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An improved estimate of the isotopic air-sea disequilibrium of CO_2: Implications for the oceanic uptake of anthropogenic CO_2

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Abstract. We reevaluate the isotopic air-sea disequilibrium of CO_2 on the basis of a new high-quality global data set of surface observations of the reduced isotopic ratio ($\delta^{13}C$) of dissolved inorganic carbon (DIC). We find a global mean isotopic air-sea disequilibrium of 0.62±0.10 permil for 1990. Inserting our estimate into an anthropogenic ^{13}C budget for the period from 1985 to 1995 yields an oceanic uptake of 1.5±0.9 Pg C yr^{-1}, within the range of most other estimates. The large uncertainty of this estimate could be reduced with (i) better characterization of the surface ocean variability in $\delta^{13}C$, (ii) improved knowledge of the air-sea gas exchange coefficient, and (iii) a significant reduction in the uncertainty associated with the air-land biosphere disequilibrium.

Introduction

Quay et al. [1992] proposed that the oceanic uptake of anthropogenic CO_2 can be estimated from the change in the oceanic inventory of ^{13}C and an assessment of the ^{13}C budget of the combined atmosphere/ocean system. Their study has led to a series of further investigations to explore the potential of the ^{13}C method. *Tans et al.* [1993] suggested a method similar to that of *Quay et al.* [1992], except that the ^{13}C budget is only expressed for the atmosphere. By this substitution, air-sea fluxes enter the equations, and in particular enter the equation for the isotopic disequilibrium flux between the atmosphere and the surface ocean:

$$F_{as}^{diseq} = F_{as}\left(\delta^{13}C_{as}^{eq} - \delta^{13}C_{atm}\right), \qquad (1)$$

where $\delta^{13}C$ refers to r/r_s-1 (r = $^{13}C/^{12}C$ ratio of a sample and r_s the $^{13}C/^{12}C$ ratio of the International PDB Standard), F_{as} denotes the gross flux of CO_2 from the atmosphere to the ocean, $\delta^{13}C_{atm}$ the $\delta^{13}C$ of atmospheric CO_2, and $\delta^{13}C_{as}^{eq}$ the $\delta^{13}C$ atmospheric CO_2 would have were it in equilibrium with the $\delta^{13}C$ of surface ocean DIC. *Tans et al.*[1993] used the global surface oceanic data set of *Kroopnick* [1985], sampled during the Geochemical Ocean Sections Study (GEOSECS) in the 1970s, and obtained an isotopic air-sea disequilibrium of 0.43 permil and a disequilibrium flux of 36.6 Pg C permil yr^{-1}. When *Tans et al.* [1993] inserted this value, along with an estimate of the terrestrial isotopic disequilibrium flux, into a global budget of anthropogenic ^{13}C, they obtained an oceanic uptake of anthropogenic CO_2 of only 0.2 Pg C yr^{-1}, much lower than generally accepted [*Schimel et al.*, 1996], and also much lower than the estimate of *Quay et al.* [1992].

We recently synthesized a new global high-quality data set of the $\delta^{13}C$ of DIC in the surface oceans [*Gruber et al.*, 1999]. These data permit us to readdress the isotopic air-sea disequilibrium of CO_2 and its role as a constraint on the oceanic uptake of anthropogenic CO_2. Here we demonstrate that our new estimate of the isotopic air-sea disequilibrium is consistent with the current knowledge of oceanic uptake of anthropogenic CO_2, but that the uncertainties are still too large to provide strong constraints on the global anthropogenic carbon budget.

Data Considerations

Our calculations are based on the surface oceanic $\delta^{13}C$ data obtained by the Scripps Institution of Oceanography between 1978 and 1997 in all major ocean basins [*Gruber et al.*, 1999]. Over this period atmospheric $\delta^{13}C$ decreased from approximately -7.4 to -7.8 permil [*Keeling et al.*, 1995]. In response, the $\delta^{13}C$ of DIC in the ocean has also decreased [*Gruber et al.*, 1999; *Sonnerup et al.*, 1999]. Such a reduction of the $\delta^{13}C$ in a reservoir caused by the addition of isotopically light CO_2 is often referred to as the ^{13}C Suess effect. We adjust the data to the common year of 1990 on the basis of the surface ocean ^{13}C Suess effects simulated by the Princeton Ocean Biogeochemistry Model (POBM) [*Murnane and Sarmiento*, 2000]. We increase the model predicted ^{13}C Suess effects everywhere by 20% to agree better with direct observations that indicate a global mean ^{13}C Suess effect between -0.015 to -0.018 permil yr^{-1} [*Gruber et al.*, 1999; *Sonnerup et al.*, 1999]. The resulting adjusted $\delta^{13}C$ surface observations are afterwards binned into 5° bins (Figure 1a). We have made no adjustments to take into account seasonal and interannual variations. This could result in a bias in our global estimate because seasonal and interannual variations exceeding 0.2 permil have been observed [*Gruber et al.*, 1999]. However, our data have been sampled relatively uniformly over the seasons and we therefore expect that the error introduced by our neglecting of seasonal variations is smaller than 0.2 permil.

We extrapolate the binned data to the global surface oceans using a combination of nearest neighbor and tracer-correlation techniques, whereby the relative weights between the two techniques are given by the number of neighboring

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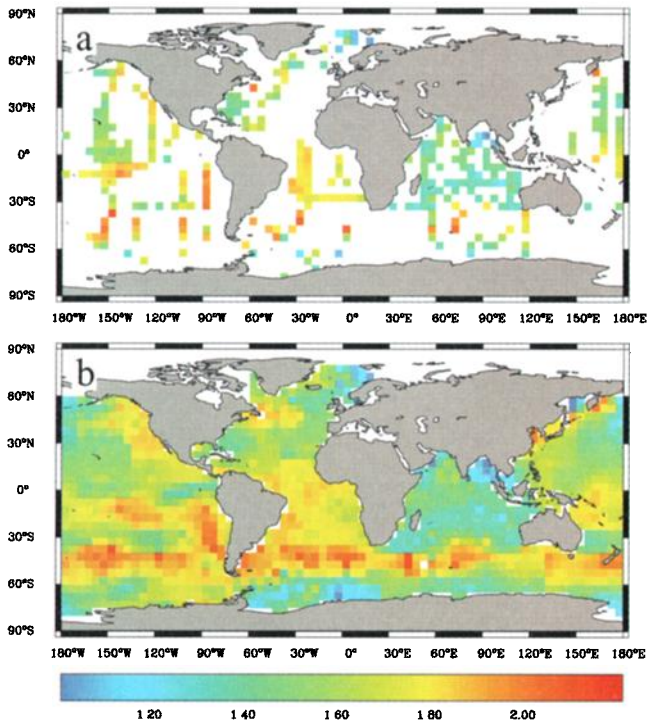

Figure 1. Maps of the surface ocean $\delta^{13}C$ of dissolved inorganic carbon (permil) for the year 1990 (see text for details).(a) Adjusted observations after binning.(b) Extrapolated observations.

observations (Figure 1b). The tracer-correlation technique is based on multiple linear regressions with concurrently measured temperature, salinity, oxygen, phosphate and silicate, which are then extrapolated to the world ocean using the NOAA NESDIS climatology (see *Gruber and Keeling* [1999] for further details).

Air-Sea Disequilibrium

We calculate the $\delta^{13}C$ of atmospheric CO_2 in isotopic equilibrium with the current surface ocean DIC as a flux weighted average, i.e.,

$$\delta^{13}C_{as}^{eq} = \frac{\int_A F_{as}(1+\varepsilon_{eq})\delta^{13}C_{oc}dA}{\int_A F_{as}dA}, \qquad (2)$$

where $(1+\varepsilon_{eq})$ denotes the term for the equilibrium fractionation. We compute the gross exchange flux, F_{as}, using the mean atmospheric CO_2 concentration for 1990 of 353 ppm [*Keeling and Whorf*, 1994], the quadratic wind-speed dependent gas exchange coefficient of *Wanninkhof* [1992], annual mean wind speeds from COADS [*da Silva et al.*, 1994], annual mean sea-ice coverage from *Alexander and Mobley* [1974], and annual mean temperature and salinity according to NOAA NESDIS. For $(1+\varepsilon_{eq})$, we chose the temperature dependent relationship of *Zhang et al.* [1995] for dissolved inorganic carbon. Finally, to calculate the isotopic air-sea disequilibrium according to (1), we adopt a global mean $\delta^{13}C$ of atmospheric CO_2 for 1990 of -7.65 permil [*Keeling et al.*, 1995], based on a mass-spectrometric correction for N_2O of 0.21 permil.

We find a global mean air-sea disequilibrium for 1990 of 0.62±0.10 permil on the basis of our adjusted and extrapolated data. This error estimate is just based on an estimated uncertainty of about 0.08 permil for $\delta^{13}C_{as}^{eq}$, inferred from a large number of sensitivity studies where various extrapolation schemes, different fractionation factors, windspeed, and sea-ice climatologies were employed, and an estimated error of about 0.06 permil for the atmospheric $\delta^{13}C$. This latter estimate includes the error that arises from the uncertain magnitude of a correction that needs to be applied to atmospheric samples of CO_2 to account for the presence of N_2O. The estimated air-sea disequilibrium is not sensitive to the details of the extrapolation. Using zonal mean distributions on the basis of the adjusted, binned data yields a similar air-sea disequilibrium of 0.57 permil.

We recognize that this isotopic disequilibrium estimate is sensitive to the gas exchange flux weighting, in particular to the relative weighting between the high and the low latitudes [*Heimann and Maier-Reimer*, 1996]. Using the *Liss and Merlivat* [1986] windspeed relationship for computing the gas exchange coefficient yields a very similar disequilibrium of 0.64 permil, but employing the cubic wind-speed relationship proposed recently by *Wanninkhof and McGillis* [1999] results in a very low disequilibrium of 0.46 permil.

We have decided to use the quadratic wind speed relationship of *Wanninkhof* [1992] as a weighting factor, because it yields most consistent results when used to simulate atmospheric oxygen observations from surface ocean oxygen data [*R. F. Keeling et al.*, 1998]. It is possible that the true isotopic disequilibrium is outside our estimated uncertainty because of systematic errors stemming from an inappropriate gas exchange coefficient. A better determination of the wind-speed dependency of the gas exchange coefficient is required to resolve this issue. Since we have used annually averaged data for our computation, a bias may arise also from seasonal co-variance between surface $\delta^{13}C$, equilibrium fractionation and air-sea gas exchange. We believe that this effect is small, but it needs to be addressed in future studies.

Our disequilibrium of 0.62±0.10 permil is substantially larger than that computed by *Tans et al.* [1993] (0.43 permil). However, their estimate applies to the period from 1970 to 1990, and our results from the POBM indicate that the air-sea disequilibrium has increased by about 0.08 permil between 1980 and 1990, bringing their estimate almost within our range. Our estimated disequilibrium agrees well with that simulated by the HILDA model (0.64 permil for 1990) [*Joos et al.*, 1999], by the POBM (0.59 permil for 1990) and, adjusted to 1990, with that of *Heimann and Maier-Reimer* [1996].

Implications for the oceanic uptake of anthropogenic CO_2

Following the notation of *Heimann and Maier-Reimer* [1996], we calculate the change in the oceanic inventory of CO_2, $\dot{N}_{oc}$ by solving

$$\begin{aligned}\dot{N}_{oc}\left(\delta^{13}C_{oc}+\varepsilon_{sa}-\delta^{13}C_{bio}\right) \approx\\ Q_{foss}\left(\delta^{13}C_{foss}-\delta^{13}C_{bio}\right)\\ -\dot{N}_{atm}\left(\delta^{13}C_{atm}-\delta^{13}C_{bio}\right)-N_{atm}\,d\delta^{13}C_{atm}/dt\\ +F_{ab}\left(\delta^{13}C_{ab}^{eq}-\delta^{13}C_{atm}\right)+F_{as}\left(\delta^{13}C_{as}^{eq}-\delta^{13}C_{atm}\right)\\ +(0.5F_{r,DIC}+F_{r,org})\left(\delta^{13}C_{oc}+\varepsilon_{sa}-\delta^{13}C_{bio}\right) \qquad (3)\end{aligned}$$

where the definitions of the different terms are given in Table 1. Solving (3) with our estimated air-sea disequilibrium

Table 1. Description and values of parameters used in equation (4)

Parameter	Description	Value	Reference
ε_{sa}	sea-air kinetic isotopic fract.	-10.2 permil	this study
$\delta^{13}C_{atm}$	δ^{13}C of atm. CO_2	-7.65 permil	[*Keeling et al., 1995*]
$d\delta^{13}C_{atm}/dt$	rate of change of $\delta^{13}C_{atm}$	-0.016 permil yr^{-1}	[*Keeling et al., 1995*]
$\delta^{13}C_{bio}$	mean δ^{13}C of terr. biosphere	-24.7 permil	[*Bakwin et al., 1998*]
$\delta^{13}C_{foss}$	δ^{13}C of fossil fuel	-28.4 permil	[*Andres et al., 1996*]
$\delta^{13}C_{oc}$	δ^{13}C of surface ocean DIC	1.651 permil	this study
N_{atm}	Atmospheric inventory of CO_2	748 Pg C	[*Keeling and Whorf, 1994*]
$\dot{N}_{atm}$	Rate of change of N_{atm}	3.07 Pg C yr^{-1}	[*Keeling and Whorf, 1994*]
Q_{foss}	Fossil fuel emissions	5.95 Pg C yr^{-1}	[*Marland et al., 1998*]
F_{as}	Gross air-sea flux of CO_2	85 Pg C yr^{-1}	this study
$\left(\delta^{13}C_{as}^{eq} - \delta^{13}C_{atm}\right)$	Isotopic air-sea disequilibrium	0.62±0.08 permil	this study
$F_{ab}\left(\delta^{13}C_{ab}^{eq} - \delta^{13}C_{atm}\right)$	Diseq. flux with terr. biosphere	24 Pg C permil yr^{-1}	[*Joos et al., 1999; Fung et al., 1997*]
$F_{r,DIC}$	River input of DIC	0.4 Pg C yr^{-1}[a]	[*Heimann and Maier-Reimer, 1996*]
$F_{r,org}$	River input of organic carbon	0.4 Pg C yr^{-1}[a]	[*Heimann and Maier-Reimer, 1996*]

All values representative for period from 1985 to 1995
[a] *a priori* values given by *Heimann and Maier-Reimer* [1996]

and with the values for the other terms as listed in Table 1, we compute an anthropogenic CO_2 uptake by the oceans, $\dot{N}_{oc}$, of approximately 1.5 Pg C yr^{-1}. An error analysis of (3) shows that the most sensitive terms are the isotopic disequilibrium fluxes. We compute an oceanic disequilibrium flux of 53±13 Pg C permil yr^{-1} (1990) on the basis of our estimated isotopic disequilibrium of 0.62±0.10 permil and a gross air to sea flux estimate of 85±17 Pg C yr^{-1} [*Heimann and Maier-Reimer*, 1996]. The latter error is based on our perceived uncertainty of 20% of the global oceanic bomb radiocarbon inventory [*Broecker et al.*, 1995]. For the terrestrial isotopic disequilibrium flux, we adopt a value of 24±7 Pg C permil yr^{-1} on the basis of the studies by *Joos et al.* [1997] and by *Fung et al.* [1997]. Inserting these values and uncertainties into (3) results in an uncertainty of 0.9 Pg C yr^{-1} for our oceanic uptake estimate.

Our estimate of the oceanic disequilibrium flux is in close agreement with predictions of the HILDA model (60 Pg C permil yr^{-1}, [*Joos et al.*, 1999]) and POBM (47 Pg C permil yr^{-1}, [*Murnane and Sarmiento*, 2000]). Our total disequilibrium flux (ocean and terrestrial combined) of 77±15 Pg C permil yr^{-1} is in good agreement with a recent estimate of 89±21 Pg C permil yr^{-1} based on atmospheric oxygen data [*Battle et al.*, 2000]. Our estimate agrees still better with 83 Pg C permil yr^{-1} derived using the method of *Ciais et al.* [1995] (updated as referenced by *Battle et al.* [2000]).

Discussion and Conclusion

We have estimated an oceanic uptake of anthropogenic CO_2 of 1.5±0.9 Pg C yr^{-1} by combining our improved estimate of the isotopic air-sea disequilibrium with best estimates of the current atmospheric ^{13}C budget. Our uptake estimate supports the conclusion of *Heimann and Maier-Reimer* [1996] that the air-sea disequilibrium estimate of *Tans et al.* [1993] is too small. Our ocean uptake estimate also removes the inconsistency between this method on the basis of the isotopic air-sea disequilibrium and that based on the change of the oceanic inventory of ^{13}C [*Quay et al.*, 1992; *Sonnerup et al.*, 1999]. Our estimate is consistent with independent estimates based on observations [*Battle et al.*, 2000; *Takahashi et al.*, 1999; *R. F. Keeling et al.*, 1996] and models [*Orr et al.*, 2000]; however, its uncertainty is too large to constrain the global budget of anthropogenic CO_2 with confidence. This uncertainty could be strongly reduced by a better spatial and temporal coverage of δ^{13}C in the ocean, by lower uncertainties associated with the gas exchange coefficient, and by improved characterization of the isotopic air-sea disequilibrium between the atmosphere and terrestrial biosphere. However, indirect methods to estimate the global oceanic uptake of anthropogenic CO_2, such as this one, will always be limited by the numerous assumptions that need to be made. More direct methods, such as monitoring the ocean to detect the increase of CO_2 over time, are ultimatively required to reliably account for the redistribution of the emitted anthropogenic CO_2 in the global carbon system. Although isotopic constraints may not be good enough to determine the global oceanic uptake of anthropogenic CO_2 with great certainty, they can aid substantially in atmospheric inversions and deconvolution studies to constrain regional sources and sinks and their temporal variability [*Keeling et al.*, 1995].

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