

References and Notes

- C. Salomon et al., *Proceedings of the 17th International Conference on Atomic Physics (ICAP 2000)*, E. Arimondo, P. D. Natale, M. Inguscio, Eds. (American Institute of Physics, Melville, NY, 2001), pp. 23–40.
- J. M. Hensley, A. Wicht, B. C. Young, S. Chu, *Proceedings of the 17th International Conference on Atomic Physics (ICAP 2000)*, E. Arimondo, P. D. Natale, M. Inguscio, Eds. (American Institute of Physics, Melville, NY, 2001), pp. 43–57.
- C. Chin, V. Leiber, V. Vuletic, A. J. Kerman, S. Chu, *Phys. Rev. A* **63**, 033401 (2001).
- C. E. Wieman, *Proceedings of the 16th International Conference on Atomic Physics (ICAP 1998)*, W. E. Baylis, G. W. Drake, Eds. (American Institute of Physics, Woodbury, NY, 1999), pp. 1–13.
- M. J. Snadden, J. M. McGuirk, P. Bouyer, K. G. Haritos, M. A. Kasevich, *Phys. Rev. Lett.* **81**, 971 (1998).
- J. Söding, D. Guéry-Odelin, P. Desbiolles, G. Ferrari, J. Dalibard, *Phys. Rev. Lett.* **80**, 1869 (1998).
- D. Guéry-Odelin, J. Söding, P. Desbiolles, J. Dalibard, *Europhys. Lett.* **44**, 26 (1998).
- J. Arlt et al., *J. Phys. B* **31**, L321 (1998).
- H. Perrin, A. Kuhn, I. Bouchoule, C. Salomon, *Europhys. Lett.* **42**, 395 (1998).
- A. J. Kerman, V. Vuletić, C. Chin, S. Chu, *Phys. Rev. Lett.* **84**, 439 (2000).
- D.-J. Han et al., *Phys. Rev. Lett.* **85**, 724 (2000).
- D. Han, M. T. DePue, D. Weiss, *Phys. Rev. A* **63**, 023405 (2001).
- S. L. Cornish, S. Hopkins, A. M. Thomas, C. J. Foot, abstract from the 7th Workshop on Atom Optics and Interferometry, 28 September to 2 October 2002, Lunteren, Netherlands (book of abstracts).
- M. Anderson, J. Ensher, M. Matthews, C. Wieman, E. Cornell, *Science* **269**, 198 (1995).
- K. Davis et al., *Phys. Rev. Lett.* **75**, 3969 (1995).
- C. Bradley, C. Sackett, J. Tollett, R. Hulet, *Phys. Rev. Lett.* **75**, 1687 (1995).
- S. Cornish, N. Claussen, J. Roberts, E. Cornell, C. Wieman, *Phys. Rev. Lett.* **85**, 1795 (2000).
- G. Modugno et al., *Science* **294**, 1320 (2001); published online 18 October 2001 (10.1126/science.1066687).
- D. G. Fried et al., *Phys. Rev. Lett.* **81**, 3811 (1998).
- A. Robert et al., *Science* **292**, 461 (2001); published online 22 March 2001 (10.1126/science.1060622).
- C. Chin, V. Vuletic, A. J. Kerman, S. Chu, *Phys. Rev. Lett.* **85**, 2717 (2000).
- P. J. Leo, C. J. Williams, P. S. Julienne, *Phys. Rev. Lett.* **85**, 2721 (2000).
- L. Khaykovich et al., *Science* **296**, 1290 (2002).
- K. Strecker, G. Partridge, A. Truscott, R. Hulet, *Nature* **417**, 150 (2002).
- R. Grimm, M. Weidemüller, Yu. B. Ovchinnikov, *Adv. At. Mol. Opt. Phys.* **42**, 95 (2000).
- D. S. Weiss, personal communication.
- A. J. Kerman et al., *C. R. Acad. Sci. Paris IV* **2**, 633 (2001).
- P. S. Julienne, E. Tiesinga, C. J. Williams, personal communication.
- We observe the narrow Feshbach resonances at 11.0, 14.3, 15.0, 19.9, 47.7, and 53.4 G (accuracy ± 0.2 G) as loss resonances in three-body decay of an uncondensed gas.
- Materials and methods are available as supporting material on Science Online.
- M. Barrett, J. Sauer, M. Chapman, *Phys. Rev. Lett.* **87**, 010404 (2001).
- P. Treutlein, K. Y. Chung, S. Chu, *Phys. Rev. A* **63**, 051401 (2001).
- D. Stamper-Kurn et al., *Phys. Rev. Lett.* **81**, 2194 (1998).
- M. Hammes, D. Rychtarik, H.-C. Nägerl, R. Grimm, *Phys. Rev. A* **66**, 051401(R) (2002).
- E. A. Donley, N. R. Claussen, S. T. Thompson, C. E. Wieman, *Nature* **417**, 529 (2002).
- D. Jaksch, C. Bruder, J. Cirac, C. Gardiner, P. Zoller, *Phys. Rev. Lett.* **81**, 3108 (1998).
- M. Greiner, O. Mandel, T. Esslinger, T. W. Hänsch, I. Bloch, *Nature* **415**, 39 (2002).
- F. Masnou-Seeuws, P. Pillet, *Adv. At. Mol. Opt. Phys.* **47**, 53 (2001).
- S. Gupta, K. Dieckmann, Z. Hadzibabic, D. E. Pritchard, *Phys. Rev. Lett.* **89**, 140401 (2002).
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Figs. S1 to S3
References

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Anthropogenic CO₂ Uptake by the Ocean Based on the Global Chlorofluorocarbon Data Set

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We estimated the oceanic inventory of anthropogenic carbon dioxide (CO₂) from 1980 to 1999 using a technique based on the global chlorofluorocarbon data set. Our analysis suggests that the ocean stored 14.8 petagrams of anthropogenic carbon from mid-1980 to mid-1989 and 17.9 petagrams of carbon from mid-1990 to mid-1999, indicating an oceanwide net uptake of 1.6 and 2.0 \pm 0.4 petagrams of carbon per year, respectively. Our results provide an upper limit on the solubility-driven anthropogenic CO₂ flux into the ocean, and they suggest that most ocean general circulation models are overestimating oceanic anthropogenic CO₂ uptake over the past two decades.

Despite improvements in our understanding of the partitioning of anthropogenic CO₂ between the atmosphere, ocean, and terrestrial biosphere, substantial uncertainties and insufficient direct observational constraints continue. Recent decadal-scale changes in oxygen concentrations that have been observed in the ocean (1, 2) imply large and uncertain corrections (3–5) to the oceanic and terrestrial sinks for anthropogenic CO₂ that have been estimated based on atmospheric O₂/N₂ observations (6, 7), which was the method used in the 2001 report by the Intergovernmental Panel on Climate Change (8). Because the O₂/N₂ technique is based on atmospheric observations, it inherently requires assumptions regarding the partitioning of anthropogenic CO₂ between the ocean and terrestrial biosphere. Ocean general circulation models (OGCMs) currently simulate oceanic anthropogenic CO₂ uptake, assuming a steady-state circulation and biological production (9). We present here an observational estimate of the decadal inventory of anthropogenic CO₂ in the ocean based on the global chlorofluorocarbon (CFC) data set. Our estimates provide in-

dependent observational insights into the contemporary global carbon budget and provide a framework that can be used for direct validation of ocean model predictions.

The most direct way of estimating anthropogenic CO₂ accumulation in the ocean is to compare dissolved inorganic carbon (DIC) measurements made at one time with those made later in the same region. To isolate the long-term trend from changes due to natural variability, DIC measurements along isopycnal surfaces (10) can be compared, or multiple linear regression (MLR) of DIC against hydrographic properties (11) can be used (Fig. 1C legend). Although these methods provide direct evidence for regional anthropogenic CO₂ accumulation (10, 12, 13), they currently cannot be used in the global context because of the lack of adequate historical DIC measurements.

Another way to estimate anthropogenic CO₂ is to study the distribution of CFCs in the ocean. The release of CFC-11 (CCl₃F) and CFC-12 (CCl₂F₂) to the atmosphere began in the 1930s and accelerated in the 1950s. CFCs are entirely anthropogenic and biologically inert in the ocean. The oceanic CFC distribution thus provides valuable information about the rates and pathways of water mass ventilation processes (14). As part of the World Ocean Circulation Experiment (WOCE) carried out in the 1990s, dissolved CFCs were measured with great accuracy and unprecedented global resolution (Fig. 2). The patterns of oceanic accumulation

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REPORTS

of CFCs and anthropogenic CO₂ are quite different as a result of regional differences in carbonate chemistry, solubilities, and rates of air-sea gas exchange. Concentrations of CFCs (Fig. 1A) are highest in the cold, high-latitude surface ocean and decrease equatorward because of lower solubility at warmer temperatures. Although CO₂ solubility also increases with colder temperatures, the observed surface water accumulation of anthropogenic CO₂ shows a different pattern (Fig. 1C), with the lowest concentrations in the Southern Ocean and concentrations increasing northward. This pattern is mainly due to regional variations in sea surface alkalinity (15). Because of lower alkalinity in surface waters, the Southern Ocean has less capacity to accumulate anthropogenic CO₂ for a given increase in atmospheric CO₂ than oceans that are located farther north. Because of these regional differences, the direct use of CFC concentra-

tions to infer anthropogenic CO₂ accumulation in the ocean is problematic.

The method used here is based on using observed CFC concentrations to estimate water "ages" (defined as the amount of time since the parcel of water was last at the surface). These age-based methods have been applied on a regional scale (16, 17). However, they have yet to be applied and validated on a global scale. The conversion to water ages requires CFC, temperature, and salinity measurements, along with CFC solubility functions (18) and CFC atmospheric histories (19). The ages used in this study were calculated using the oceanic CFC-12 concentration and atmospheric observations (20). The conversion from CFC-12 concentration to CFC age produces a tracer that normalizes the variations in the CFC-12 distribution due to oceanic temperature and salinity distributions (Fig. 1B). Because of mixing, a water

parcel is composed of a distribution of transit times since it was last in contact with the atmosphere. The mean of these transit times (or ages) is sometimes referred to as the ideal age (21). Water ages estimated from CFC-12 (22, 23) have been shown to reflect the mean transit time (the ideal age) to within 25% for waters from 0 to 25 years old because of the quasilinear atmospheric history of CFC-12 from 1970 to 1995 (23, 24). For older waters, CFC ages significantly underestimate the ideal ages because of a nonlinear mixing bias resulting from the exponential increase of CFCs earlier than 1970 (23, 24). We focus on water with CFC ages younger than 30 years, based on the good agreement with independent direct anthropogenic CO₂ estimates in the Indian Ocean (Fig. 1) and Southern Ocean (25) and the results from our modeling study. The contribution of waters older than 30 years to the decadal anthropogenic CO₂ inventory has only a modest impact on our estimates, as discussed below. We now combine our estimates of water mass ages with the atmospheric CO₂ history (26) and carbonate chemistry equilibrium equations (27) to calculate the change in DIC in the ocean from one period (t_1) to another (t_2) by using the equation

$$C_{\text{anth}}(t_2) - C_{\text{anth}}(t_1) = \text{DIC}_{\text{eq}} [S, T, \text{ALK}_0, f\text{CO}_2(t_2 - \tau)] - \text{DIC}_{\text{eq}} [S, T, \text{ALK}_0, f\text{CO}_1(t_1 - \tau)]$$

where $C_{\text{anth}}(t_2) - C_{\text{anth}}(t_1)$ is the accumulation of anthropogenic CO₂ between year (t_1) and year (t_2), DIC_{eq} is the equilibrium concentration of seawater with atmospheric CO₂ concentration ($f\text{CO}_2$) when the water parcel was last at the surface (28), T is the temperature, S is the salinity, ALK_0 is the preformed alkalinity (29), and τ is the water parcel age in years (30).

From the above equation, we can calculate the change in DIC throughout the 1980s and 1990s. When a comparison is possible, the decadal accumulation of anthropogenic CO₂ that is estimated with the CFC age method (Fig. 1D) agrees well with direct observations that have been obtained using the MLR technique (Fig. 1C). Using all available measurements in the Indian Ocean, we use the CFC age method to estimate a basinwide accumulation of 6 Pg of C from 1978 to 1995, in agreement with the direct MLR-based estimate of 6.1 ± 1 Pg of C (12). The accumulation between 20°S and 5°N was 1.5 Pg of C, also in good agreement with the independent estimate from isopycnal analyses of 1.64 Pg of C (10).

Although the CFC age technique reproduces the observed decadal accumulation of anthropogenic CO₂, we must carefully evaluate the main assumptions in the CFC age technique: (i) that CFC ages give a reasonable estimate of ideal water mass ages, (ii) that CFCs and anthropogenic CO₂ maintain similar saturation states at the surface ocean, and (iii) that the air-sea CO₂

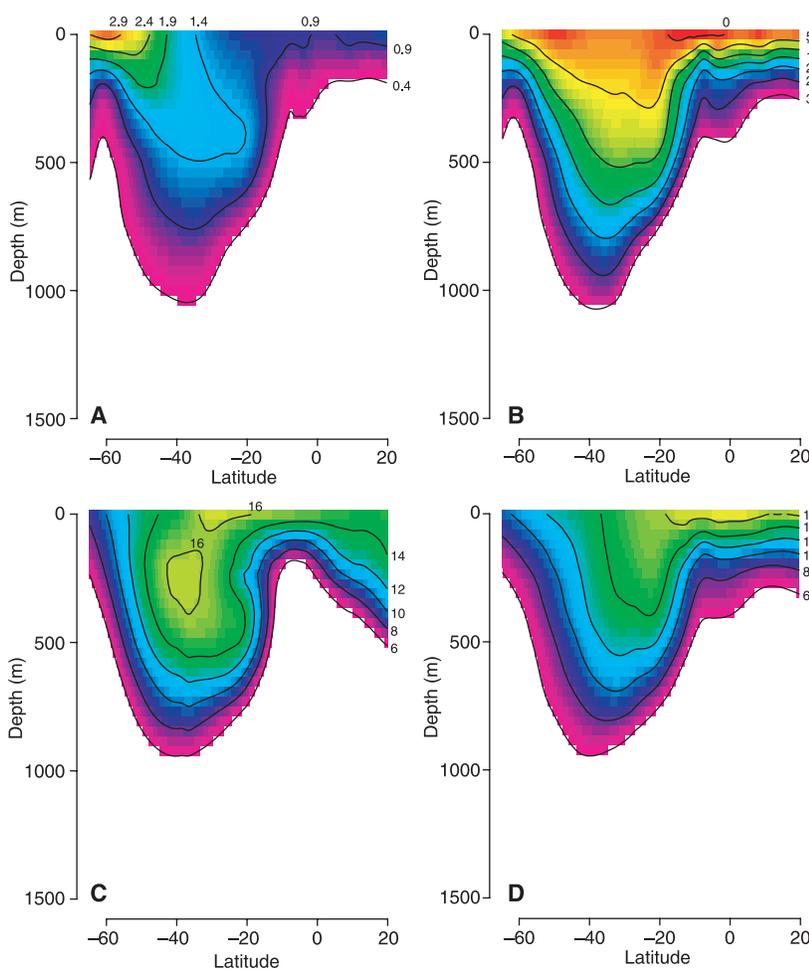


Fig. 1. (A) Zonal mean CFC-12 concentrations (in pmol/kg) for the Indian Ocean. (B) Estimated CFC-12 ages (years) for the Indian Ocean. (C) Zonal mean estimate of anthropogenic CO₂ (in $\mu\text{mol/kg}$) accumulation in the Indian Ocean from 1978 to 1995 made with the MLR method (12). The MLR method involves regressing DIC against Geochemical Ocean Sections hydrographic data from 1978 and applying the resulting regression to WOCE hydrographic data from 1995 to predict DIC concentrations that would have been present without anthropogenic CO₂ accumulation. The difference between the measured DIC during WOCE and the DIC predicted from the regression is the change in DIC due to anthropogenic CO₂ from 1978 to 1995, corrected for changes in DIC due to natural variability. (D) Zonal mean anthropogenic CO₂ accumulation (in $\mu\text{mol/kg}$) for the same period (1978 to 1995), estimated using the CFC age technique.

disequilibrium does not change over the time interval in question (31). To explore these combined uncertainties, we used results from a simulation with an OGCM forced with the observed atmospheric histories of CO₂ and CFCs (32), following the protocol of the Ocean Carbon Model Intercomparison Project (9). By comparing the simulated global anthropogenic CO₂ inventory from between 1980 and 1999 to those computed using the CFC age method in the model, we evaluated the potential biases in the CFC age method. We found that the CFC age method reproduces the simulated decadal anthropogenic CO₂ inventory to within 10% (Fig. 3A). The Southern Ocean (south of 40°S) is the region of greatest error, because the rapid exchange of surface waters with deep waters results in a violation of all three of the assumptions of the CFC age approach (Fig. 3B). However, the bias in the Southern Ocean may be overestimated in the model, given the good agreement between the observational CFC age estimates and the direct estimates south of 40°S (Fig. 1) (25). In any case, because the total anthropogenic CO₂ inventory in the Southern Ocean is small, the biases in this region have little effect on our global inventory estimates. An important factor in the relatively small bias in estimating the decadal anthropogenic CO₂ inventory using CFC ages is the relatively linear atmospheric histories between CFCs and CO₂ from 1970 to 1990 (19, 26). Any mixing effects that result in biases in the CFC age estimation for young waters (23, 24) will have a similar impact for anthropogenic CO₂ and will result in a relatively small bias when the CFC age method is used to infer decadal anthropogenic CO₂ in the ocean.

The decadal accumulation of anthropogenic CO₂ shows a similar pattern in each ocean (Fig. 4). The lowest values are found in the Southern Ocean (south of 40°S) and the equatorial regions (10°S to 10°N), associated with the upwelling of old water to the surface (12, 13). The largest inventories are found in the southern subtropical gyre (20° to 40°S) and in the North Atlantic, where anthropogenic CO₂ penetrates throughout the water column, consistent with penetration of North Atlantic Deep Water.

The global inventory (33) is estimated to be 14.8 Pg of C from mid-1980 to mid-1989 and 17.9 Pg of C from mid-1990 to mid-1999, with the Pacific (~46%) and Atlantic (~35%) contributing the most (Table 1). These estimates imply an average uptake rate of 1.6 ± 0.4 and 2.0 ± 0.4 Pg of C per year for the 1980s and 1990s (34).

The CFC age method assumes a steady-state ocean, whereas both models and recent observations (1, 2) suggest that the ocean may be changing as a result of global warming. CFCs were added to the model described above (1, 32) to evaluate climate change impacts on the CFC age-derived estimates of anthropogen-

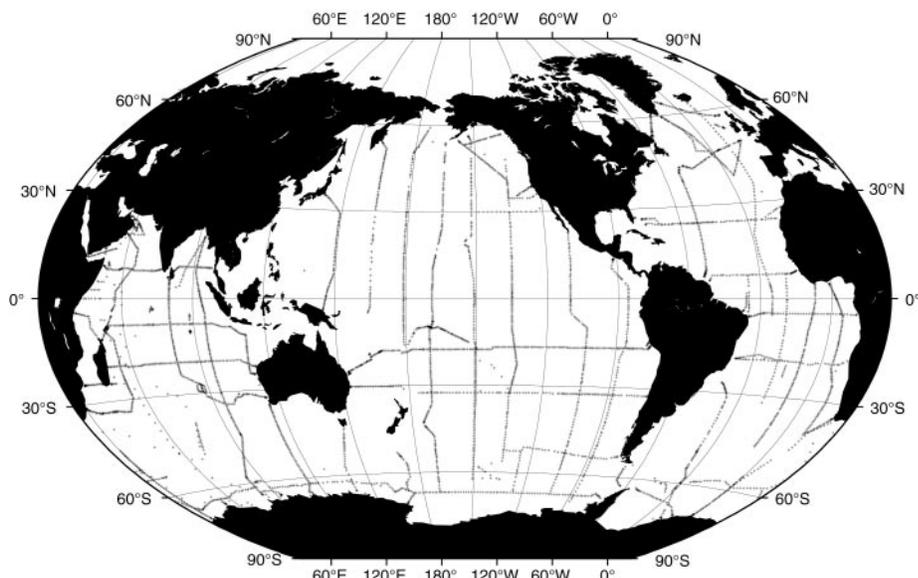
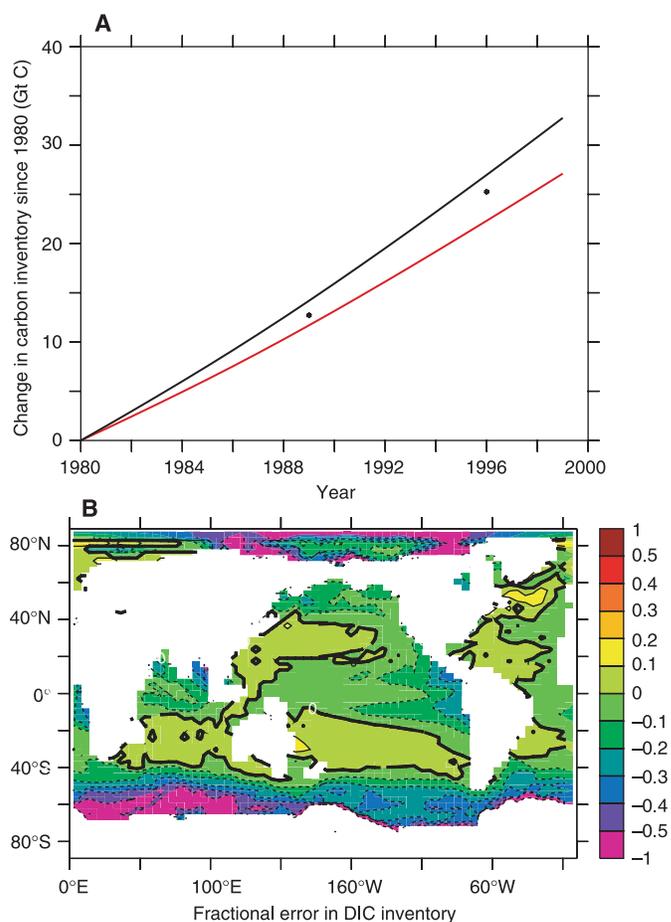


Fig. 2. Global distribution of CFC measurements used for this study and taken as part of the WOCE (see <http://whpo.ucsd.edu>).

Fig. 3. (A) The change in global inventory of anthropogenic CO₂ from 1980 to 1999 simulated in the CSIRO model (black line) compared with the model inventory in waters with an age younger than 30 years (red line) and the inventory change calculated using the CFC age technique for waters younger than 30 years (data points). From 1 June 1980 to 1 June 1999, the total simulated anthropogenic CO₂ inventory change is 32.8 Pg of C. In waters younger than 30 years, the inventory change is 27.8 Pg of C. The CFC age technique gives an inventory change of 30.1 Pg of C for waters younger than 30 years, overestimating the inventory change in waters younger than 30 years by 2.3 Pg of C because of age biases and air-sea exchange assumptions (data points versus red line), but missing the 5 Pg of C inventory in waters older than 30 years. These partially cancel to give a reasonably consistent estimate of the decadal anthropogenic CO₂ inventory (~10%) in the model. (B) Fractional error in the water column inventory of anthropogenic CO₂ from 1980 to 1999 calculated from the CFC ages as compared to the simulated inventory of anthropogenic CO₂ in the model. Negative values indicate that the CFC age estimate is too high. This plot highlights the regions with most bias in using the CFC age technique from our model analysis. In the Southern Ocean, analysis of observations south of 40°S (Fig. 1) (25) shows that the model is most likely overestimating the extent of the bias based on the CFC age approach.



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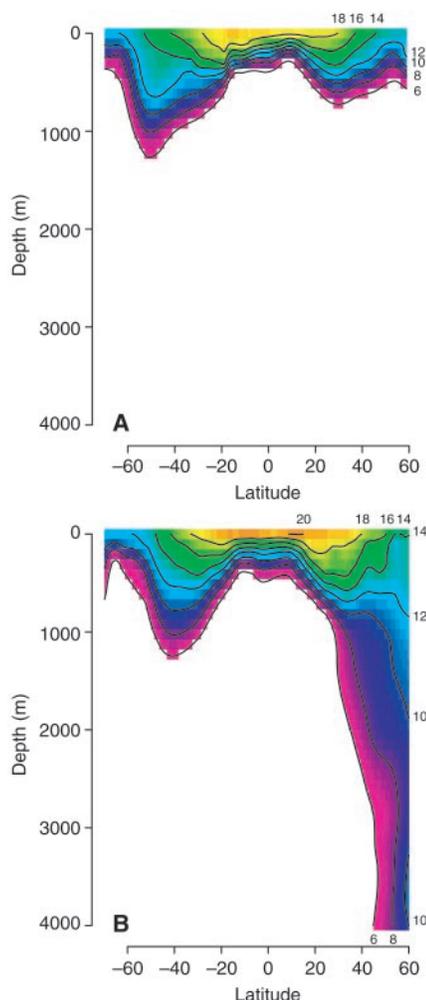


Fig. 4. Basinwide estimates of the accumulation of anthropogenic CO₂ (in μmol/kg) for the period from 1980 to 1999, using the CFC age method for the (A) Pacific and (B) Atlantic Oceans.

ic CO₂ accumulation (35). We compared the change in anthropogenic accumulation for the period from 1980 to 1999 that was estimated from the CFC simulation with and without climate change. As the ocean warms and becomes more stratified because of global warming, CFC transport into the ocean slows, which results in older water masses remaining in the ocean interior. From the model simulations, climate change reduces the oceanic uptake during the 1990s by less than 1% relative to a simulation that neglects this process. Hence, the potential error associated with climate change is small enough to neglect.

The CFC age technique used here explicitly addresses the abiotic solubility-driven anthropogenic CO₂ flux into the ocean and is independent of other observational estimates of anthropogenic CO₂ uptake. The assumptions used for our observational estimate here are exactly the same as those used to estimate anthropogenic CO₂ from models (36). The potential biases in the CFC age technique result in an overestima-

Table 1. Estimates of the inventory of anthropogenic CO₂ (in Pg of C) from 1980 to 1999 (33). Uncertainties are 20% and taken to be an upper limit (34).

Period	Ocean			
	Pacific	Atlantic	Indian	Global
Mid-1980 to mid-1989	7.0	5.0	2.8	14.8
Mid-1990 to mid-1999	8.2	6.4	3.3	17.9

Table 2. Annual mean oceanic anthropogenic CO₂ uptake (in Pg/year) from the Ocean Carbon Model Intercomparison Project (9, 37) for the past two decades. MIT, Massachusetts Institute of Technology, Cambridge, Massachusetts; NCAR, National Center for Atmospheric Research, Boulder, Colorado; IPSL, Institut Pierre Simon Laplace, Paris, France; MPIM, Max Planck Institut für Meteorologie, Hamburg, Germany; SOC, Southampton Oceanography Centre/Hadley Centre, UK Meteorological Office, England; IGCR, Institute for Global Change Research, Tokyo, Japan; AWI, Alfred Wegener Institute for Polar and Marine Research, Bremerhaven, Germany; PIUB, Physics Institute, University of Bern, Switzerland; NERSC, Nansen Environmental and Remote Sensing Centre, Bergen, Norway.

Model	1980–89	1990–99
PRINCE	1.65	2.15
LLNL	1.78	2.22
CSIRO	1.78	2.32
Upper limit from this study	1.9	2.3
MIT	1.91	–
NCAR	1.93	2.5
IPSL	2.03	2.63
MPIM	2.01	2.6
SOC	2.01	2.62
IGCR	2.07	2.7
AWI	2.14	2.75
PIUB	2.18	2.82
NERSC	2.38	–

tion of the anthropogenic storage in the ocean and can therefore provide a firm upper limit on anthropogenic CO₂ uptake in the ocean. However, our estimates only include waters younger than 30 years and will therefore miss the anthropogenic CO₂ inventory in older waters. To determine the likely contribution of waters older than 30 years, we used the results from the OGCM and also from the direct observations of Sabine *et al.* (12). Based on the model (Fig. 3), the anthropogenic CO₂ inventory for waters older than 30 years is about 18% of the total from 1980 to 1999 and only 5% using the direct observations reported by Sabine *et al.* (12). If we take the model results as the upper bound and add the contribution to our estimates, then the absolute maximum net oceanic solubility-driven anthropogenic CO₂ flux from 1980 to 1999 is about 39 Pg of C, corresponding to 1.9 and 2.3 Pg of C per year for the past two decades. In a recent intercomparison project of

12 international models that simulate anthropogenic CO₂ uptake (37), three models are close to this observational upper limit [Princeton University in Princeton, New Jersey (PRINCE), the Lawrence Livermore National Laboratory in California (LLNL), and the Commonwealth Scientific and Industrial Research Organisation in Hobart, Australia (CSIRO)], whereas the majority of models overestimated the uptake of anthropogenic CO₂ during the past two decades (Table 2). The reason for this is not clear and requires closer regional examination in the models.

References and Notes

1. R. J. Matear, A. C. Hirst, B. I. McNeil, *Geochim. Geophys. Geosys.* **1**, 1 (2000).
2. K. Keller, R. Slater, M. Bender, R. M. Key, *Deep-Sea Res.* **2** **49**, 345 (2002).
3. L. Bopp *et al.*, *Global Biogeochem. Cyc.* **16**, 1 (2002).
4. R. Keeling, H. Garcia, *Proc. Natl. Acad. Sci. U.S.A.* **99**, 7848 (2002).
5. G.-K. Plattner, F. Joos, T. F. Stocker, *Global Biogeochem. Cyc.* **16**, 4 (2002).
6. R. F. Keeling, S. C. Piper, M. Heimann, *Nature* **381**, 218 (1996).
7. M. Battle *et al.*, *Science* **287**, 2467 (2000).
8. J. T. Houghton *et al.*, *Climate Change 2001: The Scientific Basis, Contribution of Working Group 1 to the Third Assessment Report of the Intergovernmental Panel on Climate Change* (Cambridge Univ. Press, Cambridge, 2001).
9. J. C. Orr *et al.*, *Global Biogeochem. Cyc.* **15**, 43 (2001).
10. T. Peng *et al.*, *Nature* **396**, 560 (1998).
11. D. W. R. Wallace, *Monitoring Global Ocean Carbon Inventories* [Ocean Observing System Development Panel (OOSDP) Background Report 5, Texas A&M Univ., College Station, TX, 1995].
12. C. L. Sabine *et al.*, *Global Biogeochem. Cyc.* **13**, 179 (1999).
13. B. I. McNeil, B. Tilbrook, R. J. Matear, *J. Geophys. Res.* **106**, 31431 (2001).
14. M. H. England, E. Maier-Reimer, *Rev. Geophys.* **39**, 29 (2001).
15. Variations in sea surface alkalinity influence the buffer factor for the ocean. The buffer factor is a measure of how much anthropogenic CO₂ can be dissolved in the surface ocean [E. T. Sundquist, L. N. Plummer, T. M. L. Wigby, *Science* **204**, 1203 (1979)].
16. R. J. Matear, unpublished data.
17. Y. W. Watanabe, T. Ono, A. Shmamoto, *Mar. Chem.* **72**, 297 (2000).
18. M. J. Warner, R. F. Weiss, *Deep-Sea Res.* **2** **32**, 1485 (1985).
19. S. J. Walker, R. F. Weiss, P. K. Salameh, *J. Geophys. Res.* **105**, 14285 (2000).
20. To estimate water ages from CFC-12, the CFC-12 concentrations were first converted into a partial pressure (*p*) by $p\text{CFC-12} (\mu\text{atm}) = \text{CFC-12}/\alpha$, where μatm is 10⁻⁶ atmospheres and α is the temperature-dependent solubility (78). The time since last contact with the atmosphere (water age) was then estimated by finding the date at which the atmospheric *p*CFC-12 equaled the oceanic *p*CFC-12. Unlike CFC-11, CFC-12 continued to increase in the atmosphere through the mid-1990s and therefore will give less bias due to mixing (23). Mixing beyond bulk advection from a single source region is not accounted for when calculating CFC ages. The biases in using CFC ages were explored with an OGCM (34).
21. T. M. Hall, T. W. N. Haine, *J. Phys. Oceanogr.* **32**, 1987 (2002).
22. D. H. Min, J. L. Bullister, R. F. Weiss, *Geophys. Res. Lett.* **27**, 3909 (2001).
23. R. Sonnerup, *Geophys. Res. Lett.* **28**, 1739 (2001).
24. T. W. N. Haine, T. M. Hall, *J. Phys. Oceanogr.* **32**, 1932 (2002).
25. R. J. Matear, B. I. McNeil, unpublished data.
26. C. D. Keeling, T. P. Whorf, *Trends: A Compendium of*

- Data on Global Change* (Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, TN, 2002).
27. C. Goyet, P. G. Poisson, *Deep-Sea Res.* **36**, 2635 (1989).
 28. We used the dissociation constants derived from (27) to calculate the equilibrium DIC concentration. Using different constants resulted in negligible changes in our estimates.
 29. The preformed alkalinity distribution is essential because it largely determines the oceanic buffer capacity. Preformed alkalinity was derived using a multiple linear regression of the surface (<60 dbar) alkalinity distribution for each of the three ocean basins in a way similar to the technique used by Sabine *et al.* (12). Using in situ alkalinity instead of preformed alkalinity resulted in negligible changes in our results.
 30. Our method requires the oceanic biological carbon pump to be in a preindustrial steady state. Anthropogenic CO₂ is assumed to be a perturbation propagating through the ocean as a tracer inert to the biological carbon pump.
 31. Most models suggest that the CO₂ air-sea disequilibrium has been increasing with time (16). However, the extent of this increase varies between models. This bias is reduced by estimating anthropogenic CO₂ on shorter time scales (such as decadal) during which the disequilibrium is closer to constant than over longer time scales (since preindustrial times).
 32. R. J. Matear, A. C. Hirst, *Tellus* **51B**, 722 (1999).
 33. The global oceanic inventory of anthropogenic CO₂ from 1980 to 1989 and from 1990 to 1999 was calculated on a 1° grid at 100 m intervals and integrated to 5000 m for the Atlantic and 2000 m for the Indian and Pacific Oceans. The objective mapping techniques used were based on the methods described by Sarmiento *et al.* (38). For the basinwide inventories, the Southern Ocean was split from 120°E to 70°W for the Pacific, 70°W to 30°E for the Atlantic, and 30°E to 120°E for the Indian Ocean. Aside from the Southern Ocean, the major continents were taken to be main boundaries separating the ocean basins.
 34. We used the OGCM to estimate the combined bias and uncertainty of our anthropogenic CO₂ uptake estimates by comparing the simulated and estimated inventory from CFC predictions in the model. Our model analysis suggests that the CFC age technique is biased ~8% lower (30.1 Pg of C) than the total simulated anthropogenic CO₂ inventory from 1980 to 1999 (32.8 Pg of C). We are hesitant to apply this "correction" because there is good agreement between our CFC age-based inventory and those using the direct techniques (MLR and isopycnal) in the Indian Ocean (Fig. 1), which suggests that the model bias may be exaggerated. Instead, we expand our estimated uncertainty to ~15%, which includes both the predicted "bias" from our model analysis (including mixing and disequilibrium) and the uncertainty in the inventories obtained from the MLR and isopycnal analyses. The measurement precision of CFC was taken to be ±0.005 pmol/kg. Adding this value to the simulated CFCs in the model resulted in an uncertainty of about 5% in the global inventory. Repeat measurements of CFCs show differences in CFC ages from year to year (39). As model simulations may underestimate interannual variability in the ocean, we added extra "noise" to the CFC measurements in the model (±0.01 pmol/kg), which resulted in a further 10% uncertainty in our estimates. The uncertainty associated with the inventory calculation was determined using two different interpolation schemes (objective and loess) and was estimated to be 2%. The total uncertainty of our estimate (~20%) was calculated by a sum-of-squares propagation of all uncertainties with the assumption that each is independent of the others. We believe the uncertainty of our estimates to be an upper limit, given the good consistency between our estimated inventory in the Indian Ocean and those using the direct methods (10, 12) (Fig. 1). Because these direct techniques (MLR and isopycnal) are not subject to the uncertainties associated with the CFC age method (for example, in ages and disequilibrium), they provide evidence of upper-bound errors.
 35. The predicted warming of the ocean in the climate change simulation (0.3°C) was in good agreement with observations from 1950 to 1998 (40) and provides confidence that the model is simulating recent changes in the ocean.
 36. To simulate anthropogenic CO₂ uptake, models use constant climate and assume that biological processes are operating in a steady state. These are the same assumptions used for the CFC age technique.
 37. J. C. Orr *et al.*, paper presented at the International Geosphere-Biosphere Programme Open Science Conference, Amsterdam, 10 to 13 July 2001. For more details and to download the results of this study, go to www.ipsl.jussieu.fr/OCMIP/.
 38. J. L. Sarmiento, J. Willebrand, S. Hellerman, *Objective Analysis of Tritium Observations in the Atlantic Ocean During 1971–1974* (Ocean Tracer Laboratory Tech Report 1, Princeton Univ. Press, Princeton, NJ, 1982).
 39. S. C. Doney, J. L. Bullister, R. Wanninkhof, *Geophys. Res. Lett.*, **25**, 1399 (1998).
 40. S. Levitus, J. I. Antonov, T. P. Boyer, C. Stephens, *Science* **287**, 2225 (2000).
 41. This study would not have been possible without the efforts of those responsible for collecting and analyzing CFCs during WOCE and making those measurements available. They include R. Fine, M. Rhein, W. Roether, W. Smethie, M. Warner, Y. Watanabe, R. Weiss, and C. S. Wong. We also thank three anonymous reviewers and C. Sweeney for comments on the text. B.I.B. and R.M.K. were supported by NSF (grants OCE9819144 and OCE9986310). J.L.B. was supported by NOAA's Office of Global Programs and Global Carbon Cycle Program, J.L.S. was supported by the Carbon Modeling Consortium, and R.J.M. was supported by Environment Australia and the CSIRO Climate Change Program.

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Satellite Observations of Magnetic Fields Due to Ocean Tidal Flow

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The ocean is an electrically conducting fluid that generates secondary magnetic fields as it flows through Earth's main magnetic field. Extracting ocean flow signals from remote observations has become possible with the current generation of satellites measuring Earth's magnetic field. Here, we consider the magnetic fields generated by the ocean lunar semidiurnal (M_2) tide and demonstrate that magnetic fields of oceanic origin can be clearly identified in satellite observations.

In a fully magnetohydrodynamic process, the flow and electromagnetic fields are coupled. In the ocean, however, flow generates electromagnetic fields but the electromagnetic fields are not thought to affect the flow appreciably. This reduced magnetohydrodynamic case is often called "motional induction" and can be understood as follows. The dissolved salts in seawater form hydrated, electrically charged ions. As the charged ions are carried by the ocean flow through Earth's main magnetic field, they are deflected by the Lorentz force, which acts in a direction perpendicular to both the velocity and magnetic field. This leads to various combinations of two effects. First, the migrating ions can accumulate to form electrical spatial charge densities that in turn create electric fields that tend to prevent further migration of charge. Second, the spatial charge densities can be relieved by electrical shorting through surrounding sections of the water or electrically conducting sediments. The latter effect involves electrical currents and the associated secondary magnetic fields, which are the subject of this paper.

Two components of the ocean-generated magnetic field can be distinguished. The first is a "toroidal" component that has been estimated to reach maximum amplitudes of 100 nT but is confined to the ocean and sediments and is therefore not observable remotely (1–5). This component results from electric current circuits closing in planes containing the vertical axis. The second is a much weaker (1 to 10 nT) "poloidal" component with large spatial decay scales that allow the magnetic fields to reach remote land and satellite locations (4, 6–10). This component involves electric current circuits closing horizontally and is the least understood because it is generated by large-scale integrals of ocean flow transport and estimates typically require large-domain integrations.

But this dependence of the far-reaching poloidal magnetic fields on transport integrals also makes these fields attractive. In principle, information about past and present ocean variability is contained in the land and satellite magnetic records, and this variability would primarily reflect integrated transport quantities (including in ice-covered regions) that are difficult to obtain using other methods (11). Understanding such ocean variability is a key factor in addressing climate and global change concerns, and although an assessment of the potential for exploiting the magnetic fields in this way is beyond the scope of this paper, here we describe

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