.*[7] who found a land biotic sink from 30° to 60° N of ~3.5 Pg C yr^{-1} in the 1992–93 period based on the north–south gradient in CO_2 and $^{13}C/^{12}C$ isotopes. The Bender *et al.* records agree well with the Cape Grim record (Fig. 1) in the critical overlapping time periods, thus supporting our global flux estimates which are based on longer records from both hemispheres. As the Ciais *et al.* study was based on a more comprehensive observational dataset but from a slightly different time period and used a different transport model, more analysis is needed to assess if the O_2/N_2 and $^{13}C/^{12}C$ data are consistent with the same source/sink distributions. The O_2/N_2 data complement the stable-isotope data by providing additional constraints, without complications from isotope dilution effects, on the partitioning of the global CO_2 sink between the land and the oceans and among large-scale regional sources and sinks. □

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