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## Two decades of ocean CO<sub>2</sub> sink and variability

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#### ABSTRACT

Atmospheric CO<sub>2</sub> has increased at a nearly identical average rate of 3.3 and 3.2 Pg C yr<sup>-1</sup> for the decades of the 1980s and the 1990s, in spite of a large increase in fossil fuel emissions from 5.4 to 6.3 Pg C yr<sup>-1</sup>. Thus, the sum of the ocean and land CO<sub>2</sub> sinks was 1 Pg C yr<sup>-1</sup> larger in the 1990s than in to the 1980s. Here we quantify the ocean and land sinks for these two decades using recent atmospheric inversions and ocean models. The ocean and land sinks are estimated to be, respectively, 0.3 (0.1 to 0.6) and 0.7 (0.4 to 0.9) Pg C yr<sup>-1</sup> larger in the 1990s than in the 1980s. When variability less than 5 yr is removed, all estimates show a global oceanic sink more or less steadily increasing with time, and a large anomaly in the land sink during 1990–1994. For year-to-year variability, all estimates show 1/3 to 1/2 less variability in the ocean than on land, but the amplitude and phase of the oceanic variability remain poorly determined. A mean oceanic sink of 1.9 Pg C yr<sup>-1</sup> for the 1990s based on O<sub>2</sub> observations corrected for ocean outgassing is supported by these estimates, but an uncertainty on the mean value of the order of ±0.7 Pg C yr<sup>-1</sup> remains. The difference between the two decades appears to be more robust than the absolute value of either of the two decades.

#### 1. Introduction

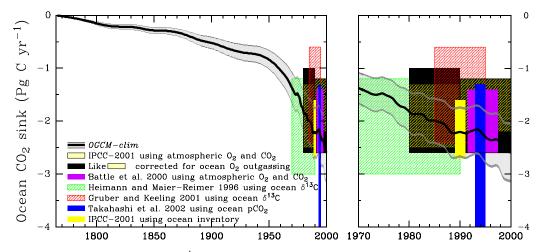
In the last 10 yr, observationally based estimates of the oceanic  $CO_2$  sink have been possible using oceanic or atmospheric measurements. According to recent estimates, the oceanic  $CO_2$  sink ranged between 1.5 and 2.8 Pg C yr<sup>-1</sup> for the past two decades (Fig. 1; Prentice et al., 2001), which is supported by box-diffusion models (Broecker et al., 1979; Oeschger et al., 1975) and by general circulation models (Orr et al., 2001; Dutay et al., 2002). Constraining the oceanic  $CO_2$  sink allows us to close the global carbon budget and estimate the net  $CO_2$  land sink, which comprises the sum of land use change including deforestation plus the residual terrestrial sink. We now go one step further and examine the temporal variability of the ocean and land  $CO_2$  sinks during the past two decades by comparing three recent estimates based on atmospheric inversions and one based on an ocean model.

In this paper we use the term "sink" to define a flux from the atmosphere to the ocean or land, and "source" to define a flux from the ocean or land to the atmosphere. We only discuss global sources and sinks. A global sink (source) does not imply that the flux is everywhere going from the atmosphere to the ocean or land (or the reverse), but only that the flux integrated over the globe is in that direction.

#### 2. Description of methods

We compare one atmospheric inversion using  $CO_2$ measurements only, two atmospheric inversions both

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*Fig. 1.* Global oceanic CO<sub>2</sub> sink (Pg C yr<sup>-1</sup>) computed using different methods. The central ocean model estimate was forced with atmospheric CO<sub>2</sub> concentration from ice cores before 1970, and from direct measurements after 1970. A range of  $\pm 20\%$  encompassing recent model results is also shown (Orr et al., 1999). The IPCC-2001 estimate (Prentice et al., 2001) is based on data from Langenfelds et al. (1999) for the 1980s and Manning (2001) for the 1990s, and includes a small correction for the solubility effect of ocean O<sub>2</sub>. The additional correction for ocean O<sub>2</sub> outgassing includes circulation as well as solubility effects of ocean O<sub>2</sub> (Table 1). The estimate of Takahashi et al. (2002) was corrected by 0.6 Pg C yr<sup>-1</sup> to account for the natural transport of CO<sub>2</sub> from land to the ocean by rivers. The IPCC-2001 estimate using ocean inventories assumes that the ratio of CO<sub>2</sub> growth rate to inventory is the same in the atmosphere and in the ocean, and uses global CO<sub>2</sub> inventory in 1990 (Gruber, 1998; Sabine et al., 1999; Sabine et al., 2002).

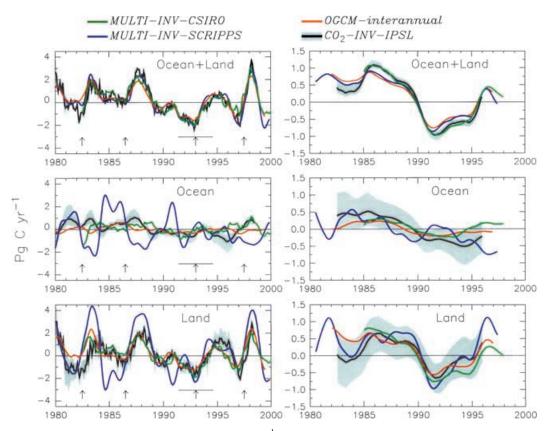
using  $\delta^{13}$ C and CO<sub>2</sub> measurements, and one ocean model forced with re-analyzed data.

The CO<sub>2</sub>-only inversion approach (hereafter referred to as CO2-INV-IPSL) is based on a threedimensional tracer inversion using the transport models TM2 and TM3 driven by observed winds for year 1990 (Bousquet et al., 2000). CO2 observations alone are used to determine both the global and regional land and ocean flux patterns and their temporal variability. The atmospheric CO<sub>2</sub> concentrations were provided by GLOBALVIEW (Masarie and Tans, 1995) CO<sub>2</sub>, including interpolated data but excluding extrapolated data. The shaded area in Fig. 2 represents the range covered by eight different sensitivity tests described in Bousquet et al. (2000). CO<sub>2</sub> transported from land to oceans by rivers is not directly reflected in atmospheric observations and therefore is not accounted for in CO2only inversions. An adjustment was made to the global sinks: 0.6 Pg C yr<sup>-1</sup> was subtracted from land uptake and added to oceanic uptake (Table 1; Sarmiento and Sundquist, 1992). This correction is constant in time.

One multi-tracer inversion (hereafter referred to as *MULTI-INV-CSIRO*) is based on a three-dimensional tracer inversion, where  $\delta^{13}$ C observations at Cape

Grim are used to separate the global land and ocean interannual variability, O2/N2 measurements also from Cape Grim to separate the mean global land and ocean fluxes for both decades together, and CO2 measurements from 12 stations to determine the regional distribution of the land and ocean fluxes. This inversion is an update of Rayner et al. (1999). The differences from that work can be summarized as follows: (1) the  $\delta^{13}$ C record has been revised using a new calibration scheme, (2) the input CO<sub>2</sub>,  $\delta^{13}$ C and O<sub>2</sub>/N<sub>2</sub> records have been extended forward in time, (3) the atmospheric response functions have been re-calculated with a different transport model, MATCH/MACCM2 (Gurney et al., 2002) and (4) a term has been added to the  $\delta^{13}$ C budget to treat the dilution of  $\delta^{13}$ C anomalies by the gross fluxes. Differences in the global anomaly fluxes between this study and Rayner et al. (1999) arise mainly from items (1) and (4), while item (3) affects regional flux anomalies.

Another multi-tracer inversion (hereafter referred to as *MULTI-INV-SCRIPPS*) is based on a double deconvolution of atmospheric CO<sub>2</sub> and  $\delta^{13}$ C data from nine stations, mainly in the Pacific Ocean basin (C. D. Keeling et al., 2001). The station data are smoothed with a spline technique and the splines are averaged



*Fig.* 2. Variability in ocean and land  $CO_2$  fluxes in Pg C yr<sup>-1</sup>. Positive values indicate a  $CO_2$  flux anomaly from the ocean or land to the atmosphere, negative values a  $CO_2$  flux anomaly from the atmosphere to the ocean or land. The curves are smoothed to remove variability less than 1 yr (left panels) and less than 5 yr (right panels). The mean of 1985–1995 is removed from each curve. Arrows indicate El Niño events. The different methods are described in the text.

globally. This inversion is an update of C. D. Keeling et al. (1995) using more  $\delta^{13}$ C stations and taking into account the proportion of C3 and C4 plants and their different isotopic fractionation. The proportion is fixed in time. The regional distribution of the fluxes is estimated using the transport model TM2 driven by observed winds from ECMWF for 1986 (Piper et al., 2001).

The ocean model estimate (hereafter referred to as *OGCM-interannual*) is based on an ocean general circulation model including biogeochemistry. The ocean general circulation model (OPA) has a mean resolution of  $1.5 \times 2^{\circ}$  increasing to  $0.5^{\circ}$  of latitude at the equator, and computes vertical turbulence explicitly. The model was forced by a combination of daily to weekly wind stresses and fluxes from the ERS satellite and NCEP and ECMWF re-analyzed data from 1979 to 1999 (Le Quéré et al., 2000). The biogeochem-

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istry model was updated to include plankton dynamics (Aumont et al., 2002) and initialized with carbon and nutrient observations (Levitus et al., 1993; Goyet et al., 2000).

To investigate the difference in oceanic  $CO_2$  sink between the 1980s and 1990s, we have done additional model simulations using the same ocean general circulation model, OPA, used in the *OGCM-interannual*, but forced with climatological wind stresses and fluxes and with observed atmospheric  $CO_2$  over both complete decades (hereafter referred to as *OGCM-clim*). These simulations reproduce the trend in the oceanic  $CO_2$  sink caused by increasing atmospheric  $CO_2$  at the observed rate. They do not reproduce the natural variability or trends which would be a consequence of climate change. Biological fluxes were not included in these simulations. A one-dimensional model (Kheshgi et al., 1999) is also used to separate the effect of

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	Ocean			Land		
	1980s	1990s	Difference	1980s	1990s	Difference
Based on atmospheric $O_2$ and $CO_2$						
IPCC 2001 <sup>a</sup>	$1.9 \pm 0.6$	$1.7 \pm 0.5$	-0.2	$0.2 \pm 0.7$	$1.4 \pm 0.7$	1.2
IPCC 2001 with O <sub>2</sub> correction <sup>b</sup>	$1.8\pm0.8$	$1.9\pm0.7$	0.1	$0.3\pm0.9$	$1.2\pm0.9$	0.9
Based on atmospheric inversions						
MULTI-INV-CSIRO <sup>c</sup>	1.6	1.8	0.1	0.6	1.5	0.9
MULTI-INV-SCRIPPS <sup>d</sup>	2.0	2.4	0.4	0.2	0.8	0.6
$CO_2$ -INV-IPSL $^{d-f}$	$1.7\pm0.7$	$2.4\pm0.6$	0.6	$0.4\pm0.9$	$0.8\pm0.9$	0.4
Based on ocean models <sup>g</sup>						
OGCM-interannual <sup>d,f</sup>	1.4	1.7	0.3	0.7	1.4	0.7
OGCM-clim <sup>h</sup>	2.0	2.3	0.3	0.1	0.8	0.7
1-D model with constant SST <sup>d</sup>	1.9	2.2	0.3	0.2	0.9	0.7
1-D model with observed SST <sup>d</sup>	1.7	1.9	0.2	0.4	1.2	0.8

Table 1. Ocean and land anthropogenic  $CO_2$  sinks (Pg C yr<sup>-1</sup>) for the past two decades

<sup>a</sup>Prentice et al. (2001) using atmospheric  $O_2/N_2$  and  $CO_2$  and including a correction for the solubility effect of ocean  $O_2$ . <sup>b</sup>Like (a) but including a further correction for the circulation effect of ocean  $O_2$  for both complete decades. Our correction is based on a  $O_2$ /heat flux ratio of 6.1 nmol J<sup>-1</sup>, a N<sub>2</sub>/heat flux ratio of 2.2 nmol J<sup>-1</sup> and an oceanic heating rate of -0.13  $10^{22}$  J yr<sup>-1</sup> in the 1980s and 0.55  $10^{22}$  J yr<sup>-1</sup> in the 1990s. The heating rate was estimated using the observed heat content of Levitus et al. (2000) until 1996, and an estimate of ocean heat content based on sea surface height for 2000 (Cabanes et al., 2001). For comparison, Bopp et al. (2002) used an  $O_2$ /heat flux ratio of 6.1 nmol J<sup>-1</sup> with a heating rate of 1.0 for the 1990–1996 period solely based on Levitus et al. (2000), whereas Keeling and Garcia (2002) used a  $O_2$ /heat flux ratio of 4.9 nmol J<sup>-1</sup> based on observations with a heating rate of 0.6  $10^{22}$  J yr<sup>-1</sup> based on model results of anthropogenic heat uptake.

<sup>c</sup>Results available from 1982.

<sup>d</sup>In these studies, the trend and variability is more reliable than the absolute mean sinks. In the *MULTI-INV-SCRIPPS*, the mean sinks result from initial assumptions. In the *CO<sub>2</sub>-INV-IPSL*, the mean sinks result from horizontal CO<sub>2</sub> gradients which are smaller than the temporal CO<sub>2</sub> variability. In the *OGCM-interannual*, the mean oceanic sink results from the initialization of the model with observations in 1979. The one-dimensional model is tuned to produce an oceanic sink of 1.9 in the 1980s when forced with constant SST.

<sup>e</sup>0.6 Pg C yr<sup>-1</sup> was added to the oceanic uptake and subtracted from land uptake to correct for the carbon transported from land to ocean by rivers (Sarmiento and Sundquist, 1992), which is not detected by inversions based on  $CO_2$  only. <sup>f</sup>Results available until 1999.

<sup>g</sup>Estimate from ocean models plus the atmospheric growth rate are subtracted from the emissions to give a land estimate. <sup>h</sup>This model estimate lies in the middle of the OCMIP model estimates. It does not resolve climate variability.

variations and trends in atmospheric  $CO_2$  and average sea surface temperature.

The monthly fluxes of the *MULTI-INV-SCRIPPS* were directly provided without the seasonal cycle. For the other estimates, the interannual fluxes were computed by applying a 12-month equal-weight running average. The 5-yr smoothing of Fig. 2 was estimated by applying a 60-month running average on all estimates.

#### 3. Variability

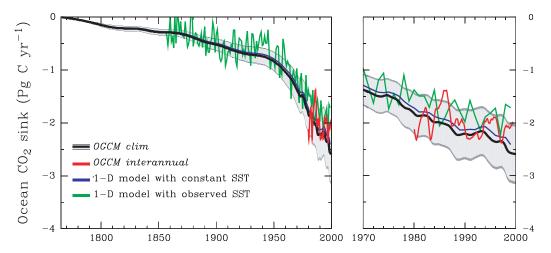
The sum of the ocean and land  $CO_2$  sinks is well constrained by atmospheric observations and fossil fuel emissions (top panels of Fig. 2). A smaller total sink (i.e. a larger atmospheric  $CO_2$  increase) by ~2 Pg C yr<sup>-1</sup> was observed following the El Niño events of 1982–1983, 1986–1987 and 1997–1998, with the 1997–1998 El Niño showing sharper features than the previous ones. When variability less than 5 yr is filtered out, a larger total sink appears from 1990 to 1995, and disappears in the later years. The average total sink was 1 Pg C yr<sup>-1</sup> larger in the 1990s than in the 1980s.

The interannual variability of the global oceanic  $CO_2$  sink between the tracer inversions shows large discrepancies both in amplitude and in phase (Fig. 2). The discrepancies in amplitude become smaller after 1991/1992, which may be due to new calibration

techniques introduced in the two  $\delta^{13}$ C observational programs at that time. However, a longer comparison covering a few El Niño events after 1991/92 need to be done before it can be concluded that the different methods converge. Comparing the atmospheric inversions with the ocean model also shows poor correspondence for the global ocean. These estimates only agree in allowing more variability for the land  $(\pm 1.6 \text{ to } \pm 3.7 \text{ Pg C yr}^{-1} \text{ for peak-to-peak smoothed})$ monthly values) than for the ocean ( $\pm 0.7$  to  $\pm 2.8$  Pg C yr<sup>-1</sup>). At the regional level, atmospheric inversions indicate at least as much variability in the northern hemisphere oceans (poleward of 20°) than in the south, opposite to the results of the ocean model (Le Quéré et al. 2000, not shown). Convergence of the estimates of variability by atmospheric inversions and the ocean model is seen in the Southern ocean in the 1990s only, where all estimates give a variability of  $\pm 0.2$  to  $\pm 0.5$  Pg C yr<sup>-1</sup>, roughly in phase with each other.

When the global estimates are compared after removing variability less than 5 yr, the agreement for the global variability is much better (right panels of Fig. 2, note the change in scale). On this time scale, all estimates show an increase in the oceanic sink by  $\sim 0.3$  Pg C yr<sup>-1</sup> per decade and attribute the residual variability in atmospheric CO<sub>2</sub> to the land. The land shows variability of  $\pm 0.6$  to  $\pm 1$  Pg C yr<sup>-1</sup> (for peakto-peak smoothed monthly values) with a smaller sink during 1986–1989 and a larger sink during 1990–1994. Whereas a trend is apparent in the oceanic sink, oscillating variability dominates the land sink. The difference between the oceanic sink for the 1990s and the 1980s ranges between 0.1 and 0.6 Pg C yr<sup>-1</sup>, with a mean value of 0.3 Pg C yr<sup>-1</sup> (Table 1). The corresponding 1990s–1980s difference for the land  $CO_2$  sink ranges between 0.4 and 0.9 Pg C yr<sup>-1</sup>, with a mean value of 0.7 Pg C yr<sup>-1</sup>.

Ocean models estimate a larger oceanic sink of 0.3 Pg C yr<sup>-1</sup> in the 1990s than in to the 1980s (Table 1). This larger sink in the 1990s is driven by the increase in atmospheric CO<sub>2</sub> between the two decades, and by the fact that oceanic CO<sub>2</sub> is not in equilibrium with the atmosphere and continues to take up CO<sub>2</sub> emitted during the previous decades. This trend of 0.3 Pg C yr<sup>-1</sup> per decade is determined by the relative time scales of the CO<sub>2</sub> increase in the atmosphere, versus that of ocean mixing. The trend is almost the same whether or not the ocean model includes a representation of natural variability and global warming (Fig. 3). The effect of warming alone, however, decreases the mean oceanic sink by 0.2 and 0.3 Pg C yr<sup>-1</sup> for the 1980s and the 1990s, respectively. However, the model considered here does not include the more complex interactions between ocean circulation, marine biology and the carbon cycle which may result from global warming (Sarmiento et al., 1998).



*Fig. 3.* Global oceanic CO<sub>2</sub> sink (Pg C yr<sup>-1</sup>) computed using different ocean models forced by observed atmospheric CO<sub>2</sub> measurements. The *OGCM-clim* is described in Fig. 1. The *OGCM-interannual* is based on the same model as *OGCM-clim* but it is forced with reanalysis and satellite fields of heat and water fluxes and wind stress. The one-dimensional model estimate is forced with constant sea surface temperature (blue) and with the observed global temperatures anomalies as estimated by Jones et al. (1999).

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#### 4. Mean sink and uncertainty

Thirteen ocean models participating in the Ocean Carbon-cycle Model Inter-comparison Project (OCMIP) recently estimated an oceanic sink ranging between 1.6 and 2.4 Pg C yr<sup>-1</sup> for the 1980s (Orr et al., 2001; Dutay et al., 2002; Prentice et al., 2001), in agreement with observational estimates (Fig. 1) and consistent with inventories of anthropogenic carbon in the ocean (Gruber, 1998; Sabine et al., 1999; 2002). This range of  $\pm 0.4$  Pg C yr<sup>-1</sup> represents the minimum uncertainty in ocean circulation and mixing because other carbon cycle parameters were the same for all models. A larger estimate of 3.9 Pg C yr<sup>-1</sup> was recently published using an ocean model with simplified ventilation (Thomas et al., 2001). However, Thomas et al. assume that the partial pressure difference between the ocean and the atmosphere remained the same in the past century, an assumption which is not valid at high latitudes (Takahashi et al., 2002) and leads to an overestimation of the oceanic sink. Other sources of uncertainty must be added which we roughly estimate as follows. Uncertainties in gas exchange can add  $\pm 0.1$  Pg C yr<sup>-1</sup> to the range of possible model results (Sarmiento et al., 1992). Uncertainties in the mean surface alkalinity add  $\pm 0.1$  Pg C yr<sup>-1</sup>. Uncertainties more difficult to quantify are associated with natural or anthropogenic changes in marine biology, carbon fluxes in the coastal area, river fluxes, eddies and extreme storm events. The estimated model uncertainty around the mean also varies over time. Large decadal variability in ocean temperature was observed since 1950 (Levitus et al., 2000) which translates to a variability in the oceanic  $CO_2$  sink of at least  $\pm 0.2$  Pg C  $yr^{-1}$  based on the coupled model results of Bopp et al. (2002). Furthermore, climate change may have already altered the ocean carbon cycle in ways that are difficult to quantify. Decadal variability and trends do not translate into uncertainties, but into biases of the mean value which can be estimated by assessing the sink for different time periods.

To further constrain estimates of the oceanic  $CO_2$ sink, atmospheric measurements appear promising because of the rapid mixing of the atmosphere. This rapid mixing allows us to deduce information on the global ocean based on relatively few atmospheric measurements. Atmospheric  $\delta^{13}C$  was first used to separate the ocean and land sinks, but the uncertainty in  $\delta^{13}C$  disequilibrium fluxes and in the measurement itself was found to be large (Tans et al., 1993). Recent CO<sub>2</sub>-only inversion approaches have also the potential to constrain the mean  $CO_2$  sinks, but at present the associated uncertainty is greater than  $\pm 1$  Pg C yr<sup>-1</sup> because of uncertainties in the atmospheric transport models (particularly associated with the rectifier effect) and in the station network (Gurney et al., 2002).

The ocean and land sinks can also be separated using atmospheric O<sub>2</sub>/N<sub>2</sub> and CO<sub>2</sub> observations (R. F. Keeling and Shertz, 1992). The largest uncertainty of this method concerns the long-term oceanic O<sub>2</sub> flux. As the ocean surface warms, oceanic O<sub>2</sub> is out-gassed to the atmosphere due to its reduced solubility, to changes in ocean circulation and mixing and to changes in marine production and respiration. This impact was recently assessed using empirical (R. F. Keeling and Garcia, 2002) or modeled (Bopp et al., 2002) flux relationships applied to observed heat fluxes (Table 1). Both methods give a total oceanic O<sub>2</sub> out-gassing roughly four times larger than the effect of the solubility alone. For any one decade since 1950, the maximum estimated correction which must be added to the oceanic CO<sub>2</sub> sink (and subtracted from the land CO<sub>2</sub> sink) estimated from atmospheric  $O_2/N_2$  is -0.2 to 0.6 Pg C  $yr^{-1}$ , with an additional uncertainty of  $\pm 0.5$  associated with oceanic  $O_2$  outgassing (Bopp et al., 2002). This correction increases the uncertainty in the decadal mean sink based on atmospheric  $O_2$  to  $\pm 0.7$  Pg C yr<sup>-1</sup> in the 1990s around a mean value of 1.9 Pg C yr<sup>-1</sup> (Table 1). When the impact of changes in ocean circulation on  $O_2$  outgassing is considered (Table 1), oceanic sink estimates based on atmospheric O<sub>2</sub> give a larger oceanic sink in the 1990s than in the 1980s, but with an uncertainty larger than the difference of the two decades.

#### 5. Discussion

Each approach presented in this paper has its limitation. The multi-tracer inversion approach using  $\delta^{13}$ C is very sensitive to data quality, and may be affected by variable isotopic discrimination associated with photosynthesis of terrestrial plants (Fung et al., 1997). The CO<sub>2</sub>-only inversion approach critically depends on the position of the stations to separate the ocean from the land. The ocean model approach tends to underestimate variability in ocean circulation, especially at high latitudes, and is subject to further uncertainties in parameters and climate feedbacks.

Nevertheless the comparison of these different estimates for the past two decades highlights three results where a consensus is reached. First, estimates of the

global mean oceanic CO2 sink have converged towards a value of  $\sim$ 2 with an uncertainty of around  $\pm$ 0.7 Pg C  $yr^{-1}$  for the past two decades (Table 1). This estimate is supported by oceanic and atmospheric observations, ocean models and ocean carbon inventories. Second, all models indicate an increasing oceanic CO2 sink of  $\sim 0.3$  Pg C yr<sup>-1</sup> per decade driven by the sustained atmospheric CO2 increase, and this is not contradicted by any of the observational-based approaches. Third, the interannual variability of the oceanic CO<sub>2</sub> sink is less than that of the land. Although atmospheric inversions and ocean models show quite different variability for ocean CO<sub>2</sub>, they all allocate smaller variability for the ocean than for the land, with the ratio of land/ocean variability increasing in the 1990s and for variability smoothed over 5 yr or longer.

The comparison of various estimates also highlights unresolved controversies. First the amplitude of the interannual variability in the oceanic CO<sub>2</sub> sink is not reliably determined: although all estimates examined here allocate less variability to the ocean than to the land, the amplitude of this variability ranges from  $\pm 0.7$ to  $\pm 2.8$  Pg C yr<sup>-1</sup> for peak-to-peak smoothed monthly values. Second, regional variability in the oceanic CO<sub>2</sub> flux is poorly known. For these two aspects, atmospheric tracer inversions differ amongst each other and from the ocean model, and we are not yet in a position to say which of these estimates is closer to reality. The differences between tracer inversions on interannual timescales may be due to differences in the early measurement methods of  $\delta^{13}$ C. The location and number of stations, particularly those with matching  $CO_2$  and  $\delta^{13}C$  records, and differences in prescribed spatial distribution of the sources and sinks may also be a factor. Further differences between tracer inversions approaches and ocean models can come from the possible underestimation of variations in ocean circulation, and from the compensation of variability by different processes in certain ocean regions (Le Quéré et al., 2000). Such differences can be resolved in the future by a more accurate quantification of oceanic CO<sub>2</sub> flux and variability at the regional level. Until regional  $CO_2$  flux patterns can be better constrained, it will be difficult to reduce the uncertainty in the mean ocean and land CO<sub>2</sub> sinks below the value of  $\pm 0.7$  Pg C yr<sup>-1</sup> estimated using atmospheric O<sub>2</sub>/N<sub>2</sub> measurements.

#### 6. Acknowledgments

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